PHYSICAL REVIEW B VOLUME 1, NUMBER 1 1 JANUARY 1970

Descriptive Model for the Magnetic Behavior of Au-Fe Alloys*

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(Received 21 August 1969)

Antiferromagnetic Au-Fe alloys acquire a ferromagnetic character, i.e., demonstrate remanence, possess a positive Curie θ, etc., in the presence of an external field. A phenomenological model is presented which purports to describe the mixed magnetic behavior of these alloys. The model satisfactorily reconciles Mössbauer and magnetization results and provides an insight into the nature of the magnetic order.

THE occurrence of magnetic order in Au-Fe alloys has been well established by means of both Mössbauer[1–3] (ME) and magnetization (MAG) experiments.[4,5] Below ~15-at.% Fe, the ordering temperature is a unique function of composition,[6,7] the order corresponding to a random distribution of the magnetic moments. The basis for the latter statement is the observation that the intensity ratios of the Mössbauer hyperfine spectrum of the Fe occur in the ratio 3:2:1:1:2:3 as required of a random array of spins. However, a significant departure from randomness can be caused by the application of an external field, whose magnitude is a function of field strength, alloy composition, and temperature. This polarization is clearly reflected in the Mössbauer spectra of the 0.26 and 18% Fe alloys in Fig. 1. Graphical integration yields a more quantitative comparison of the variation in the intensity ratios as a function of composition in a 50-kOe field. The results of this calculation are shown in Fig. 2. Because of asymmetries arising from quadrupolar interactions[8] as well as the probable distribution of the molecular field, we have simply combined the intensities of lines 1 and 6 and similarly lines 2 and 5 to obtain the ratio I_{16}/I_{25}. No claim is made for the accuracy of the individual values, yet the functional dependence of the polarizability upon composition is clearly evident. A completely polarized ferromagnet would make $I_{16}/I_{25}=\infty$, whereas the random array would yield $I_{16}/I_{25}=1.5$. It is apparent from the results shown in Fig. 2 that the 0.26 and 18% Fe alloys have been significantly polarized, whereas the intermediate compositions are scarcely altered; in fact, from 2 to 5% Fe the ratio nearly corresponds to 1.5 in spite of the presence of the polarizing field.

TABLE I. Mössbauer, magnetization, and calculated results.

at.% Fe	μ^{MAG}	μ^{ME}	I_{16}/I_{25} (expt)	I_{16}/I_{25} (calc)	$H_n/H_n{}^0$ (expt)	$H_n/H_n{}^0$ (calc)
0.26	1.15	1.52	2.23	2.77	0.91	0.94
2.0	0.73	1.56	1.33	1.17	0.97	0.97
5.0	0.85	1.66	1.36	1.29	0.96	0.97
8.0	1.26	1.76	1.50	2.40	⋯	⋯
10.5	1.64	1.83	1.93	6.87	0.93	0.93
18.0	⋯	⋯	4.79	⋯		

In contrast to the unvarying values of the intensity ratios, the magnitude of μ^{MAG}, the effective moment per Fe atom, as determined by conventional magnetization measurements[9] passes through a pronounced minimum in the vicinity of ~2% Fe as shown in Fig. 3. The calculation of μ^{MAG} is for the same field and temperatures as for I_{16}/I_{25}. A previous Mössbauer investigation[6] showed the hyperfine field to be a weak linear function of the Fe concentration over this region. Hence we suspect that the minimum in μ^{MAG} must result from the change in spin order although, for the moment, this appears to contradict the polarization experiments. We may add that the contradiction is apparently enhanced by the fact that stable hysteresis loops are obtainable[5,9] at sufficiently low temperatures, indicating a significant degree of ferromagnetic character.

The foregoing contradiction, however, can be quite simply explained. First, let us assume that we are able to calculate the moment on the Fe by use of

$$\mu_A{}^{\mathrm{ME}}=\mu_{\alpha-\mathrm{Fe}}[\Gamma_A/(\Gamma_{\alpha-\mathrm{Fe}})]. \tag{1}$$

This relationship seems to have been first proposed by Marshall *et al.*,[10] and although it has not proven to be universally valid, we will, nevertheless, accept it for the following calculations. Equation (1) assumes the magnitude of the hyperfine splitting to be directly proportional to the atomic moment; hence, $\Gamma_{\alpha-\mathrm{Fe}}$ and Γ_A are the magnitudes of the hyperfine splitting for pure Fe and the alloys, respectively, extrapolated to 0°K. We now propose that the discrepancy between μ^{MAG} and μ^{ME} at each composition merely reflects the

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* Work performed under the auspices of the U. S. Atomic Energy Commission. A portion of this work was done while the author was visiting Professor of Physics, Queens University, Flushing, N. Y.

