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Relaxation in the $Co_{50+x}Al_{50-x}$ system

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Abstract. Results of ageing experiments on the $Co_{50+x}Al_{50-x}$ alloy system are reported. All alloys display a maximum in the temperature dependence of the maximal rate of the relaxation, $S_{max}(T)$. The temperature of this maximum, T_{max}, is $\approx 0.60\ T_m$ (T_m is the freezing temperature) for the investigated spin-glass and cluster-glass materials. T_{max} depends on the concentration of the dopant, and may depend on the waiting time used in the experiment.

1. Introduction

$Co_{50+x}Al_{50-x}$ alloy is an example of a random-exchange magnetic material (REMM). Through increase of the dopant concentration the system gradually sweeps through the spin-glass (SG), cluster-glass (CG) and re-entrant region (RE) of the magnetic phase diagram (MPD) [1, 2]. Each sample type displays specific behaviour when appropriate experiments are performed.

It is known that in a random-exchange magnetic material the exchange couplings are positive and random in magnitude.

This investigation is to examine the ageing and relaxation behaviour [3] of the random-exchange $Co_{50+x}Al_{50-x}$ spin-glasses. Using a long-observation-time experiment to investigate an alloy system creates a picture of the gradual development of the relaxation process through doping.

Problems such as the dependence of the maximal speed of relaxation S_{max} on the temperature of the investigation T, the dependence of $S_{max}(T)$ on the waiting time used in the experiment, t_w, and the influence of the concentration of the dopant x on $S_{max}(T)$ are discussed.

The development of the relaxation process in the low-temperature SG–RE region through proceeding to the 'high'-temperature soft-ferromagnetic (FM) RE phase is traced.

2. Experiment

Polycrystalline $Co_{50+x}Al_{50-x}$ samples ($x = 2.5$, 5, 12 and 13) [1, 2], prepared by arc-melting, are used for magnetic measurements.

Structural x-ray diffraction, reported in [1], showed the CsCl single-phase crystal structure in $Co_{50+x}Al_{50-x}$, for the investigated concentration range.

The magnetic measurements were made with a commercial Quantum Design (San Diego) SQUID magnetometer with sensitivity of about 10^{-6} emu, temperature measuring interval from 1 to 800 K and applicable magnetic field interval $(-5, 5)$ T.

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To investigate the ageing and relaxation in a material [3, 4], the sample was cooled down in zero field from a temperature well above the freezing temperature T_m to the measuring temperature T and aged (kept) at T for a waiting time t_{wait}, before a probing magnetic field in the linear region of the magnetization M–H was switched on. After a period of about 60 s, the magnetic field was stable. Then the measuring procedure of the zero-field cooled magnetization, M_{ZFC}, against time, t, for a period of $\sim 5 \times 10^3$ s, started. Time $t = 0$ is defined as the moment when the magnetic field becomes saturated in the non-overshoot mode. The resultant waiting time, t_w, is defined as the sum of the times of keeping the sample at a constant temperature t_{wait}, and the time to obtain stable magnetic field.

The SG freezing temperature T_m is determined as the temperature of the maximum of the dc M_{ZFC}–T curve, measured using a small dc magnetic field of 1 Oe. The rate of warming during the dc magnetization measurement was usually 0.0018 K s^{-1}. No relaxation was observed at and above T_m.

3. Results and discussion

The spin-glasses were investigated at a set of temperatures below their freezing temperature T_m. After a waiting time of ∼5, 15, 30 or 50 min a weak probing magnetic field of ∼1 Oe was applied to study the behaviour of the SG at the different temperatures. The M_{ZFC}–t plots are typical, and do not expose any specific peculiarities for all investigated alloys, though ascribed to different regions of the MPD [1].

3.1. The $Co_{52.5}Al_{47.5}$ spin-glass

$Co_{52.5}Al_{47.5}$ has a freezing temperature at ∼13.5 K [1]. The spin-glass was investigated for the waiting times of ∼8, 13, 22, 33 and 53 min.

