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Nuclear orientation of Mn and Co in spin glass and re-entrant AuFe

M Trhlík†‡, P De Moor‡, J Camps‡, A L Erzinkyan§, G M Gurevich‖, V P Parfenova§, P Schuurmans‡, N Severijns‡, W Vanderpoorten‡ and L Vanneste‡

† Department of Low Temperature Physics, Charles University, V Holešovičkách 2, 180 00 Prague 8, Czech Republic

‡ Instituut voor Kern-en Stralingsfysika, K U Leuven, Celestijnenlaan 200D, B-3001 Leuven, Belgium

§ Institute of Nuclear Physics, Moscow State University, 119899 Moscow, Russia

‖ Institute for Nuclear Research, Russian Academy of Sciences, 117312 Moscow, Russia

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Abstract. Low-temperature nuclear orientation has been used to study the magnetic behaviour of ^{54}Mn and ^{57}Co impurities in the **Au**Fe alloy, below (14 at.% Fe—spin-glass state) and above (18 at.% Fe—re-entrant state) the percolation threshold. The anisotropy of the γ-rays was monitored in the direction of the external magnetic field, as a function of the external magnetic field B_{ext} up to 8.5 T and of the temperature in the range 4–40 mK. In $Au_{86}Fe_{14}$ we have found total misalignment of the Mn and Co spins when $B_{ext} \rightarrow 0$ and a rather easy alignment of Mn and Co with the application of B_{ext}, which is typical for a spin-glass behaviour. On the other hand, both Mn and Co spins in $Au_{82}Fe_{18}$ show an alignment (mean value ~32°), even at $B_{ext} \rightarrow 0$, and are turned only slowly by B_{ext} which corresponds to the re-entrant state. The difference in the Mn and Co results can be interpreted as an indication of the existence of local correlations between the Fe spins, supporting the necessity of using the finite-range model for the re-entrant state.

1. Introduction

While **Au**Fe alloys with Fe concentration c_{Fe} below the percolation threshold of 15 at.% are generally accepted as spin glasses, the magnetic behaviour of the more concentrated alloys (c_{Fe} = 15–24 at.%) is much more complicated and has not been completely established until now. Magnetic [1] and Mossbauer spectroscopy (ME) [2–4] results have shown that after a paramagnetic-ferromagnetic transition at higher temperature T_C (~160 K for Au + 18 at.% Fe) there exists a second transition at lower temperature T_f (~30–50 K for Au + 18 at.% Fe). This transition is characterized by a decrease of susceptibility and by an anomalous increase of an average hyperfine field B_{hf} on Fe, as was deduced from the ME measurements. Moreover, ME [2, 3, 5] shows a noncollinearity of the Fe spins below T_f. These experiments were interpreted as a proof for the existence of a new canted phase (i.e., re-entrant spin glass) below T_f. Also neutron depolarization studies (e.g., [6]) have found a re-entrant state within the domains at low temperature.

The infinite-range mean-field 3D Heisenberg spin model of Gabay and Toulouse (GT) [7] has given a good explanation for the behaviour mentioned above. It predicts that, when both ferromagnetic and antiferromagnetic exchanges are present in a system, with

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the ferromagnetic interactions dominating weakly, a spin-glass ordering in the transverse magnetic moment components is established below some temperature T_f, while the longitudinal ones are already ferromagnetically ordered at higher temperature T_C. Thus, at very low temperatures the magnetic moments are supposed to be locally misaligned with each other and even a rather high external magnetic field B_{ext} up to 3 T does not break this disorder [5].

This picture was questioned by Beck [8], who proposed an alternative model of Fe-rich ferromagnetic clusters, present in Au–Fe alloys, interacting weakly with each other as superparamagnetic particles and being responsible for the observed magnetic double transition. Nevertheless, on the basis of later experimental data [9, 10] a crucial role of atomic clustering in the re-entrant transition can be excluded.

