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LETTER TO THE EDITOR

Broad peak of the non-linear susceptibility at the spin-glass temperature of amorphous $Fe_{93}Zr_7$

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Abstract. The broad peak of the non-linear AC susceptibility around the spin-glass temperature (T_g) of amorphous $Fe_{93}Zr_7$ suggests that the paramagnetic to spin-glass transition at T_g is determined by the superparamagnetic behaviour of magnetic clusters.

Amorphous $Fe_{100-x}Zr_x$ (x = 7–12) alloys show peculiar magnetic properties at low temperatures in low magnetic fields, including spin-glass (x = 7), re-entrant (x = 8–10) and ferromagnetic-like behaviour (x = 12) [1]. The alloy near the critical concentration (x = 7) looks like a typical spin glass as seen from the temperature dependence of the susceptibility. It is a cusp-like function around a temperature called the spin-glass temperature (T_g) after cooling the sample through T_g in zero field. The width of the peak is rather broad (about 20–30 K) and it is only slightly dependent on the measuring frequency.

In usual ferromagnets the linear AC susceptibility χ_0 exhibits a divergent behaviour at the Curie temperature (T_c). However, in real measurements the transition is smeared out because of the non-vanishing measuring field and due to the magnetic disorder in systems like crystalline AuFe and amorphous Fe–Zr alloys. It should be noted that the non-linear AC susceptibility χ_2 (defined from the expansion of the magnetization M with respect to the applied field H in the vicinity of T_C: $M = \chi_0 H + \chi_2 H^3 + \chi_4 H^5 + \ldots$ represents the critical behaviour more clearly than χ_0 [2, 3]. χ_2 diverges for ferromagnetic phase transitions and changes its sign across T_C: it is positive below T_C and negative above T_C. For spin-glass transitions a negative divergence of χ_2 both below and above T_g is predicted. The non-linear AC susceptibility χ_2 for the classical spin glass $Au_{96}Fe_4$ has been shown to behave accordingly: it has a very sharp negative peak at T_g [4]. This transition was compared to that of ferromagnetic fine cobalt particles precipitated in a $Cu_{97}Co_3$ alloy [4]. In the latter system χ_0 also shows a spin-glass-like maximum at low temperature while χ_2 is negative with a peak around the temperature where χ_0 exhibits the maximum. However, the peak in χ_2 is very broad compared with that of $Au_{96}Fe_4$. THis behaviour can be well described by the blocking of superparamagnetic particles.

Ma *et al* [5] investigated the Curie and spin-glass transitions of the re-entrant $Fe_{100-x}Zr_x$ glasses (x = 8–11) measuring both the linear and the non-linear susceptibilities. The peaks in the non-linear susceptibility at the spin-glass temperature were rather broad (the

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width is about 30–40 K), showing no divergent behaviour. Despite this observation the authors suggest this transition to be a real phase transition and explain the non-divergent feature of the non-linear susceptibility by the dynamic nature of the measurements and the complications associated with transverse spin freezing thought to be present in re-entrant amorphous Fe-Zr alloys.

In this letter the linear (χ_0) and non-linear (χ_2) AC susceptibilities of amorphous $Fe_{93}Zr_7$ were measured at low ($f = 1$ kHz) and high ($f = 30$ MHz) frequencies to clarify the nature of the paramagnetic to spin-glass transition at T_g. An amorphous ribbon of the nominal composition $Fe_{93}Zr_7$ was prepared by melt-spinning. Static (DC) susceptibility measurements reveal a paramagnetic to spin-glass transition at $T_g \cong 105$ K (figure 1) in good agreement with literature data [1].

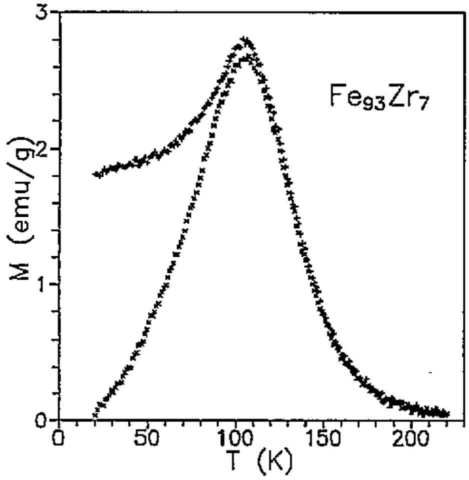

Figure 1. DC magnetization M (in emu g^{-1}) as a function of temperature T (in K) for amorphous $Fe_{93}Zr_7$, measured in external magnetic field H_{ext} = 10 Oe after cooling the sample in zero field (×) and in the measuring field (+).