A typical development of the magnetization with time at temperatures below T_m, for the $Co_{52.5}Al_{47.5}$ SG, is shown in figure 1(a). The plots were taken after a waiting time of ∼8 min. Figure 1(b) shows the gradual change of the relaxation rate $S = \mathrm{d}M/\mathrm{d}\ln(t)$ against $\ln(t)$ [3, 4], with the temperature of the investigation. It can be observed that the maximal value of this rate, $S_{max}(T)$, changes with the temperature of the investigation, too (see figure 2). $S_{max}(T)$ can be descriptive, in a way, of the system's response to an external influence such as ageing. The maximal rate of relaxation for $Co_{52.5}Al_{47.5}$ is highest at the temperature of ≈8.05 K, which is 0.60 of T_m, for the waiting time of ∼8 min.

The set of the maximal rates of change of the magnetization against $\ln(t)$, measured at different temperatures, fits well to a Gaussian distribution (figure 2), and, consequently, the relaxation process is best observed at $T \approx$ the temperature of the maximum of the Gaussian curve. Further, by T_{max} we denote the temperature at which a set of $S_{max}(T)$ values has a maximum.

If the measurements are performed in the linear region of M against H, and express the relaxation process in the system correctly [5], then the position of the maximum in $S_{max}(T)$ should not depend on the magnitude of the applied field. However, the growth of the domains in the SG is dependent on the waiting time, and hence so is the relaxation process.

Figure 3 presents the temperature dependences of $S_{max}(T)$ for the waiting times of 8, 13, 22, 33 and 53 min for $Co_{52.5}Al_{47.5}$. It is seen that the position of $S_{max}(T)$ is practically constant within the experimental error, as the waiting time increases. Physically this means that the rate at which M against $\ln(t)$ is greatest stays the same as the system is aged longer. This implies a quasi-stable picture of the domains in this sample, reached already at ∼8 min.

