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Neutron spin depolarization studies in Au–Fe alloys near the percolation limit

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Abstract. Neutron spin depolarization measurements have been made on Au–Fe alloys as a function of temperature in the concentration range between 15 and 19% Fe. Results show that alloys containing 17 and 19% Fe do not cease to be ferromagnetic at any temperature below T_c. The 15% Fe alloy, which lies just below the percolation limit for ferromagnetic ordering, reveals an anomalous magnetic property of a distinct type at two different temperatures. On cooling below the temperature T^* ($\simeq 90$ K), a large depolarization in the neutron beam occurs owing to the formation of large but non-divergent ferromagnetic clusters. On further cooling, these clusters break up into small regions which then freeze into a spin-glass behaviour below the temperature T_f ($=25$ K). This change in the nature of the clusters in the 15% Fe alloy is due to the significant effects of the frustrations caused by the long-range RKKY interactions between the spin system, whereas such effects are far less important than the strong d–d interactions responsible for the pure percolation process in the 17 and 19% Fe alloys.

1. Introduction

The magnetic properties of dilute Au–Fe alloys with a concentration of iron near and above the magnetic percolation limit for ferromagnetic order have been most extensively studied by means of a variety of experimental techniques (see, e.g., Murani *et al* 1976, Coles *et al* 1978, Sarkissian 1979, 1981, Mirza and Loram 1985). Alloys above the percolation limit show many complex magnetic features which are found to be based on the idea of the formation of an infinite ferromagnetic cluster at T_c, coupled to finite clusters by exchange fields which are much smaller than the strong direct d–d interaction which couples iron spins within the infinite cluster. At temperatures close to the T_c of the infinite cluster, the relaxation time of finite clusters is short and the dynamic behaviour of the alloy is dominated by the slow relaxations of the spins within the infinite cluster. On cooling below T_c, the finite clusters slow down and freeze into spin-glass-like magnetism below the temperature T_f, but the state of the infinite cluster is not broken up (Coles *et al* 1978, Sarkissian 1979, 1981). The T-dependence evolution of the freezing process of the finite clusters manifests itself in a variety of experiments; for example, it reveals anomalies in the T-dependent AC susceptibility and specific heat curves (Sarkissian 1981, Mirza and Loram 1985) which are analogous to those causing the increase in the intensity

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of the small-angle neutron scattering (Murani *et al* 1976). Such clusters also influence the resonance relaxation process and the spin-wave excitation of the infinite cluster as seen in ferromagnetic resonance (Sarkissian 1979, Continentino 1983) and inelastic neutron scattering studies (Murani 1983), respectively. Alloys containing less than or about 15% Fe (15–13% Fe) also have a rather complex magnetic behaviour at T^* different from that of the infinite-cluster magnetism, but correspond more closely to the behaviour associated with very large but finite clusters at temperatures well above the spin-glass freezing temperature T_f. Alloys with less than 13% Fe are close to canonical spin glasses at sufficiently low temperatures (Sarkissian 1981).

Remarkably, various systems (Rainford *et al* 1982, Aeppli *et al* 1983, Sarkissian 1984) also have features similar to those of Au–Fe and thus show similar physical phenomena. Various models, different from that of the cluster model discussed above, have been proposed to describe the magnetism of the dilute alloys. In the random field picture (Sherrington and Kirkpatrick 1975), the high-temperature ferromagnetic order is destroyed at low temperatures, giving way to the so-called re-entrant spin-glass state. In contrast with the above model, the mean-field theory of Gabay and Toulouse (1981) suggests that the long-range ferromagnetic order is preserved at all temperatures below T_c, but only along the longitudinal spin components, while the transverse component remains paramagnetic and freezes into a spin-glass state at a lower temperature T_{GT}; in the Gabay–Toulouse model, the freezing of the transverse spin components gives rise to the appearance of magnetic irreversibility at T_{GT} and then, at a temperature lower than T_{GT}, another transition occurs at T_f which is marked by the onset of a stronger-irreversibility regime. The Gabay–Toulouse state can be identified as a random ferrimagnetic state characterized by the coexistence of a non-vanishing net magnetization and random static fluctuations in the spin direction.

In order to have a clear picture of the state of order in Au–Fe alloys, we have carried out neutron spin depolarization studies on alloys containing 15, 17 and 19% Fe. In such a study, the change in the polarization of the transmitted neutron beam through the specimen gives direct information on the characteristics of domains and clusters. The details of these studies will be presented and discussed in section 3.