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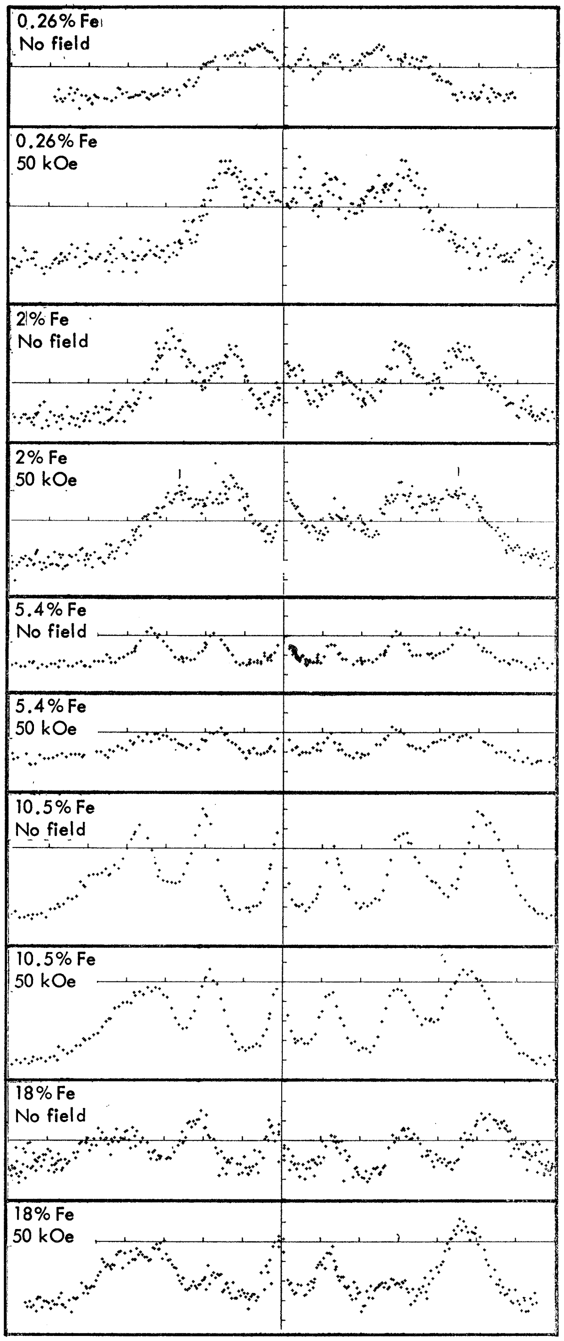

FIG. 1. Magnetic hyperfine spectra of Fe^{57} of five Au-Fe alloys as observed by the Mössbauer effect. In all cases the temperature is well below the ordering temperature. Note that an external field of 50 kOe causes substantial polarization of the 0.26- and 18-at.% alloys whereas the others are relatively unchanged. The 0.26% alloy was measured at 2.0°K; all others were measured at 4.2°K.

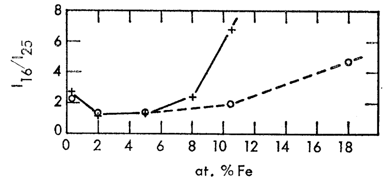

FIG. 2. Observed intensity ratios I_{16}/I_{25} of lines 1 and 6 to lines 2 and 5 (open circles) as obtained from the graphical integration of the spectra shown in Fig. 1. These are compared with the values calculated from the model (crosses).

state of order. Our model merely represents the true angular distribution of moments about the axis of the applied field by a single average angle of inclination $\bar{\theta}$, which is given by

$$\cos\bar{\theta}=\mu^{\mathrm{MAG}}/\mu^{\mathrm{ME}}. \tag{2}$$

Now, using the appropriate angular distribution functions for the γ intensities, viz., $I_{16}\propto 9/4(1+\cos^2\theta)$ and $I_{25}\propto 3\sin^2\theta$, one can easily derive

$$I_{16}/I_{25}=\tfrac{4}{3}(1+\cos^2\bar{\theta})/(1-\cos^2\bar{\theta}). \tag{3}$$

The results of applying Eq. (3) to the various alloys are tabulated in Table I and displayed in Fig. 2. Considering the simplicity of the model, the agreement for alloys containing less than ~5% Fe is quite convincing. Before discussing the reason for the dramatic disagreement at higher concentrations, we will test the validity of our model in another wholly independent manner.

An applied field causes a reduction in the hyperfine field which for Fe is oppositely directed with respect to the atomic moment. If our model indeed offers a good approximation to the magnetic behavior of these alloys, we should be able to calculate the magnitude of the decrease in the hyperfine splitting using the previously calculated values of $\bar{\theta}$. The results of these calculations are listed in the last column of Table I as the ratios of H_n/H_n^0, where H_n and H_n^0 are the values of the hyperfine field in 50 kOe and in zero field, respectively. The agreement between calculation and measurement is seen to be excellent, although it must be admitted that this criterion is rather insensitive to variations in $\bar{\theta}$.