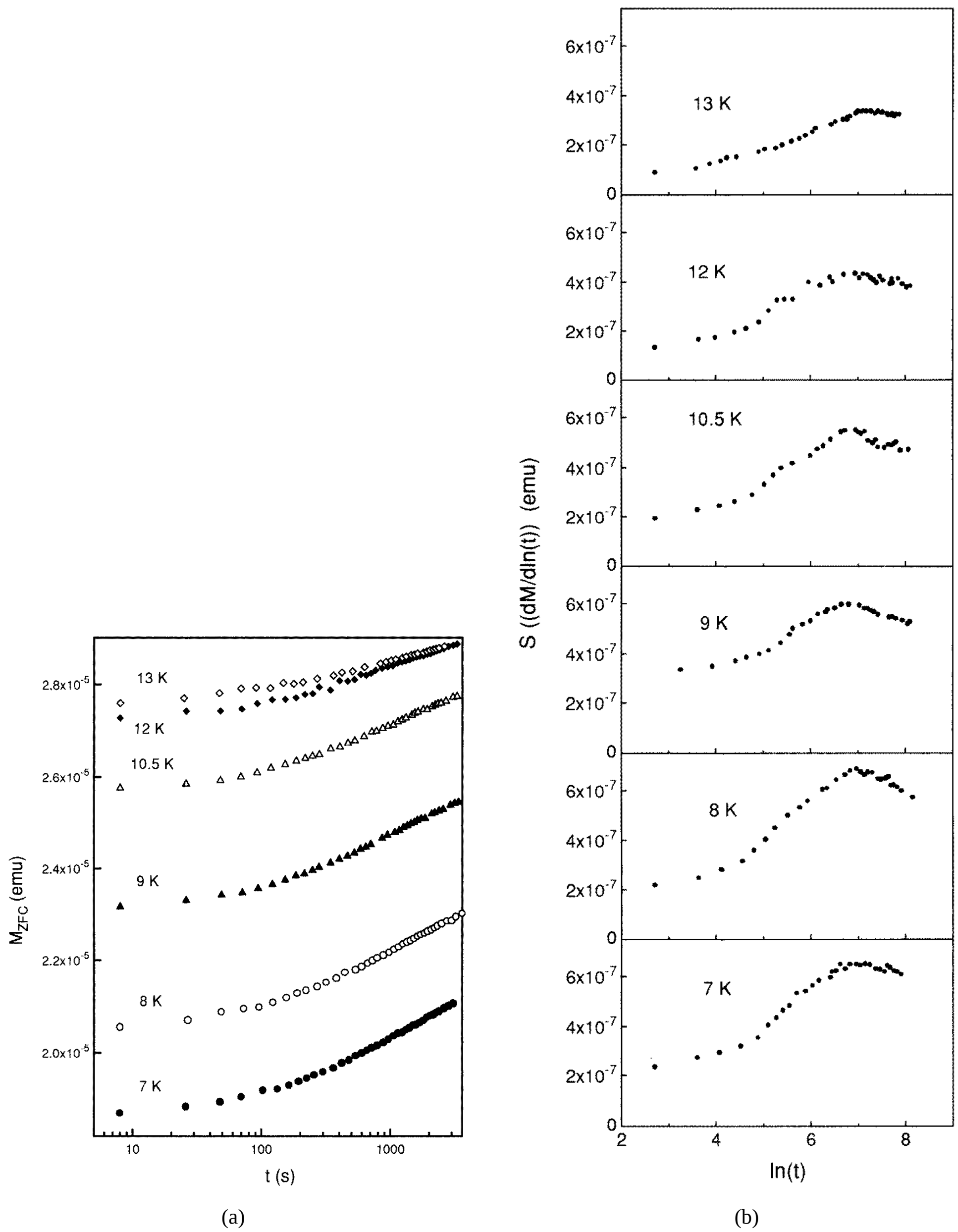

Figure 1. (a) Relaxation of the M_{ZFC} isotherms at the temperatures of 7, 8, 9, 10.5, 12 and 13 K for $Co_{52.5}Al_{47.5}$, measured after $t_w \sim 8$ min, using a probing magnetic field of 1 Oe. (b) The rate of change of the magnetization $S = dM/d\ln(t)$ against $\ln(t)$ for the temperatures of 7, 8, 9, 10.5, 12 and 13 K for $Co_{52.5}Al_{47.5}$.

3.2. The $Co_{55}Al_{45}$ cluster-glass

$Co_{55}Al_{45}$ cluster-glass [1] has a transition temperature at ≈46 K, and was investigated for $t_w \approx 7$, 10.5, 14, 24 and 34 min. The relaxation of the M_{ZFC}, measured at temperatures

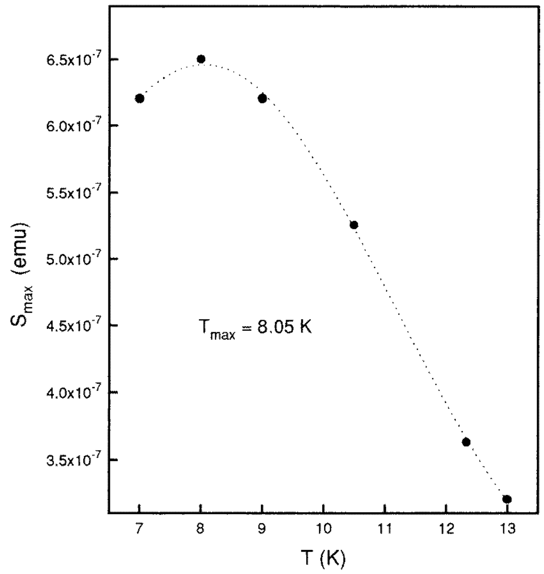

Figure 2. Temperature dependence of the maximal rate of the relaxation $S_{max}(T)$ for $Co_{52.5}Al_{47.5}$, measured after $t_w \sim 8$ min, using 1 Oe. The dotted line presents the fit of the $S_{max}(T)$ data to a Gaussian approximation.

below T_m after applying 1 Oe, produces similar results for the dependence of $S_{max}(T)$ as the results for the $Co_{52.5}Al_{47.5}$ SG.