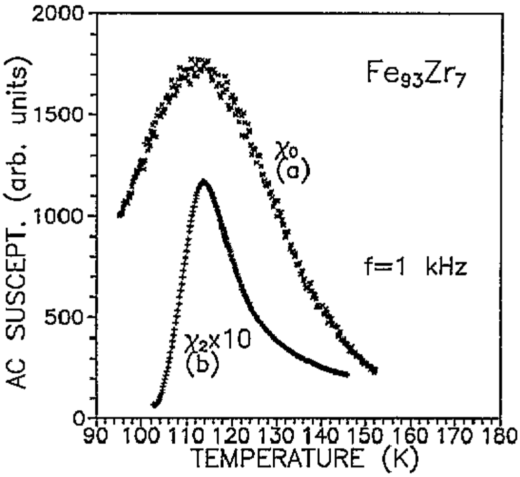

Figure 2. Linear χ_0 (a) and non-linear χ_2 (b) AC susceptibility as a function of temperature T (in K) at f = 1 kHz for amorphous $Fe_{93}Zr_7$ alloy. The excitation field is H_{exc} = 10 Oe (1 mT).

The AC susceptibility was measured in the radio-frequency (RF) range at $f = 30$ MHz with the help of equipment which consists of three main parts: (i) an excitation and pick-up coil working in a usual inductive transmission mode; (ii) a home-made wide-band RF lock-in of 50 Ω input impedance which amplifies the voltage detected by the low-impedance pick-up coil wound around the sample and measures its two components by two phase-sensitive detectors (PSDs); (iii) a commercial crystal-controlled RF generator of 50 Ω impedance which supplies the RF drive to the excitation coil (H_{exc} = 1 mOe or 0.1 μT) and the reference signals to the PSDs.

The reference signals are connected to the reference inputs of the two PSDs through two different delay lines whose lengths are selected in such a way that the phase difference between them at the reference inputs is $\pi(n + \frac{1}{2})$ at 30 + n60 MHz where n is an integer. The PSD outputs are amplified and filtered by low-frequency amplifiers with an upper cut-off frequency of 150 kHz. In the case of the linear susceptibility (χ_0) the two components of the complex susceptibility with respect to a random oriented coordinate system were measured at the PSDs' outputs (as DC signals). The phase angle of the RF magnetic moment of the sample relative to the excitation field was determined by the signal of the empty pick-up coil. In this way we can calculate the exact real and imaginary part and also the amplitude (absolute value) of χ_0 (the latter is used here). In the measurements the excitation signal was chopped to separate the sample response.

The non-linear susceptibility (χ_2) measurements were performed in a different way. We used two RF generators of the same type, offset from each other in frequency. While one of them provided the reference signal with a frequency of $3f + 9$ kHz for the RF lock-in, the other supplied a continuous signal of frequency f and controlled amplitude which was connected to the excitation coil. The non-linear response of the sample is given by one of the (equivalent) 9 kHz output signals of the PSDs which was amplified in AC mode and measured in AC RMS mode. The exact amplitude of χ_2 was calculated by subtracting the background of the empty pick-up coil.

The measurements in the low-frequency range at $f = 1$ kHz were made in a similar way. The only exception is the use of a commercial lock-in amplifier instead of the home-made one. Because of the diminished sensitivity at low frequencies the excitation magnetic field had to be increased ($H_{exc} = 10$ Oe or 1 mT).