2. Experimental details

Measurements were carried out on the IN11 neutron spin echo spectrometer using polarized neutrons of wavelength 4.7 Å at the high-flux reactor of the Institut Laue–Langevin, Grenoble, France. The specimens used in this study were those used previously in other neutron scattering studies (Murani *et al* 1976). To avoid effects associated with the field cooling, we first cooled the specimens to the lowest temperatures (about 1.5 K) in a truly zero field (i.e. the earth field was eliminated). The measurements were then carried out while warming the specimen gradually to the paramagnetic temperature region. The quantity measured is the flipping ratio $R = I^+/I^-$, defined as the ratio of the transmitted neutron intensities for the two neutron spin orientations, parallel and antiparallel, respectively, to the small (2 Oe) applied field. The polarization P is simply related to the measured flipping ratio by $P = (R-1)/(R+1)$.

Measurements of the elastic diffuse scattering intensities $I(Q)$ at a fixed Q were also made by observing the scattering cross section as a function of T with the instrument set to measure both the spin-up (I^+) and the spin-down (I^-) cross-section at a fixed scattering angle. The sum of I^+ and I^- (i.e. $I^+ + I^-$) intensities represents an integration of the

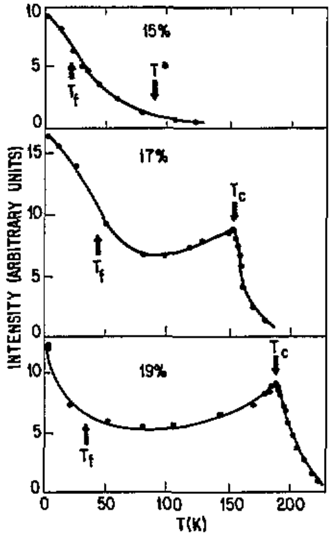

Figure 1. Temperature dependence of the scattering intensity $I(Q)$ ($=I^+ + I^-$; see section 2) at a Q-value of 0.04 Å for the 15, 17 and 19% Fe alloys. The arrows show the temperatures at which the AC susceptibility measurements (Sarkissian 1979) reveal the corresponding magnetic behaviour.

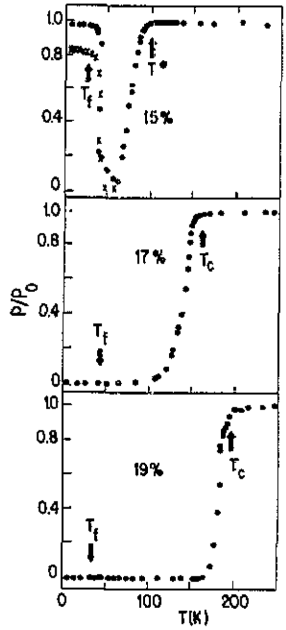

Figure 2. Relative depolarization ratio P/P_0 as a function of the temperature for 15, 17 and 19% Fe alloys: ×, data for the 15% Fe alloy obtained after cooling to the lowest temperature in a field of 2 Oe. The arrows have the same meaning as in figure 1.

scattering at a fixed scattering angle over the final neutron energies and is equivalent to the cross section $I(Q)$ obtained by unpolarized small-angle neutron scattering experiments.

3. Results and discussion

On subtracting the background (determined from the high-T data), the magnetic $I(Q)$ data alone were revealed for alloys containing 15, 17 and 19% Fe. These are shown in figure 1, as a function of temperature for a Q-value of 0.04 Å^{-1}. Critical scattering peaks were observed for alloys with 17 and 19% Fe, but not for the 15% Fe alloy. Instead the 15 Fe alloy showed only an increase in the intensity with a decrease in temperature. This confirms the fact that long-range ferromagnetic ordering occurs in the 17 and 19% Fe specimens but not in the 15% Fe specimen. The results in figure 1 also show a continuous increase in the intensity with a decrease in temperature for the 17 and 19% Fe alloys due to freezing of the finite clusters as initially observed by Murani *et al* (1976). Figure 2 shows the depolarization data for the 19, 17 and 15% Fe alloys plotted as P/P_0, as a function of temperature, where P_0 is the polarization measured at high temperatures. The data indicate that, when the 17 and 19% Fe specimens are cooled towards their T_c, P/P_0 rapidly decreases and, on further cooling below T_c, the neutron beam becomes completely depolarized. This is due to the onset of ferromagnetic domain walls below T_c and is caused by the averaging effects of many spin rotations over the number of domains passed by the neutrons. These results are very similar to those obtained for amorphous $(Fe_{0.765}Mn_{0.235})_{75}P_{16}B_6Al_3$ alloy of the same type using polarized neutrons of a much smaller wavelength (Mirebeau *et al* 1986).