Figure 4 shows the dependences of S_{max} on T for the smallest, and the largest waiting times. The inset in figure 4 shows the dependence of T_{max} on t_w. A peculiarity is observed, if figure 4 is compared to figure 3: there is an initial increase in T_{max} against t_w, which is followed by a saturation. This initial increase can possibly be assigned to a more complex relaxation process ascribed to the clusters, and the magnetic moments inside them, present in a cluster-glass material [6].

The picture of the domain growth develops further during the longer waiting time. This results in the higher temperature of the maximum of the $S_{max}(T)$ values. Larger quasi-equilibrated domains need higher thermal energy to overcome the higher energy barriers initiated in the sample during the longer waiting time.

For the $Co_{55}Al_{45}$ cluster-glass (see figure 4), T_{max} for the waiting time of $\approx$7 min is about 0.56 T_m, which increases for the saturating waiting times to $\approx$0.60 T_m. This value of T_{max} is the same as the value of T_{max} for $Co_{52.5}Al_{47.5}$ (figure 3).

The gap, defined as the difference between the t_w at which T_{max} is saturated and $t_w = 0$, is further denoted by Δt_w. Δt_w can be large in the case of SG-2-type [6] material. Small clusters and separate magnetic moments exist in the SG-2 state [6]. This more complex magneti structure can possibly need a longer time to reach the saturation in T_{max} against t_w. Thus, the value of Δt_w can be descriptive of the place of the sample on the magnetic phase diagram.

3.3. $Co_{62}Al_{38}$ and $Co_{63}Al_{37}$ re-entrant alloys

To use the wider opportunities of the relaxation measurements as an investigation tool, re-entrant $Co_{62}Al_{38}$ and $Co_{63}Al_{37}$ alloys were measured at sets of low temperatures, after

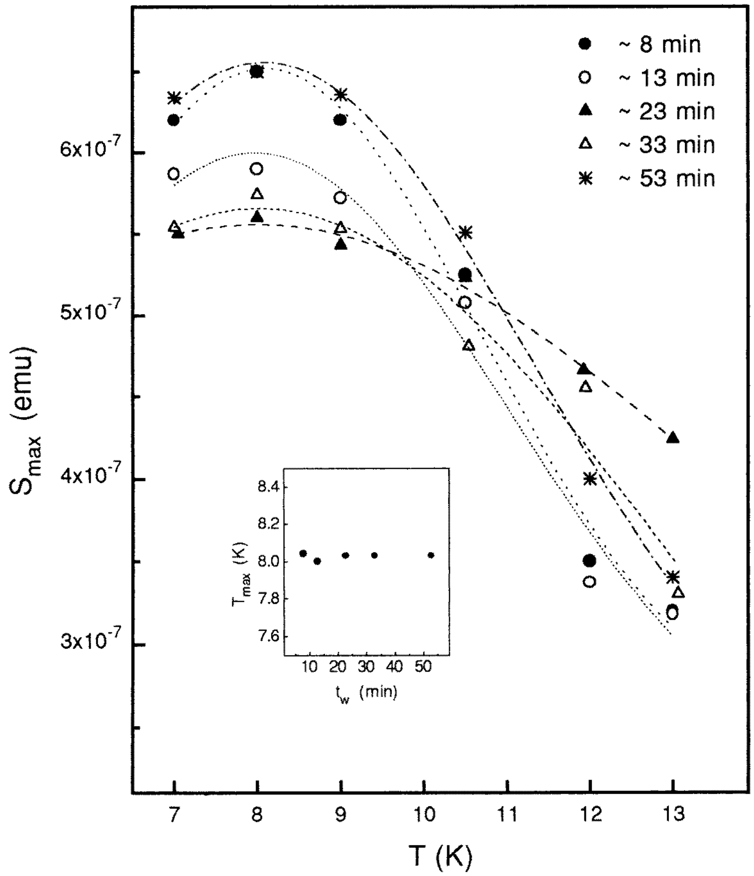

Figure 3. $S_{max}(T)$ for $Co_{52.5}Al_{47.5}$, measured with the waiting times of ~8 min (solid circles), ~13 min (open circles), ~23 min (solid up triangles), ~33 min (open up triangles) and 53 min (stars), using 1 Oe. The inset shows the dependence of T_{max} on t_w. (T_{max} is the temperature of the maximum of the $S_{max}(T)$ dependence.) The different dot and dash lines present the fits of the $S_{max}(T)$ data for the corresponding t_w, to a Gaussian approximation.

