Fast thermoremanent magnetization in compounds of the modified pyrochlore structure $CsM_1^{2+}M_2^{3+}F_6$

1994 J. Phys.: Condens. Matter 6 L607

(http://iopscience.iop.org/0953-8984/6/40/001)

J. Phys.: Condens. Matter 6 (1994) L607–L611. Printed in the UK

LETTER TO THE EDITOR

Fast thermoremanent magnetization in compounds of the modified pyrochlore structure $CsM_1^{2+}M_2^{3+}F_6$

D Enkler and M Steiner†

Institut für Physik, Johannes Gutenberg-Universität, D-55099 Mainz, Germany

Received 12 August 1994

Abstract. The thermoremanent magnetization in the insulating spin glasses $CsNiFeF_6$, $CsCoFeF_6$, and $CsCoCrF_6$ was measured with a superconducting quantum interference device magnetometer below the spin glass temperature. The time range of the observed decay of the magnetization after removing an applied magnetic field between 2 and 43 G was from 10^{-2} to 10^3 s. It is shown that the experimental results follow closely the form predicted by a new theory of continuous phase transitions.

Relaxation effects in spin glasses can be observed in the entire time range experimentally accessible, from 10^{-14} to 10^{-8} s with neutron scattering, 10^{-6} to 10^1 s with AC susceptibility, and 10^{-2} to 10^5 s with thermoremanent magnetization (TRM). Especially the TRM provides the possibility to observe the relaxation in spin glasses after switching off the magnetic field directly in the time domain. The decay of the magnetization can be compared with the forms predicted by theories and models. In the present study, measurements of the TRM in compounds showing spin glass behaviour are compared with the predictions of a new theory.

The sample substances $CsNiFeF_6$, $CsCoFeF_6$, and $CsCoCrF_6$ are crystalline, isolating compounds of the modified pyrochlore structure [1] with the space group $Fd3m$. The transition metal ions form a corner sharing tetrahedral network with antiferromagnetic coupling via superexchange between nearest neighbours and negligible higher-order coupling. Thus spin glass behaviour can be expected because of the frustration in the magnetic system and because of the random distribution of the magnetic ions on equivalent lattice sites 16*c*. The samples used in this investigation are single crystals [2].

The spin glass state in the sample substances has been confirmed previously. In the case of $CsNiFeF_6$ detailed investigations of the magnetic properties in powdered samples exist [3, 4, 5, 6]. Experimental results on $CsCoFeF_6$ and $CsCoCrF_6$ are published in [7, 8]. All substances show a Curie–Weiss behaviour in the vicinity of the cusp temperature T_{cusp}, which differs from the temperature T_{split} at which irreversibilities, marked by a splitting of the field cooled and zero-field cooled curves, occur. The relevant numbers from [8] are shown in table 1.

The superconducting quantum interference device (SQUID) magnetometer used in this experiment was especially designed for the measurement of magnetic relaxation beginning at small times after a rapid change of the magnetic field, in order to obtain a large time range for comparison with models. Magnetic fields up to 200 G can be switched within

† Present address: Technische Universität Berlin and Hahn-Meitner-Institut, Glienickerstrasse 100, D-14109 Berlin, Germany.

0953-8984/94/400607+05$19.50

Table 1. Magnetic properties of the sample substances. $T_<$ indicates the lower bound of the Curie–Weiss law.

Substance	T_{cusp} (K)	T_{split} (K)	Θ (K)	$T_<$ (K)
$CsNiFeF_6$	4.59(2)	4.49(3)	4.95	6.5
$CsCoFeF_6$	4.45(2)	4.58(3)	−0.9	6
$CsCoCrF_6$	3.38(4)	3.55(5)	−57.5	20

10 μs, while the recovery time of the SQUID system is below 10 ms. Further details of the apparatus are published in [9].

To measure the TRM, the following procedure was applied. At the starting temperature T_s well above the splitting temperature ($T_s/T_{split} > 1.4$) a magnetic field is applied. The sample is cooled with constant rate down to the measuring temperature T_m. Before the field is switched off, the sample is kept for 1000 s at full field in order to reduce non-equilibrium effects as much as possible at longer measuring times, which range to 1000 s. After switching the magnetic field off, the decay of the magnetization is recorded. In order to establish the base line of the magnetization, the signal is also recorded when the sample is heated up after the measuring time to T_s again.