Further hyperfine interaction studies with nonmagnetic probes in **Au**Fe alloys of the re-entrant concentration region (^{197}Au [11], ^{119}Sn [12] and ^{100}Rh [13]) detected both paramagnetic–ferromagnetic and ferromagnetic–re-entrant transitions proposed by GT. Simultaneously however, they gave an argument for a certain degree of local correlation among the Fe spins. This was concluded from the fact that the temperature dependence of B_{hf} on these nonmagnetic impurities, which should feel the transferred hyperfine field proportional to $|\sum_i \langle S_i \rangle|$ (S_i are the Fe spins in the nearest neighbourhood of the nonmagnetic impurity), closely followed that on Fe. This local correlation cannot be predicted by the GT model because of its infinite-range character. These experiments have again revealed the weakly coupled superparamagnetic clusters model with correlated spins within a cluster [12], which now is believed to be established even when the Fe atom distribution is completely random. Although this cluster model could successfully explain the large change of T_C with increasing B_{ext}, which had been observed in the ME experiments [12, 14], it could give no explanation for the observed B_{hf} anomaly below T_f.

Although much theoretical effort has been devoted to tackling more realistic spin systems, i.e. the 3D dilute Heisenberg spins with finite-range competing coupling, where the ferromagnetic coupling prevails (see e.g. review [15]), the problem of the re-entrant state has not yet been solved completely. Up until now the re-entrant state has been obtained numerically for the 3D Heisenberg spins with competing short-range coupling, where the ferromagnetic coupling dominates [16], but no percolation threshold has been found. Recently, numerical simulations of the dilute classical Heisenberg spins (concentration 16 at.%) in a fcc lattice with finite long-range RKKY coupling [17] have shown a spin transverse component freezing below the temperature $\sim 0.2T_C$, which should correspond to the ferromagnetic–re-entrant transition.

The low-temperature nuclear orientation (NO) technique can, in principle, give information on the local directions of chosen magnetic moments and distinguish them from other moments of a given alloy (see e.g., the review in [18]). NO techniques can study their behaviour under B_{ext} at low temperature, which in some cases (including **Au**Fe) corresponds to the magnetic moment's ground state. In NO experiments the angular distribution, $W(\theta, T)$, of the γ-rays of radioactive probe nuclei at sufficiently low temperature T is measured. The anisotropy $W(\theta, T)$ can be expressed by the formula

$$W(\theta, T) = 1 + Q_2 A_2 B_2 (E_m/kT) P_2(\cos\theta) + Q_4 A_4 B_4 (E_m/kT) P_4(\cos\theta) \tag{1}$$

where Q_k and A_k (B_k) are the known constants (functions) conventionally defined in the literature on NO techniques (e.g. [18]). E_m is the hyperfine splitting energy defined by

$$E_m = g_N \mu_N B_{tot} \tag{2}$$

with

$$B_{tot} = B_{hf} + B_{ext} \tag{3}$$

where B_{tot} is the total magnetic field acting on the nuclei, while B_{hf} and B_{ext} are the hyperfine and external magnetic fields, respectively. The functions P_k are the Legendre polynomials and θ is the angle between B_{tot} and the γ-ray observation direction.

The hyperfine interaction studies of AuFe alloys (ME on Fe [2–5], ME on nonmagnetic atoms of Au [11] and Sn [12] and PAC on Cd [19] and Rh [13]) have provided fruitful, but in some aspects controversial, information about their magnetic properties. In this paper we present an NO experimental study of the behaviour of magnetic Co and Mn impurities in two Au–Fe alloys with the Fe concentration just around the percolation threshold (i.e. 14 at.% Fe and 18 at.% Fe), in B_{ext} up to 8.5 T (the results of the measurements in low external field were published in [20, 21]). A misalignment parameter at the ground state as a function of B_{ext} can be unambiguously derived. Some information about the spin correlation was also obtained.

2. Experiment

The alloys $Au_{86}Fe_{14}$ and $Au_{82}Fe_{18}$ were prepared by melting the corresponding quantities of Au and Fe in an Ar arc-furnace. The alloys were then remelted with the ^{57}Co and ^{54}Mn activities in vacuum (the total amount of Co and Mn impurities introduced was estimated to be several ppm), cold-rolled, annealed for several hours at 900 °C under Ar atmosphere and then rapidly quenched in water. The samples, in the form of foils, were soft-soldered with Wood's metal onto the cold finger of a dilution refrigerator with the foil surfaces parallel to the B_{ext} direction.

