Magnetic field dependence of the magnetization of a 29 nm thick AuFe spin glass film

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Abstract. We performed polarized neutron reflectometry (PNR) experiments on a 29 nm thick Au$_{93}$Fe$_7$ film. The measurements were done in a temperature range from 300 K to 2 K in a magnetic field up to 6 T applied in the sample's plane, at 2 K we took PNR data up to 11 T. The magnetization as determined by PNR can be described with a Brillouin function from 295 K down to 50 K assuming the magnetic moment of isolated Fe atoms, i.e. 4 $\mu_B$ per Fe atom. Below 50 K the onset of the spin-glass freezing was observed as a strong reduction of the magnetic moment compared to the isolated Fe atom.

1. Introduction
The magnetic and transport properties of non-magnetic metals with magnetic impurities have been of large scientific interest for the last five decades, both for theorists and experimentalists [1]. The prerequisite for a spin glass is a competition among the different interactions between the spins with a random (glass-like) distribution of these interactions. The random spin arrangement in a spin glass is frozen when cooling the system below a certain temperature $T_f$, called the freezing temperature. It is still under discussion whether the oscillating Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction is the only origin for this freezing or frustration. Khmelevskyi et al. [2] pointed out that the long-range RKKY interaction is not a prerequisite for a spin-glass system. They conclude that only very dilute systems can be described by the RKKY interaction properly, whereas for higher concentrations the magnetic properties in AuFe spin glass systems are dominated by short-range interactions.

Depending on the concentration of the magnetic solute the AuFe system shows a very peculiar behaviour as a function of temperature. A paramagnetic to spin glass state is observed in the concentration range from 0.005 % to 10 at.% Fe [3], a paramagnetic to ferromagnetic phase transition for an Fe concentration above 10 at.% [3] and a reentrant spin-glass behavior in the range from 10 to 24 at.% [4]. An overview of bulk AuFe properties can be found in Cannella et al. [3].
Thin alloy film properties have been studied mainly by means of resistivity and Hall effect measurements which are among the few techniques applicable to single layer spin glasses. From the experimental point of view it is very important to measure directly the magnetic moment of the impurity, which is extremely difficult to obtain from the magnetometery techniques for layer thicknesses in the nm range. Indeed, that has led to many investigations of multilayered samples consisting of spin glass layers alternated with decoupling layers, in order to increase the magnetization signal [5]. PNR has turned out to be an excellent tool to determine the absolute magnetic moment of single spin glass films avoiding the complications of multilayer samples such as inhomogeneity and an incomplete decoupling between the individual spin glass layers. In earlier PNR studies on Au$_{93}$Fe$_7$ [6, 7] and Au$_{97}$Fe$_3$ films [8, 9] we determined the temperature dependence of the absolute magnetic moment in a temperature range from 295 down to 2 K in a field of 6 T. Only recently we were able to show that the temperature dependence of the magnetization in AuFe films shows a size effect. The spin glass magnetization deviates from bulk behaviour below 10 nm but it still shows a typical spin-glass behaviour [9]. The present study is an extension of the previous work by focusing on the magnetic-field dependence of the spin glass film magnetization.

2. Experimental

The 29 nm thick AuFe alloy film was prepared by co-deposition of Au and Fe onto thermally oxidized silicon wafers by molecular beam epitaxy. The PNR experiments were performed on the C5 spectrometer at the neutron research reactor NRU in Chalk River, Canada and the instrument Asterix at LANSCE, Los Alamos, USA.

On C5 we used a Cu$_2$MnAl Heusler crystal at $\lambda = 0.237$ nm as monochromator and analyzer along with a PG filter to reduce the higher order contamination ($\lambda/2$, $\lambda/3$). The magnetic field was provided by a cryomagnet [10] with the magnetic field being in the sample plane, which coincides with the polarization direction of the incident neutron beam and perpendicular to the scattering plane. The superconducting split-pair magnet provides a maximum field of 7.2 T in the asymmetric mode where the bottom coil is energized with a higher current, which shifts the null point (location of zero stray field) above the neutron beam. In a field of 6 T with this setup [10] we achieved a 96 % polarization of the incoming neutron beam, as determined from the measured flipping ratio of 25:1. Prior to the PNR measurements, the sample was cooled down in a field of 6 T.

Since the Asterix instrument is located at a pulsed neutron source, neutron wavelength was measured using time-of-flight techniques. Neutrons with wavelengths varying from 4 to 12 Å were provided by a l-H$_2$ cold moderator [11]. Magnetic fields up to 11 T were produced by an Oxford cryomagnet operating in symmetric mode (the field null for the cryomagnet was compensated using permanent magnets). With a field of 11 T applied in the sample plane and perpendicular to the scattering plane, the polarization of the incident neutron beam changed from 93 to 96% depending upon neutron wavelength [12]. The sample was field cooled from 300 K to 2 K in a field of 6 T, then measurements of the reflectivity using polarized neutron beams were taken at 2 K in fields from 6 to 11 T.