An explanation for the discrepancy between the predicted values of I_{16}/I_{25} must be based upon something other than the change in the degree of order or a radical change in the intrinsic moment of the Fe. The extent of the disagreement is further emphasized by

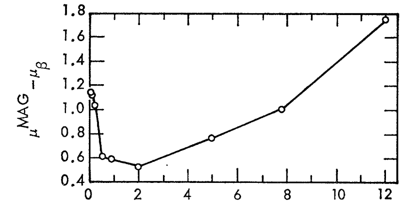

FIG. 3. Effective moment per Fe atom in μ_B as a function of composition. All values were calculated on the basis of a 50-kOe applied field at temperatures well below the respective ordering temperatures.

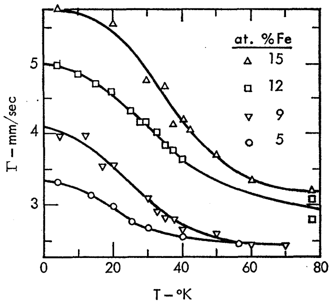

FIG. 4. Linewidth of the Au^{197} Mössbauer spectrum plotted as a function of temperature for four Au-Fe alloys. The linewidth is assumed to be a measure of the magnetic hyperfine field; it increases with decreasing temperature and increasing Fe concentration.

the fact that for the 12% alloy μ^{MAG} is slightly greater than μ^{ME} and this occurs in spite of the lack of complete polarization of the Mössbauer spectrum as well as the obvious failure of the alloy to achieve saturation at 50 kOe, as attested to by the magnetization measurements. It thus becomes necessary to postulate an additional source of magnetism. With this in mind, Mössbauer studies were extended[11] to include Au^{197}. The results of this study are summarized in Fig. 4. It is clear that the unresolved linewidth increases markedly with increasing concentration of Fe. The inflection points of these curves correspond exactly with the previously determined magnetic ordering temperatures. The increase in Γ below the ordering temperature is too great to be attributed to any source other than a genuine increase in the magnetic hyperfine field of the Au. This observation in conjunction with the previous arguments strongly suggests that above ~5% Fe, the Au itself becomes magnetic. It might be argued that this field arises solely from the polarization induced by proximate Fe atoms. However, simple polarization is not sufficient to account for the discrepancy between the calculated and measured values of I_{16}/I_{25}.

In the spirit of the foregoing approximations, one can calculate the necessary additional moment necessary to resolve the discrepancy shown in Fig. 2 which arises because μ^{MAG} cannot be attributed solely to the Fe. At 10.5-at.% Fe a moment of $\sim 0.044\,\mu_B$ (Bohr magnetons shared equally by all the Au atoms will resolve the apparent difference. At present it is not possible to decide with certainty whether the additional moment is localized upon those atoms which possess Fe nearest neighbors or whether a delocalized band provides a truer picture. The fact that at least up to 8% Fe the temperature dependence of the hyperfine field is best fit with unvarying spin of 1 appears to argue against the existence of giant moments,[6] but further investigation is needed.

In summary, we believe that the magnetic behavior of Au-Fe is qualitatively described as follows: In the concentration range 0 to approximately 18% Fe, the ordering in the absence of an applied field is random antiferromagnetic. Partial ferromagnetic alignment can be induced by an applied field, and the degree of order depends upon field strength, temperature, and composition. If the temperature is sufficiently low, hysteresis loops are obtained, presumably as the result of exchange anisotropy. This partial alignment with the field cannot be the result of a complete alignment of a substantial fraction of the Fe moments with the remaining fraction preserving its initial random orientation. Such a mechanism would lead to a straightforward diminution in the intensities of lines 2 and 5; this is not observed. The polarized Mössbauer spectra are consistent with a partial realignment of all the moments so as to present on the average a component parallel to the applied field. For these reasons as well as the absence of a blocking temperature, and also because of lack of evidence for giant moments, we do not regard this system as a conventional superparamagnet. Finally, for alloys containing more than ~5% Fe, it is necessary to invoke an additional moment beyond that which may reasonably be ascribed to Fe alone in order to reconcile the Mössbauer and magnetization experiments. This is equivalent to saying that the host lattice, i.e., Au, acquires a magnetic moment. Additional evidence for the latter proposal is the large apparent increase in the Au hyperfine field with increasing concentration of Fe below the magnetic ordering temperature.

It is a pleasure to acknowledge collaborations with Dr. Tom Kitchens, Brookhaven National Laboratory, and Dr. Charles Violet and Dr. David Pipkorn, Lawrence Radiation Laboratory.

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