applying 1 Oe. It is known [7] that re-entrant alloys have two specific regions: a 'low'-temperature SG-like, called SG–RE, and a 'high'-temperature soft FM region, called RE–FM [8]. The 'low'-temperature relaxation of the magnetization has a maximum in the rate of change of the magnetization [7]. This maximum, and the corresponding collective relaxation behaviour, do not exist in the 'high'-temperature re-entrant region. Ac magnetic susceptibility measurements [7] prove the existence of two peaks in the plots of the re-entrant samples, indicative of a low-temperature SG–RE transition, and a higher temperature RE–FM transition.

Low-temperature relaxation magnetization measurements for $Co_{62}Al_{38}$ were made at 15, 18, 22, 25, 30, 35 and 40 K for the waiting times of ~8 min, and at some additional temperatures for $t_w \sim 13$ min. The dependence of $S_{max}(T)$ shows an analogous maximum in $S_{max}(T)$ to the maxima in $S_{max}(T)$ for $Co_{52.5}Al_{47.5}$ and $Co_{55}Al_{45}$. Re-entrant $Co_{62}Al_{38}$ exhibits this maximum at $T_{max} \approx 32.1$ K for $t_w \sim 8$ min.

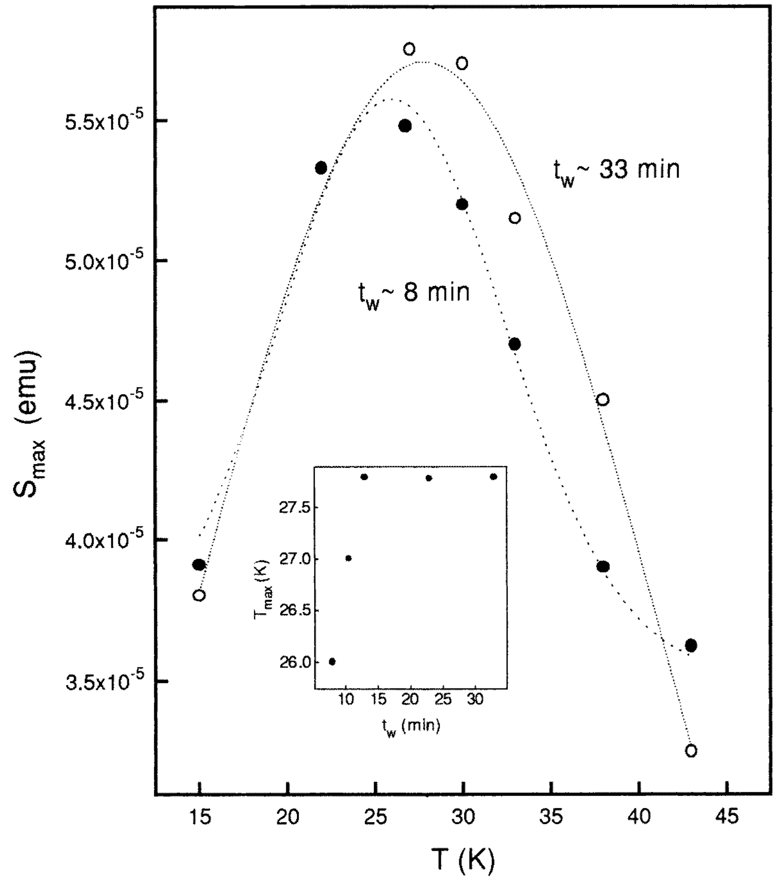

Figure 4. $S_{max}(T)$ for $Co_{55}Al_{45}$, measured with the smallest $t_w \sim 8$ min (solid circles), and the largest $t_w \sim 33$ (open circles). The dotted lines present the fits of the $S_{max}(T)$ data for the relevant t_w, to a Gaussian approximation. The inset shows the dependence of T_{max} or t_w.