3. Results

In neutron reflectometry the interaction of neutrons with a film, assuming the sample’s magnetization is parallel to the external field, can be described by the Fermi pseudopotential $V^\pm$:

$$V^\pm = \frac{2\pi \hbar^2}{m} N(b_{\text{nuc}} \pm b_{\text{mag}}),$$

where $m$ denotes the neutron mass, $N$ the atomic density, $b_{\text{nuc}}$ the nuclear scattering length, and $b_{\text{mag}}$ the magnetic scattering length. The superscripts + and - indicate that the scattering potential is different for neutrons with their spins aligned parallel (+, up-neutrons) or antiparallel (-, down-
neutrons) to the external field. Therefore, the measured reflectivity curve $R^+$ for spin-up neutrons is different from the reflectivity curve $R^-$ for spin-down neutrons, which is then used to determine the magnetization of the sample [13,14,15] because the difference between $R^+$ and $R^-$ is proportional to the sample’s magnetization. The magnetic scattering length is directly proportional to the magnetization of the sample with $b_{\text{mag}} = c \mu$, where the conversion constant is $c = 2.695 \text{ fm} / \mu_B$ with $\mu_B$ the Bohr magneton and $\mu$ the magnetic moment per atom. In the specular reflection geometry the scattering vector $\mathbf{q} = \mathbf{k}_f - \mathbf{k}_i$ is perpendicular to the sample’s surface with $k_f = k_i = 2 \pi / \lambda$, $k_f$ and $k_i$ being the wave vector of the reflected and incoming beam, respectively. Therefore, perpendicular magnetization components do not contribute to the potential $V^\pm$. The calculation of the neutron reflectivity is analogous to the calculation of the Fresnel reflectivity in light optics. We used the software Parratt32 [16] which is based on the recursion formula developed by Parratt [17].

Fig. 1 shows the polarized neutron reflectivities $R^+$ and $R^-$ as a function of the wave vector transfer $q$ (i.e. the difference between the incoming and outgoing neutron wave vectors) for specular scattering measured at 10 K in magnetic fields of a) 6 T and b) 0.2 T for the 29 nm thick Au$_{93}$Fe$_7$ film. The solid and dashed lines are the fits to $R^+$ and $R^-$, respectively. From the fits we can deduce a magnetic scattering length density $N_{\tilde{b}_{\text{mag}}} = 0.11 \times 10^{-6} \frac{\text{Å}^{-2}}{\text{Å}^{-2}}$ in a field of 6 T and $N_{\tilde{b}_{\text{mag}}} = 0.02 \times 10^{-6} \frac{\text{Å}^{-2}}{\text{Å}^{-2}}$ in a field of 0.2 T. This corresponds to a magnetic moment of 0.99 $\mu_B$ per Fe atom and 0.22 $\mu_B$ per Fe atom, respectively.

In a dilute AuFe alloy above $T_f$ the Fe impurities do not interact and should behave as paramagnetic moments which are proportional to the magnetic moment of the impurity times the Brillouin function $\tilde{B}(J, \mu B / k_B T)$ with the total angular momentum $J$, the magnetic moment $\mu$, the external magnetic field $B$, the Boltzmann constant $k_B$, and the temperature $T$. Earlier PNR
measurements on the same sample [6] showed that the Fe atoms are partially dispersed homogeneously into the Au matrix and partially aggregated as clusters with a typical size of about 1 nm. The average magnetic moment $\bar{\mu}$ of the AuFe film as determined by PNR can be described as [6]

$$\bar{\mu}(B) = f\mu_{cl}(B) + (1-f)\mu_{is}(B)$$  \hspace{1cm} (2)

with

$$\mu_{cl} = g\mu_B J_{cl} \tilde{B}(n_{cl} J_{cl}, \mu_B / k_B T)$$  \hspace{1cm} (3)

and

$$\mu_{is} = g\mu_B J_{is} \tilde{B}(J_{is}, \mu_B / k_B T)$$  \hspace{1cm} (4),

where a fraction of $f = 7\pm 1\%$ of the total amount of Fe atoms is aggregated in clusters with an averaged angular momentum of $J_{cl}$ per Fe atom and $n_{cl} = 60$ Fe atoms per cluster. There are $(1-f) = 93\%$ isolated Fe atoms with a magnetic saturation moment of $\mu_{is} = gJ_{is}\mu_B$, where $g$ denotes the gyromagnetic ratio.

Fig. 2: averaged magnetic moment per Fe atom versus applied magnetic field as derived from the fit to the PNR data at a) 10 K and b) 280 K. The dotted line represents the simulation for the clustered Fe atoms only, the dashed lines are the simulations for the sum of clustered and isolated Fe atoms with $J_{is} = 2$, the solid line in a) corresponds to the sum of clustered and isolated Fe atoms with $J_{is} = 0.95$.