It is interesting that even for the 15% Fe alloy, just below the temperature $T^* =$ 90 K, P/P_0 data show a feature similar to that observed for 17 and 19% Fe alloys with the difference that P/P_0 stays non-zero below T^*; thus, the large clusters of a finite size reveal their presence in the 15% Fe alloy; this is consistent with the large but finite response observed earlier in the low-field AC susceptibility measurements at the same temperature in an alloy of the same composition (Sarkissian 1981). On further reduction in the temperature, this alloy shows an interesting anomalous feature of a distinct nature at 50 K; P/P_0 increases abruptly owing to a break-up of the large clusters into smaller regions. At lower temperatures, P/P_0 saturates to values close to its paramagnetic value, reflecting a spin-glass-like onset below the temperature $T_f = 25$ K just where the AC susceptibility data show the spin-glass property (Sarkissian 1981). The neutron spin echo studies performed on the 15% Fe alloy show that the cluster dynamics associated with the low-temperature blocking process (39 K $\geqslant T \geqslant$ 1.5 K) have a Kohlrausch 'stretched'-exponential form $\exp[-(t/\tau)\beta]$ (with $\beta = 0.6$), which is similar to those observed in glasses of broken ergodicity (Sarkissian 1990).

To study further the low-temperature spin-glass behaviour of the 15% Fe alloy, similar P/P_0 measurements were performed after the specimen had been cooled to the lowest temperature (about 1.5 K) in a field of 2 Oe. The data obtained (figure 2) indicate that P/P_0 saturates below T_f to values much smaller than those obtained in the case of zero-field-cooled state. The results show a strong magnetic-thermal-history-dependent property which can be identified with the blocking of the clusters below T_f. This may also indicate a considerable increase in the size of the frozen clusters. However, the case is rather different in the 17 and 19% Fe alloys, where similar 2 Oe field-cooling studies have failed to detect any changes in the behaviour of the depolarization curves (figure 2). This suggests the absence of magnetic irreversibility in the depolarization behaviour for the 17 and 19% Fe alloys for a field of 2 Oe.

We have attempted to analyse the P/P_0 data of the 15% Fe alloy in terms of the formula of Halpern and Holstein (1941), in order to determine the average cluster size Λ below the temperature T^*. Here the depolarization $\Delta P/P_0$ may be expressed as

$$\Delta P/P_0 = \exp(-\gamma^2 B_s^2 \Lambda d/3v^2) \tag{1}$$

where γ is the neutron gyromagnetic ratio, B_s is the saturation magnetization, d is the specimen thickness and v is the neutron velocity. To estimate Λ, we proceed by using the B_s-value of $1.5\mu_B$ for the magnetic moment of Fe obtained from the saturation magnetization measurements of Crangle and Scott (1964). Equation (1) then gives a Λ-value of the order of 3000 Å. An interesting quantity that can be also extracted from the P/P_0 data for the 15% Fe alloy is the average frozen cluster size Λ_{cl} for the zero-field-cooled and field-cooled states at $T = 0$. This can be obtained using Maleyev's (1982) depolarization expression for spin glasses, where Λ_{cl} is related to the change ΔP in depolarization as

$$\Lambda_{cl} = 3/(2\pi)^2(\lambda^2/dc_0^{1/3})(\varepsilon_0/\mu_n B_{cl})^2\, \Delta P. \tag{2}$$

Here c_0 is the atomic concentration of the iron and B_{cl} is the average induction of the cluster. If we take $B_{cl} = 5$ Oe (this value is deduced from the change in the neutron

Larmor precession angle observed in the neutron spin echo studies on the same 15% Fe specimen (Sarkissian 1991)), then equation (2), for the zero-field-cooled state data and for $\lambda = 4.7$ Å, gives a Λ_{cl}-value of about 30 Å at $T = 0$, whereas a similar analysis for the 2 Oe field-cooled state data gives $\Lambda_{cl} = 200$ Å at $T = 0$.

Our depolarization data confirm directly that alloys containing 17 and 19% Fe do not cease to be ferromagnetic at any temperature below T_c. This is because there is no indication that the large ferromagnetic domains formed below T_c break down or are reduced in size at low temperatures. This rules out the earlier ideas of the 're-entrant' transition from the ferromagnetic to the spin-glass state in the Au–Fe alloy system. This is likely to be proved true for other alloy systems of the same type. Our depolarization data do not indicate any anomaly which could be associated with the occurrence of the low-temperature Gabay–Toulouse random ferrimagnetic state (see section 1) in the 17 and 19% Fe alloys; however, these studies alone cannot also disprove the existence of such a state below T_f. This is because the freezing effects of the transverse spin components at T_f do not give rise to a further reduction in the complete depolarization of the neutron beam which is dominated by the domain walls associated with the ferromagnetic ordering of the longitudinal spin components.