Figures 2(a) and 3(a) show the linear susceptibility χ_0 of the sample as a function of temperature at 1 kHz and 30 MHz, respectively, in the latter case normalized with respect to the signal of the empty pick-up coil. The broad peaks indicate that the paramagnetic to spin-glass transition occurs at $T_g \cong 113$ and 125 K at 1 kHz and 30 MHz, respectively, reflecting its frequency dependence. The width of the peak of χ_0 is 35–40 K at 1 kHz and 20–25 K at 30 MHz. The temperature dependence of the non-linear susceptibility χ_2 for the sample is shown in figures 2(b) and 3(b) at 1 kHz and 30 MHz, respectively, in the latter case after having subtracted the background of the empty pick-up coil. Broad peaks appear again at both frequencies at a somewhat (1–2 K) higher temperature than T_g. At 1 kHz the peak of χ_2 is narrower (with a width of 15 K) than that of χ_0 while at 30 MHz it has about the same width (20–25 K) as that of χ_0. Since the skin penetration depth in the RF range and the corresponding sample size (the ribbon is about 10 μm thick) are comparable, diamagnetic background shielding can be observed in the temperature dependence of both χ_0 and χ_2 at 30 MHz (figure 3). As mentioned above, similar broad peaks were found both in χ_0 and χ_2 by Ma *et al* [5] in the re-entrant amorphous $Fe_{100-x}Zr_x$ ($x = 8, 9$) alloys.

The AC non-linear susceptibility χ_2 reflects properties concerning the static phase transition (such as a divergence of the static non-linear susceptibility) only in the $f \to 0$ limit [6]. The measurements of Bitoh *et al* [4] were made at 80 Hz which is close to our lower frequency ($f = 1$ kHz). Thus χ_2 measured at 80 Hz seems to reflect the divergence of the static non-linear susceptibility at T_g in $Au_{96}Fe_4$. Estimating from the frequency dependence, the width of χ_2 in $Fe_{93}Zr_7$ would not be less than 10 K even at 80 Hz. Therefore, the peak of about 15 K width in χ_2 of $Fe_{93}Zr_7$ at 1 kHz (figure 2(b)) is expected to show the non-divergent character of the static non-linear susceptibility.

The experimental facts shown in figures 2 and 3, keeping in mind the results of Bitoh *et al* [4] mentioned above, strongly support the idea that the spin-glass-like properties of amorphous $Fe_{93}Zr_7$ are determined by the blocking of superparamagnetic clusters. This explanation was first suggested for classical spin glasses by Wohlfarth [7]. There is also evidence for cluster-glass behaviour from Mössbauer data near T_g of the same $Fe_{93}Zr_7$ ribbon and near T_c of the re-entrant amorphous Fe–Zr alloys, observing a large hyperfine field increase on introducing a small external magnetic field [8]. The bulk magnetization against magnetic field of the same alloys can also be described by interacting magnetic clusters [9]. The non-divergent behaviour of χ_2 at T_g in the re-entrant amorphous Fe–Zr alloys was interpreted [5] by the presence of a ferromagnetic component. According to

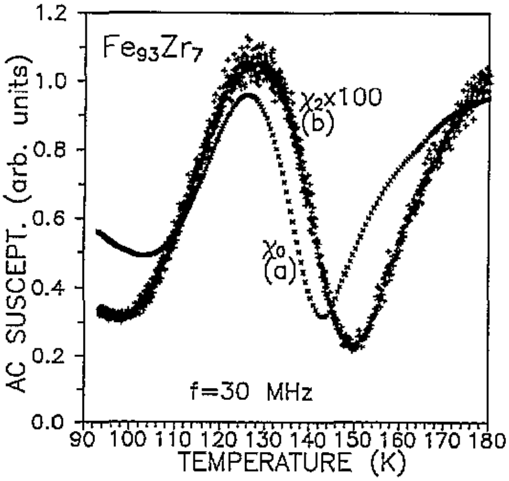

Figure 3. Linear χ_0 (a) and non-linear χ_2 (b) AC susceptibility as a function of temperature T (in K) at $f = 30$ MHz for amorphous $Fe_{93}Zr_7$ alloy. The excitation field is $H_{exc} = 1$ mOe (0.1 μT). χ_0 is normalized with respect to the signal of the empty pick-up coil while χ_2 is given after having subtracted the background of the empty pick-up coil.

our measurements (figures 2(b) and 3(b)) χ_2 is also non-divergent at T_g in the spin-glass-like amorphous $Fe_{93}Zr_7$ though no ferromagnetic component is present. Therefore, since there is a continuous change in the magnetic properties of amorphous Fe–Zr alloys, the features of the non-uniform magnetic structure (clusters) are expected to determine also the ferromagnetic to spin-glass transition of the re-entrant alloys.

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