The NO experiments were performed using the off-line NO facility at Leuven in the temperature range 4–40 mK and with B_{ext} up to 8.5 T. A ^{60}Co:Co single-crystal NO thermometer and a pure Ge detector, placed in the B_{ext} direction, were used.

The γ-ray anisotropies W of the 835 keV ^{54}Mn and 136 keV ^{57}Co lines were measured as functions of T and B_{ext}. A typical example of such results is given in figure 1.

3. Results and their analyses

3.1. Mn case

The most straightforward analysis of the experimental W-data supposes a full collinearity between B_{hf} and B_{ext} ($\theta = 0°$ in (1)). Using (1–3), values for B_{hf} can then be extracted from the experimental anisotropies W (one can suppose that the B_{hf}-value on Mn nuclei is negative). When doing this, a strong temperature dependence of B_{hf} for fixed B_{ext} was found. Moreover, the B_{hf}-values were much lower than one could expect from comparison with the B_{hf}-value on Mn in pure Au (i.e. 39.8 T [22]). The strong temperature dependence of B_{hf} indicates that the extraction of B_{hf} from W using (1) with $\theta = 0°$ is not correct. To describe the data properly, some noncollinearity of B_{hf} with respect to B_{ext} (= the 0° direction) must be assumed (see, for example [23]).

As a next step we used the fact that a possible misalignment between B_{hf} (= the Mn magnetic moment direction) and B_{ext} should be constant in our very low temperature range. In this case we can obtain both the B_{hf}-value and some misalignment parameter when fitting the temperature dependence of W at fixed B_{ext} using (1).

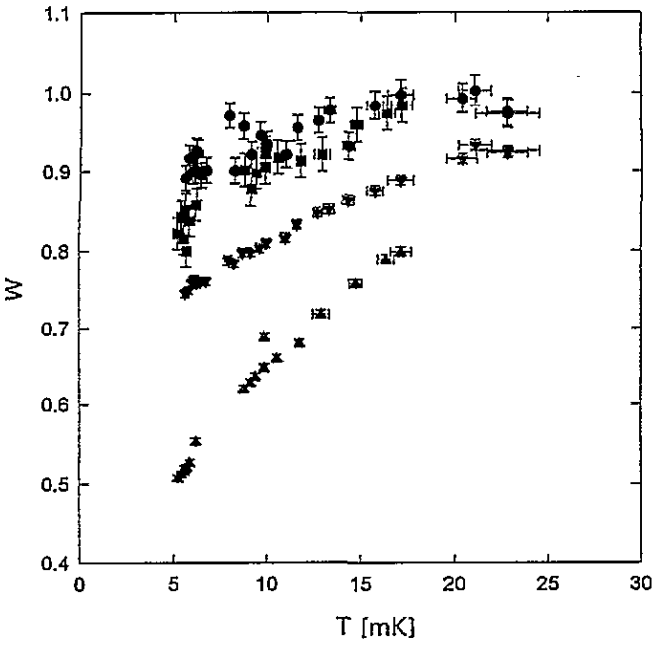

Figure 1. The temperature dependence of W for $B_{ext} = 7$ T. ▼—^{54}Mn:Au$_{86}$Fe$_{14}$, ▲—^{54}Mn:Au$_{82}$Fe$_{18}$, ●—^{57}Co:Au$_{86}$Fe$_{14}$, ■—^{57}Co:Au$_{82}$Fe$_{18}$. Where not shown, the errors are smaller than the symbol size.

First, we supposed there to be only one B_{hf} and one misalignment angle α_1 (the angle between B_{hf} and B_{ext}; θ is then calculated using (3)) and performed a two-parameter fit on the temperature dependencies of W. This resulted in rather bad χ^2-values and no reasonable values for B_{hf} and α_1 nor their B_{ext} dependencies were found, such that we had to exclude this simple model, too.