We believe that the cluster model discussed above provides a natural explanation for our depolarization data for the 17 and 19% Fe alloys. The freezing effects of finite clusters do not seem to show up in the P/P_0 data of these alloys. The reason for this is that these effects are masked by the depolarization of the domain walls within the infinite cluster. The strong effects which are seen at low T, e.g. in the AC susceptibility, magnetic resonance and inelastic neutron scattering studies mentioned above, must then be identified with the constraint imposed on the spin dynamics of the infinite cluster by the freezing effects of the finite clusters (Coles *et al* 1978, Sarkissian 1979, Sarkissian 1981, Continentino 1983, Murani 1983).

As argued above, the percolation cluster model is adequate for describing the magnetic behaviour of the Au–Fe alloys but besides the ferromagnetic d–d couplings there are also the long-range RKKY interactions (between more distant Fe spins) which yield a competing ferromagnetic and antiferromagnetic couplings within the spin system. As a result, some spins will be highly frustrated and may thus experience relatively weak overall interactions. Thus these frustrations can weaken direct d–d coupling of the spins within the clusters and may cause the clusters to be decoupled into smaller regions as their strength increases with decreasing temperature. Our experimental results of alloys containing 17 and 19% Fe confirm that frustration effects do not dominate the interactions responsible for the pure percolation process. This is because the frustration effects can be significant only on length scales less than the infinite ferromagnetic correlation length within the infinite cluster. In contrast with the 17 and 19% Fe alloys, in the 15% Fe specimen, the break-up of the size of finite clusters below 50 K (which are formed at T^*) suggests that frustration effects dominate over the direct d–d coupling of the spins within the finite clusters and thus prevent such clusters from maintaining their low-energy configuration at low temperatures.

4. Conclusion

In conclusion, the neutron spin depolarization data obtained for Au–Fe indicate that alloys which show a long-range ferromagnetic order, e.g. Au–Fe alloys with 17 and

19% Fe, keep their state of order at all temperatures below T_c. We believe that the low-T anomaly observed in various experimental studies is due to the constraint imposed on the spin dynamics of the infinite cluster by the freezing effect of finite clusters. The 15% Fe specimen, which lies just below the percolation limit, shows a significant change in the magnetic behaviour with changes in temperature. This is manifested as a break-up in the size of the finite clusters which then freezes into a spin-glass-like behaviour at low temperatures. We have estimated the average size of the frozen clusters to be of the order of a few tens of ångströms. The change in the nature of the clusters in the 15% Fe alloy is due to the frustration caused by the long-range RKKY interactions within the spin system. A theory which incorporates both the dynamic constraint and the frustration effects relevant to the Au–Fe system has yet to be found.

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References

Aeppli G, Shapiro S M, Birgeneau R J and Chen H S 1983 *Phys. Rev.* B **28** 5160
Coles B R, Sarkissian B V B and Taylor R H 1978 *Phil. Mag.* B **37** 489
Continentino M A 1983 *J. Phys. C: Solid State Phys.* **16** L71
Crangle J and Scott W R 1964 *Phys. Rev. Lett.* **12** 126
Gabay M and Toulouse G 1981 *Phys. Rev. Lett.* **47** 201
Halpern O and Holstein T 1941 *Phys. Rev.* **59** 960
Maleyev S V 1982 *J. Physique Coll. Suppl. Coll. 12* C7 23
Mirebeau I, Jehanno G, Campbell I A, Hippert F, Hennion B and Hennion M 1986 *J. Magn. Magn. Mater.* **54–7** 99
Mirza K A and Loram J W 1985 *J. Phys. F: Met. Phys.* **15** 439
Murani A P, Roth S, Radhkrishna P, Rainford B D, Coles B R, Ibel K, Goeltz G and Mezei F 1976 *J. Phys. F: Met. Phys.* **6** 425
Murani A P 1983 *Phys. Rev.* B **28** 432
Rainford B D, Burke S K, Davis J R and Haworth W 1982 *Neutron Scattering* (*AIP Conf. Proc. 89*) (New York: American Institute of Physics) p 232
Sarkissian B V B 1979 *Phil. Mag.* B **39** 413
—— 1981 *J. Phys. F: Met. Phys.* **11** 2191
—— 1984 *J. Appl. Phys.* **55** 1738
—— 1990 *J. Phys.: Condens. Matter Phys.* **2** 7873
—— 1991 to be published
Sherrington D and Kirkpatrick S 1975 *Phys. Rev. Lett.* **35** 1792