The position of T_{max} was investigated for additional treatment of the sample—ageing for a longer time, or further doping with Co.

The maximum in $S_{max}(T)$ at $T_{max} \approx 32.1$ K for $t_w \sim 8$ min moves to 32.7 K for the waiting time of 13 min (figure 5). This maximum can be initiated by the large low-temperature clusters, which possess rigid magnetic moments, and possibly act like the clusters in $Co_{55}Al_{45}$.

Further, more precise experiments for investigating the development of the gap Δt_w when using longer waiting times are desirable. At present, the existence of a maximum in $S_{max}(T)$, which depends on t_w, can only be reported. However, it is not possible by the present results to relate the temperature of the maximum in $S_{max}(T)$, T_{max}, to any of the well known temperatures of the SG–RE phase, derived by e.g. ac measurements [7].

M_{ZFC} relaxation measurements for the higher-Co-concentration sample, $Co_{63}Al_{37}$, were made for 10, 15, 18, 21, 25, 30 and 35 K, after ageing the sample for ~8 min, and afterwards using a magnetic field of 1 Oe. The peak in $S_{max}(T)$ is at ≈21.4 K. $Co_{63}Al_{37}$ is reported to be closer to the FM border of the MPD [1], and therefore the temperature range of existence of the SG–RE phase is smaller.

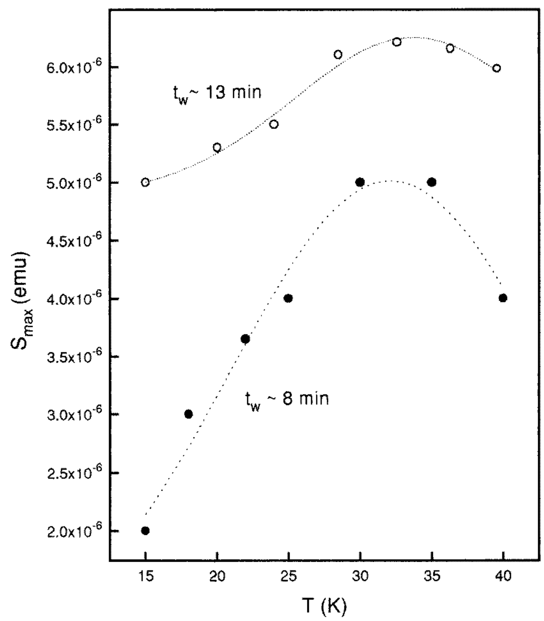

Figure 5. $S_{max}(T)$ for $Co_{62}Al_{38}$, measured with $t_w \sim 8$ min (solid circles), and $t_w \sim 13$ min (open circles). The dotted lines present the fits of the $S_{max}(T)$ data for the corresponding t_w, to a Gaussian approximation.

If enough samples with different concentrations of the dopant x are available, the dependence of T_{max} on x for the low-temperature spin-glass–re-entrant phase can be determined. Thus, the method allows independent assessment of the development of the low-temperature SG–RE phase through doping.

Summarized, all investigated alloys show a maximum in the temperature dependence of the maximal rate, $S_{max}(T)$. The temperature of this maximum, T_{max}, is $\approx$0.60 T_m for the investigated spin-glass and cluster-glass samples. This temperature decreases with increase of the dopant concentration for the low-temperature spin-glass–re-entrant phase.

T_{max} against t_w is practically constant for the spin-glass, and shows an initial increase followed by saturation for the cluster-glass and the low-temperature SG–RE phases.

The value of the gap Δt_w, defined as the time from 0 to the waiting time, at which T_{max} becomes saturated, can also be used to distinguish the position of an alloy on the MPD.

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