Frequency dependence of the superparamagnetic transition in a Finemet-type nanocrystalline alloy

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The transition to superparamagnetism in a Cr-containing Finemet alloy has been studied by means of ac susceptibility and muon spin relaxation experiments. The influence of bias field and measuring frequency has been analyzed. The transition temperature is controlled by the interaction between the particles. These results are consistent with previous static magnetic measurements.

1 Introduction

The microstructure of nanocrystalline Finemet-type alloys consists of Fe–Si grains with a mean size of about 10 nm, embedded in an amorphous matrix. The Curie transition of the matrix can be controlled by the partial substitution of Fe by refractory elements, like Cr. Apart from their technological importance, these alloys are adequate for studying the magnetic behaviour of a multiphase granular magnetic system.

Recently, the superparamagnetic transition of one of these alloys has been modelled [1], showing the importance of dipolar interactions between the particles. In this work, the superparamagnetic transition will be studied with the help of ac techniques and µSR.

2 Experimental

Amorphous ribbon 1 cm wide and 25 µm thick of nominal composition Fe₆₃.₅Cr₁₀Nb₁₂Si₁₃.₅B₉Cu₁ was prepared by single roller melt spinning. The devitrification process was studied by Perkin–Elmer DSC-7 differential scanning calorimeter. The samples were annealed in halogen lamp furnace under argon stream and the crystalline volume fraction of the samples was calculated by fitting the profile of the main X-ray diffraction peak by the superposition of two pseudo-Voight functions. The field, frequency and temperature dependences of the complex susceptibility were measured by SQUID magnetometer (Quantum Design MPMS-5S) from 10 K up to 700 K, with an excitation field of 1 Oe. Zero and longitudinal field muon spin relaxation (µSR) experiments were performed on the EMU spectrometer at the ISIS pulsed neutron and muon facility at the Rutherford Appleton Laboratory, UK. Measurements were made...
at temperatures ranging from 300 to 700 K. Samples were mounted on a Ti sample holder. The spectra were collected up to 10 million events per run and were analyzed over the time range \( t < 5 \mu s \) using the WiMDA computer program [2]. Two nanocrystalline samples with \( \sim 16\% \) and \( \sim 20\% \) crystalline fractions (annealed for 1h at 815 and 825 K, respectively) were studied.

3 Results and discussion

3.1 Susceptibility

The typical behaviour of this alloy, from the point of view of ac susceptibility, is represented in Fig. 1. At temperatures below the Curie temperature of the amorphous matrix, \( T_{C\text{am}} \), the system behaves as a soft magnetic material. As the temperature is increased, there is a frequency independent decrease in susceptibility due to the paramagnetic transition of the matrix, which prevents the exchange coupling between the nanocrystals. However, as there is still a dipolar coupling between the particles [1], the superparamagnetic transition cannot be found until the interactions are reduced further, as evidenced by the frequency dependent susceptibility maximum in the high temperature limit.

Fig. 1 (online color at: www.pss-a.com) Temperature dependence of the real part of the susceptibility of the sample with 16% crystalline fraction (annealed at 815 K/1h), measured with different excitation frequencies and a bias dc field of 100 Oe.

Fig. 2 (online color at: www.pss-a.com) Influence of the applied bias field (left: 0 Oe; right: 100 Oe) on the temperature dependence of the complex susceptibility of the sample with 16% crystalline fraction (815 K/1h), measured with different excitation frequencies.
The effect of a bias dc field is to displace the susceptibility maximum to lower temperatures, i.e. the opposite direction of the frequency effect (Fig. 2). Therefore, to be able to detect the susceptibility maximum for all the used frequencies, a field of 100 Oe was necessary, as the measuring temperature could not be increased further to avoid any change to the microstructure of the samples during measurements.

Figure 3 shows the main differences in the susceptibility of the samples with ~16% and ~20% crystalline volume fractions (annealed at 815 and 825 K for 1h), respectively. As the crystalline fraction increases, a slight increase in susceptibility and a displacement to lower temperatures of the susceptibility decrease associated to $T_{C(\text{am})}$ are found. However, these two samples show no significant displacement of the superparamagnetic peak. Although the reduction in $T_{C(\text{am})}$ facilitates the decoupling between the particles, a higher crystalline fraction compensates this effect [1].

### 3.2 Muon spin relaxation

An overview of the technique and its capabilities is given in Ref. [3]. The measured spectra were fitted to a phenomenological exponentially decaying function, with the addition of a constant term considered as a baseline:

$$P(t) = a_t \exp(-\lambda t) + a_0.$$

Figure 4 shows some typical spectra, together with the fits to the data, while Fig. 5 represents the relevant parameters of the fittings.
Due to the pulsed character of ISIS, the muon spectrometers cannot measure fast relaxation rates (>10 MHz) which prevents the detection of the depolarization of muons implanted in the sample in an environment where a very strong static field exists. Therefore, superparamagnetism can be detected as a maximum in the relaxation rate ($\lambda$) when the transition temperature is approached from below, and an increase in the amplitude associated to the exponential term [4]. For lower temperatures, muons implanted in a ferromagnetic environment fluctuate too fast for the spectrometer, contributing to the base line asymmetry. The effect of a field is to slow down the fluctuation rates. On the one hand, this displaces the increase of $a_r$ to lower temperatures, but on the other hand, $\lambda$ is progressively reduced.

As the characteristic measuring time of the muon is of the order $10^{-8} - 10^{-6}$ secs, susceptibility data can explain why the whole $\lambda$ maximum is not observed in the muon spectra: the transition temperature is too high for this characteristic time. It has also been reported that bias field is less effective in displacing the transition temperature in the case of $\mu$SR [5], which explains that the transition cannot be completed even for 4500 Oe. Muon experiments (in agreement with the ac susceptibility data) do not hint at a relevant displacement of the transition temperature with the increase of the crystalline fraction as Fig. 6 shows.

### 5 Conclusions

The study of the superparamagnetic transition of a Cr-containing Finemet alloy by means of ac susceptibility and muon spin relaxation confirms the interpretation given for the static magnetization measurements. The transition temperature is controlled by dipolar interactions between the particles, and not by their intrinsic blocking temperature.
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