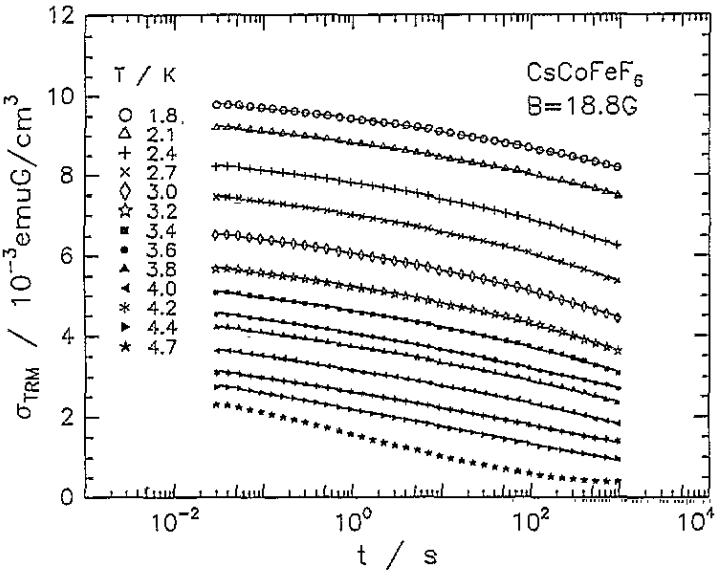

Figure 1. Thermoremanent magnetization in $CsCoFeF_6$ at $B = 18.8$ G. The solid lines are best fits to equation (1).

Experimental results of the TRM in $CsCoFeF_6$ and $CsCoCrF_6$ are shown in figures 1 and 2. At high temperatures, the inflection point between convex and concave form of the decay at longer times is visible, while at lower temperatures the relaxation is purely convex. Results of the TRM in $CsNiFeF_6$ have already been published in [9].

Figure 3 shows the TRM for all three substances and all measured magnetic fields divided by the field cooled (FC) magnetization at the measuring temperature T_m as a function of the relative temperature T_m/T_{split} at one arbitrarily chosen time $t = 40$ s.

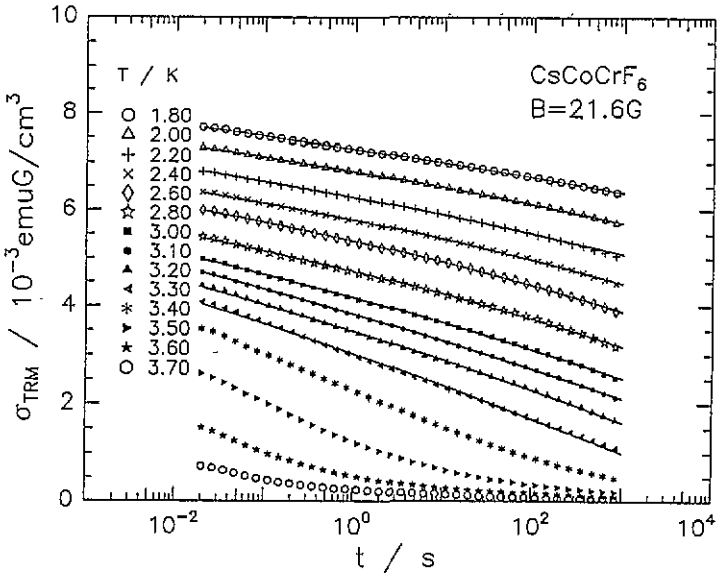

Figure 2. Thermoremanent magnetization in $CsCoCrF_6$ at $B = 21.6$ G. The solid lines are best fits to equation (1).