As a further improvement of this approach we then supposed some distribution of misalignment angles. We applied the same distribution that was used already before for describing the misalignment of Fe spins in the similar re-entrant alloy Au$_{83.2}$Fe$_{16.8}$ [5], where the canting-angle distribution $P(\alpha)$ has a maximum at $\alpha = 0°$ and goes to zero at the upper limit α_0:

$$P(\alpha) = \begin{cases} \cos^2(\pi\alpha/2\alpha_0) & \text{for } \alpha < \alpha_0 \\ 0 & \text{for } \alpha \geqslant \alpha_0. \end{cases} \tag{4}$$

The mean value $\langle\alpha\rangle$ is given by

$$\langle\alpha\rangle = \int_0^{\alpha_0} \sin\alpha P(\alpha)\alpha \, \mathrm{d}\alpha \Big/ \int_0^{\alpha_0} \sin\alpha P(\alpha) \, \mathrm{d}\alpha. \tag{5}$$

Using this distribution for the angle α, we fitted the temperature dependencies of W using (1–4) with two free parameters, i.e. B_{hf} and α_0. The fits gave reasonable results with χ^2-values of approximately unity, supporting the proposed canting-angle distribution. The resulting values of B_{hf} and $\langle\alpha\rangle$ are shown in figures 2 and 3.

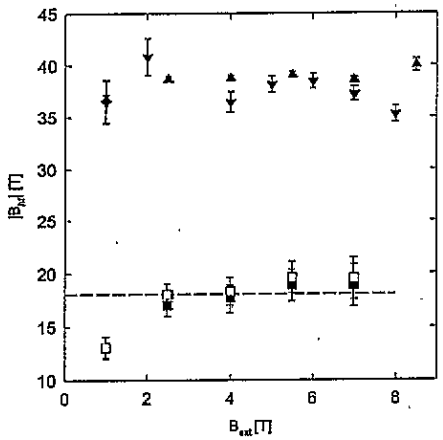

Figure 2. The B_{ext}-dependence of B_{hf}—as is obtained from the two-parameter fits (see the text). ▼—^{54}Mn:Au$_{86}$Fe$_{14}$, ▲—^{54}Mn:Au$_{82}$Fe$_{18}$, ■—^{57}Co:Au$_{82}$Fe$_{18}$ (fraction f_{cl} used), □—^{57}Co:Au$_{82}$Fe$_{18}$ (fraction f_{is} used). The dotted bar corresponds to fixed $B_{hf} = -18$ T. When not shown, the errors are smaller than the symbol size.

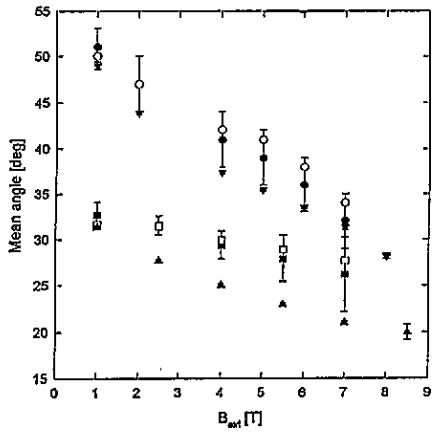

Figure 3. The B_{ext} dependence of the mean angle (α)—as is obtained from the two-parameter fits (see the text). ▼—^{54}Mn:Au$_{86}$Fe$_{14}$, ▲—^{54}Mn:Au$_{82}$Fe$_{18}$, ●○—^{57}Co:Au$_{86}$Fe$_{14}$, ■, □—^{57}Co:Au$_{82}$Fe$_{18}$. Open symbols—fraction f_{cl} used, fixed symbols—fraction f_{is} used. When not shown, the errors are smaller than the symbol size.