The field dependence of the magnetization of the Au$_{93}$Fe$_7$ film is shown in Fig. 2 for 10 and 280 K. The data points represent the average magnetic moment $\bar{\mu}$ as derived from fits to the reflectivity curves. This field dependence can be described in both cases by the field dependence of the Brillouin function. The dotted line represents the contribution of only the Fe atoms in clusters. The dashed line is the averaged magnetic moment calculated according to Eq. 2 using $J_{is} = 2$ corresponding to $4\mu_B$ per Fe atom for non-interacting isolated Fe atoms. The dashed line describes the data at 280 K reasonably.
well. A fit of $J_{is}$ does not make sense because the experimental errors are larger than the expected contribution of the isolated Fe atoms. At 10 K the contribution of the isolated Fe atoms is much larger and we can conclude that our experimental data can no longer be fitted with a Brillouin function using $J_{is} = 2$ (dashed line in Fig. 2a). A Brillouin function with $J_{is} = 0.95$ (solid line in Fig. 2a), however, can fit the data well.

![Graph showing magnetic moment vs. applied field at 2 K](image)

**Fig. 3:** Averaged magnetic moment per Fe atom at 2 K as a function of applied magnetic field. The dotted line represents the simulation for the clustered Fe atoms only, the solid and dashed lines are the simulations for the sum of clustered and isolated Fe atoms with $J_{is} = 0.35$ and $J_{is} = 2$, respectively.

In order to get more insight into the magnetic-field dependence of the spin glass magnetization we measured at 2 K in applied magnetic fields up to 11 T. The measurements from 0.1 T to 6 T were performed in Chalk River, the measurements from 6 T to 11 T were performed at Los Alamos National Laboratory. The magnetic moment inferred at 6 T from the two instruments is the same within errors. The data of both instruments are displayed in Fig. 3. Again, the field dependence can be described with the field dependence of the Brillouin function. The dotted line represents the contribution of the Fe atoms in clusters, the dashed line represents the averaged magnetic moment using $J_{is} = 2$, the solid line is the fit with $J_{is} = 0.35$. Because of the large energy associated with the frustration in the AuFe spin-glass system, the applied field of 11 T is not enough to saturate the magnetization well below the freezing temperature. Measurements on bulk AuFe samples in pulsed high fields showed that even a magnetic field of 40 T is not high enough to saturate the magnetization at 4 K [18].

The fraction of clustered Fe atoms as deduced earlier [6] is $7 \pm 1\%$. The absolute error of $\pm 1\%$ induces an error in $J_{is}$ of $\pm 0.03$. An increase in the averaged magnetic moment due to an increase in clustered Fe atoms would have to be compensated with a decrease of the magnetization of the isolated Fe atoms and vice versa.

4. **Discussion**

At 280 K, far above $T_s$, the magnetic-field dependence can be well described with the Brillouin function using the magnetic moment of a free Fe atom. This proves that the Au$_{93}$Fe$_{7}$ film behaves like
a paramagnet above the freezing temperature. The existence of the magnetic frustration in the spin-glass state can be concluded from the fact that the magnetic-field dependence can no longer be described with the Brillouin function using $J_{s} = 2$ of a free atom. At 10 K the data can be fitted with $J_{s} = 0.95$, at 2 K this value is further reduced to $J_{s} = 0.35$. This nicely demonstrates that the magnetic frustration is increasing with decreasing temperature. This was also observed in the temperature dependence of the identical Au$_{93}$Fe$_{7}$ film [7, 8]. The magnetic moment as determined with PNR in a field of 6 T can be fitted with a Brillouin function using $J_{s} = 2$ down to 50 K, i.e. well above $T_{f}$. The onset of spin-glass behaviour can be clearly observed below 50 K, where the magnetization deviates from the Brillouin function [7]. Within the errors the magnetization stays constant with a value of about 1 $\mu_{B}$ per Fe atom. This deviation from the Brillouin function at temperatures below 50 K has also been observed in Au$_{97}$Fe$_{3}$ films [8, 9].

5. Conclusion
We successfully performed PNR measurements in large magnetic fields on a 29 nm thick Au$_{93}$Fe$_{7}$ film. The sample shows paramagnetic behaviour of isolated Fe atoms at 280 K that can be described with a Brillouin function using $J = 2$ or the magnetic moment of a free Fe atom of 4 $\mu_{B}$, respectively. The spin glass behaviour can be clearly observed at 10 K and 2 K, where the magnetic-field dependence of the magnetization can only be fitted with reduced $J$-values of $J_{s} = 0.95$, and $J_{s} = 0.35$, respectively.

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