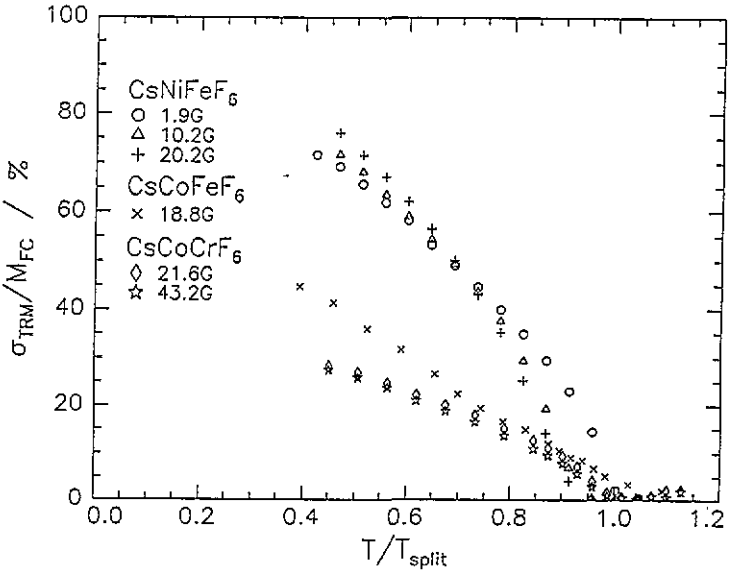

Figure 3. Thermoremanent magnetization relative to the field-cooled magnetization versus the measuring temperature relative to the splitting temperature at $t = 40$ s.

From figure 3, several observations can be made. The dependence of the TRM on the applied field is rather weak, and all curves for one substance coincide quite well. This indicates that the applied field just probes the spin glass state, and does not alter it. The magnetization has decayed fully around the splitting temperature. The magnitude

of the TRM relative to the FC magnetization at the measuring temperature varies from 75% in $CsNiFeF_6$ to 30% in $CsCoCrF_6$ at lowest measured temperatures. This can be attributed to the sign of the interactions as reflected in the Curie–Weiss behaviour at higher temperatures [9], meaning that a ferromagnetic coupling in $CsNiFeF_6$ preserves the field-induced alignment of the spins, while strong antiferromagnetic interaction in $CsCoCrF_6$ leads to a rapid compensation of most spins and thus a lower TRM.

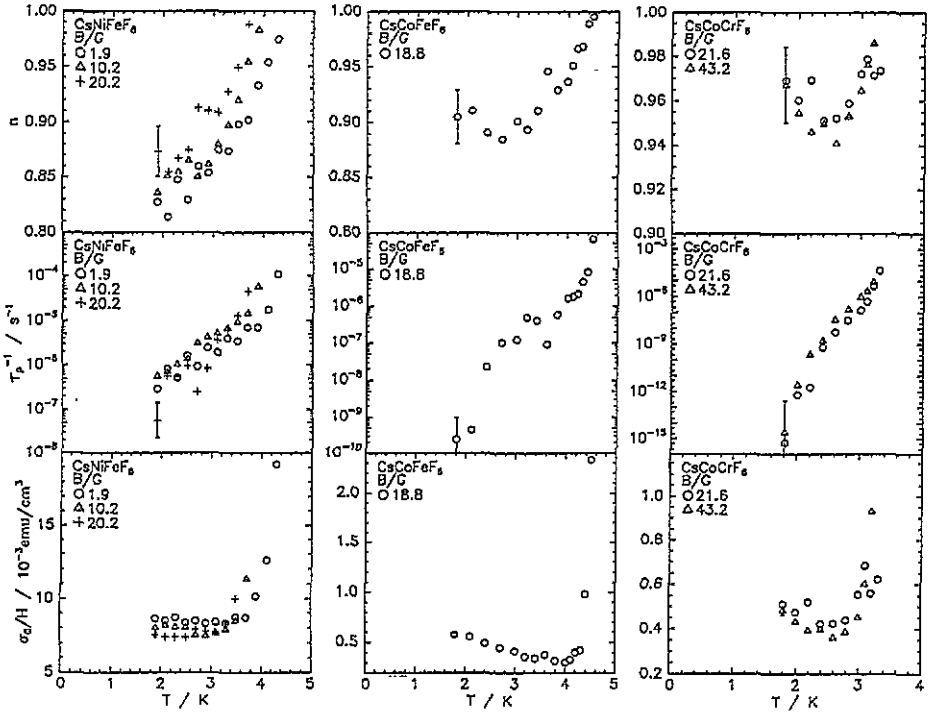

Figure 4. Variation of fit parameters as a function of temperature.