3.2. Co case

The magnetic state of Co in Au alloys is more complicated. Unlike in the Mn case, where the Kondo coupling is very weak ($T_K \sim 2.5$ mK [22]), the Kondo coupling of Co in Au is very large ($T_K \sim 700$ K for isolated Co impurities and $T_K \sim 20$ K for isolated Co pairs in Au [24]). Boysen *et al* [25] have interpreted their NO data of ^{60}Co:**Au**Co supposing that the Co atom is nonmagnetic ($B_{hf} = 0$) if it does not belong to a 'Co cluster', which they define as consisting of at least three Co atoms with first-neighbour interaction. The Co atoms in these 'Co clusters' are supposed to be fully magnetic ($B_{hf} \sim -18T$). An application of the binomial law for randomly distributed fcc alloys gives the fraction f_{cl} of magnetic Co atoms as

$$f_{cl} = 1 - (1 - c_{Co})^{12} - 12c_{Co}(1 - c_{Co})^{18} \tag{6}$$

If we follow this approach for our **Au**Fe alloys and assume the behaviour of Co and Fe atoms to be equal, we have $f_{cl} = 0.725$ and 0.847 for Au$_{86}$Fe$_{14}$ and Au$_{82}$Fe$_{18}$, respectively.

On the other hand, the Fe magnetic moments do not show Kondo coupling in Au and one could therefore object that condition (6) is too strong, and that for magnetic Co atoms it is enough to have at least one Fe atom in its first-neighbour region. Condition (6) is then replaced by

$$f_{is} = 1 - (1 - c_{Fe})^{12} \tag{7}$$

which leads to $f_{is} = 0.836$ and 0.908 for Au$_{86}$Fe$_{14}$ and Au$_{82}$Fe$_{18}$, respectively.

Taking into account Co Kondo coupling (only a fraction f_{cl} or f_{is} of the Co atoms feels nonzero B_{hf}), we have carried out the same analyses for our NO on Co data as for the Mn ones. Similarly, the first two models (full collinearity and only one misalignment angle) appeared to fail. Finally, the temperature dependencies of W have been fitted using (1)–(4) with two free parameters—B_{hf} and α_0. Because of the above mentioned uncertainty in the

fraction of magnetic Co atoms, we have carried out fits with both fractions (f_{cl} and f_{is}). The resulting values of B_{hf} and $\langle\alpha\rangle$ for $^{57}Co{:}Au_{82}Fe_{18}$ are shown in figures 2 and 3.

For $^{57}Co{:}Au_{86}Fe_{14}$ the effect on W was rather small (see figure 1). Therefore the two-parameter fits of the temperature dependencies of W resulted in B_{hf}- and $\langle\alpha\rangle$-values with large errors. Hence in the case of the $^{57}Co{:}Au_{86}Fe_{14}$ sample we have fitted only one parameter (i.e. α_0) with B_{hf} fixed at -18 T. The corresponding $\langle\alpha\rangle$-values are again shown in figure 3.

4. Discussion

As one can see from figure 2, the B_{hf}-values of ^{54}Mn in both **Au**Fe alloys lie around the value of -38 T, which is close to the B_{hf}-value of Mn in pure Au [22] and also in other alloys based on similar metals (e.g. in **Pt**Fe $B_{hf} = -40(1)$ T [23]). Similarly, the B_{hf}-values of $^{57}Co{:}Au_{82}Fe_{18}$ are close to the value of -18 T that is found in the **Au**Co alloys [25]. This coincidence indicates that our presumption of misaligned localized magnetic moments, the canted-angle distribution of which could be described by (4), is not far from the real situation. We have found that in the case of Co, the B_{hf}-values (see figure 2) (and also the $\langle\alpha\rangle$-values—see figure 3) are not too sensitive to the fraction used (i.e. (6) and (7)).

As one can see from figure 3, there is a large misalignment of Co and Mn spins in both alloys studied, which is present in the whole B_{ext}-range. In the case of $Au_{86}Fe_{14}$ the misalignment is larger but also more easily affected by B_{ext} than in the case of $Au_{82}Fe_{18}$.

We can suppose that due to the Co–Fe ferromagnetic interactions the Co spins take the direction of the Fe ones and can therefore give information about the misalignment of the Fe spins in the ground state ($T \to 0$). Taking this into account, we see a typical spin-glass behaviour for $Au_{86}Fe_{14}$—the angles are nearly randomly distributed at low B_{ext} and are slowly aligned by higher B_{ext} (compare the measurements in high magnetic field of the Au–Fe spin glasses in, e.g., [26]).