For the decay of the TRM, quite a number of forms have been proposed (see [10] and references therein) and compared with experimental data. Many of these forms lack a theoretical justification and are based on models whose connection to real spin glass systems is not always straightforward. Also, since the TRM generally is a slowly varying function of time, it is possible to obtain a reasonable fit to the data with most functions that possess three or more free parameters. A new general theory of multiscaling and classification of continuous phase transitions by Hilfer [11, 12] allows phase transitions having orders less than unity in Ehrenfest's classification scheme. It is conjectured that these so called anequilibrium phase transitions [13] are candidates for glass transitions. Typical relaxation functions exactly at anequilibrium transitions were found [13] to have the following temporal behaviour:

$$\sigma_{\mathrm{TRM}} = \sigma_0 \sum_{k=0}^{\infty} \frac{t^{k\varpi_t}}{\Gamma(k\varpi_t + 1)} \mathcal{L}^k \tag{1}$$

closely resembling the Kohlrausch–Williams–Watt (KWW) stretched exponential [14] with the substitutions $\varpi = 1-n$ and $\mathcal{L} = \tau_p^{-(1-n)}$, which gives a good description of the TRM for a number of different spin glasses. The advantage of equation (1) is that the fit parameters

follow directly from the theory and allow a physical interpretation. The exponent n is related to the order of the phase transition; τ_p is a decorrelation time, which diverges at the glass transition. Macroscopically, it is the typical one-particle timescale for relaxation.

A fit of the above function to the experimental data yields in all cases a better description than the stretched exponential with a slightly smaller mean square deviation. At temperatures close to the splitting temperature where the form of the curve changes to concave behaviour, both functions cannot be satisfactorily fitted to the data. This form might be better described by a model of dynamically correlated domains following Chamberlin and Haines [10]. The variation of the fit parameters with the temperature is shown in figure 4.

The exponent n shows a slight increase towards higher temperatures. The characteristic frequency τ_p^{-1} increases over three decades in $CsNiFeF_6$, seven decades in $CsCoFeF_6$, and over ten decades in $CsCoCrF_6$ from low to high temperatures. This large difference in the characteristic frequency can again be attributed to the different sign of the magnetic coupling above the spin glass temperature. σ_0 indicates the initial thermoremanent magnetization.

In conclusion, systematic measurements of the TRM over five decades in time on $CsNiFeF_6$, $CsCoFeF_6$, and $CsCoCrF_6$ have enabled a comparison with a new theory by Hilfer for phase transitions. The prediction of the decay of the TRM by Hilfer's new theory and the good coincidence with the experimental data on spin glasses of the modified pyrochlore structure indicates the validity of this theory for spin glasses. Further theoretical work is desired to investigate aging effects, which might lead to the observed deviation of the fit from the experimental data at long times and high temperatures close to the spin glass transition.

Important discussions with Dr R Hilfer and technical assistance by A Roters are gratefully acknowledged.

References

[1] Babel D, Pausewang G and Viebahn W 1967 *Z. Naturf.* b **22** 1219
[2] Wanklyn B M, Wandre F R, Garrard B J, Cermak J and Davison W 1981 *J. Mater. Sci.* **16** 2303
[3] Pappa C and Hammann J 1985 *J. Physique* **46** 637
[4] Pappa C, Hammann J and Jacoboni C 1984 *J. Phys. C: Solid State Phys.* **17** 1303
[5] Pappa C, Hammann J, Jehanno G and Jacoboni C 1985 *J. Phys. C: Solid State Phys.* **18** 2817
[6] Ocio M, Alba M and Hammann J 1985 *J. Physique Lett.* **46** 1101
[7] Meißner B 1991 *PhD Thesis* Technische Universität Berlin
[8] Enkler D, Roters A and Steiner M 1994 *Solid State Commun.* at press
[9] Enkler D, Roters A and Steiner M 1993 *J. Appl. Phys.* **73** 5466
[10] Chamberlin R V and Haines D N 1990 *Phys. Rev. Lett.* **65** 2197
[11] Hilfer R 1992 *Phys. Rev. Lett.* **68** 190
[12] Hilfer R 1993 *Int. J. Mod. Phys.* B **1** 1
[13] Hilfer R 1993 *Phys. Rev.* E **48** 2466
[14] Chamberlin R V, Mozurkewich G and Orbach R 1984 *Phys. Rev. Lett.* **52** 867