The $Au_{82}Fe_{18}$ alloy, on the other hand, shows some spin alignment ($\langle\alpha\rangle \sim 32°$) even at low B_{ext}. This alignment is rather fixed and can be changed only slowly by increasing B_{ext}. This result brings independent evidence in support of the existence of a re-entrant state at lower temperatures. We have used a different experimental techniques and a different analysis method, but still obtained a similar misalignment as has been previously obtained by e.g. Lange *et al* [5], who have studied the misalignment of the Fe spins in the alloy $Au_{83.2}Fe_{16.8}$ by ME at temperature of 4.2 K. Using the same canting-angle distribution (4–5) they have obtained $\langle\alpha\rangle = 25.0(15)°$ for B_{ext} up to 3 T. Taking into account the somewhat different composition of the alloy we consider this value to be in reasonable agreement with our $\langle\alpha\rangle$ -value for low B_{ext}. Their reported change in the mean canting angle ($\Delta\langle\alpha\rangle \leqslant 1.5°$ in B_{ext} up to 3 T) does not contradict our results. On the other hand, when looking at our values of $\langle\alpha\rangle$ for high B_{ext}, we see a nonzero slope (B_{ext} slowly aligns the spins), which can be estimated as $\sim 0.5°$–$1.0°$ per tesla (depending on the fraction used). Unfortunately, this effect has not yet been estimated by the theory.

Unlike those in Co, the Mn spins should couple antiferromagnetically with their first Fe neighbours, in a way similarly to, e.g., Mn in **Pt**Fe, where the first-neighbours Mn–Fe exchange parameter has been found to be negative [23]. Then the Fe spin subsystem should behave differently from the Mn one and the response of Mn spins to B_{ext} should be different from the response of the Fe ones that were monitored by Co. This behaviour is indeed observed in our results, where the Mn spins are somewhat more easily turned by B_{ext} than the Co ones (see figure 3). On the other hand, one can notice that the $\langle\alpha\rangle$-values of both Mn and Co are practically identical for $B_{ext} \to 0$ in the case of re-entrant

$Au_{82}Fe_{18}$ (the coincidence in the case of spin-glass $Au_{86}Fe_{14}$ is natural as both Mn and Co spins should be randomly distributed there). If there is no local correlation of the Fe spins, the Mn ones, which are embedded in this magnetic system and have a predominantly antiferromagnetic coupling with it, should have their own angle distribution around the B_{ext} direction and the coincidence of the Mn and Co distribution parameters at $B_{ext} \rightarrow 0$ would have been only accidental. On the other hand, if we suppose the existence of 'magnetic' regions, which consist of locally correlated Fe spins and which are misaligned to each other, we can imagine that the Mn spins will have some distribution within this region around its magnetization direction. The mean Mn spin misalignment will then be identical to that of the Co spins which is indeed what we observe. Therefore, we interpret our results as a further support for the occurrence of local correlations of the Fe spins in the re-entrant **Au**Fe alloys, as was also concluded in [11–13].

Although the GT model has explained both paramagnetic–ferromagnetic and ferromagnetic–re-entrant transitions successfully, local correlations are not compatible with it since its infinite-range character does not allow for any difference between local and non-local spin coupling. The opposite approximation, i.e. coupling with the nearest neighbours only, may be closer to reality. Unfortunately, using this approximation Thomson *et al* [16] have not given any estimates of an Fe correlation parameter. Up to now the correlation effect has been estimated in models using lower dimensions only. The older numerical simulations [27], which dealt with the 2D nearest-neighbour Heisenberg spins having both antiferromagnetic and ferromagnetic coupling, have shown that the ground state is of re-entrant type and the correlation function has been calculated, too. A rough estimation gives a correlation length of ~2 lattice spacings, which could ensure a significant correlation in the nearest-neighbour region. However, similar calculations using the 2D XY spins [28] were later criticized by Thomson *et al* [16] because some states were found to be metastable when the calculations were extended to higher dimensions. Therefore it is not quite clear whether the theory using the lower dimension could yield a good description of correlations.

The most realistic theoretical approach (3D Heisenberg spins with long-range RKKY coupling) has given promising results [17], but the detailed description of the ground state of the Fe spins in Au including an estimation of local correlations is, again, not available yet. Further theoretical and experimental effort is needed to clear the matter up completely.

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