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SPIN-GLASS BEHAVIOUR IN Zn$_x$Cd$_{1-x}$Cr$_2$S$_4$ SPINELS

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Abstract. - AC susceptibility measurements have been performed between 110 Hz and 11 kHz on the concentrated insulating spin-glass Zn$_x$Cd$_{1-x}$Cr$_2$S$_4$ (0.4 < x < 0.9). The frequency dependence of the freezing temperature $T_f$ has been analysed in terms of the dynamic scaling models $T = 0$ and $T \neq 0$ transition) and of the Fulcher law.

1. Introduction

Among spin glasses a great deal of interest has been recently devoted to materials with spinel structure, where different types and degrees of frustration may exist. In particular chromium spinels, which are normal in type (i.e. the chromium ions occupy the octahedral sites) appear good candidates to exhibit spin-glass behaviour because they can present a topological frustration (like in ZnCr$_2$Ga$_{2-2x}$O$_4$ [1], where all the interactions are antiferromagnetic) and a frustration due to the competition between n.n. ferromagnetic interactions ($J_1$) and higher order neighbours antiferromagnetic interactions ($J_2$) (like in ZnCr$_2$Ga$_{2-2x}$O$_4$, CdCr$_2$In$_{2-2x}$S$_4$ [2] and Zn$_x$Cd$_{1-x}$Cr$_2$S$_4$ [3]).

The system Zn$_x$Cd$_{1-x}$Cr$_2$S$_4$ seems to be particularly attractive because the disorder in the distribution of the magnetic interactions is not due to the magnetic dilution, as usually, but to the substitution between non magnetic ions (Zn, Cd) on tetrahedral sites. These ions play a role of intermediates in the super-exchange interactions between higher order neighbours and therefore the $J_1 / J_2$ ratio depends on their relative distribution on the tetrahedral sites. Moreover it is very sensitive to fluctuations of concentration. These effects stabilize a spin-glass phase for 0.4 < x < 0.9. In this paper we report a study of the frequency dependence of the freezing temperature $T_f$ for some compositions x.

2. Results and discussion

The samples preparation and characterization is reported elsewhere [3]. We have performed A. C. susceptibility measurements at different frequencies (110 Hz < $\nu$ < 11 kHz) by using a mutual inductance bridge. The discussion of the magnetic phase diagram is reported elsewhere [3]. It includes for 0.0 < x < 0.1 a ferromagnetic region ($T_c = 85$ K for CdCr$_2$S$_4$); for 0.1 < x < 0.4 a region with an intermediate behaviour, characterized by the existence of short range ferromagnetic order and by the appearance of a spin glass-like state at low temperature; for 0.4 < x < 0.9 a spin-glass region indicated by both static [3] and dynamic measurements (Fig. 1); for 0.9 < x < 1 an antiferromagnetic region ($T_N = 20$ K for ZnCr$_2$S$_4$, a complex helicoidal antiferromagnet). We have analysed the frequency dependence of the freezing temperature for some compositions x (0.40; 0.50; 0.85) (Fig. 2) by testing the different laws proposed for describing the spin-glass dynamics. Different criteria are commonly used to define $T_f$ accounting for the existence of a distribution of relaxation times in spin-glasses.

Fig. 1. - $\chi'$ vs. $T$ for different compositions x.

Fig. 2. - $\chi'$ vs. $T$ at different frequencies (1: 120 Hz; 3: 1 150 Hz; 3: 5 140 Hz; 4: 11 550 Hz) for x = 0.5.

$T_f(\nu)$ is taken either as the temperature at which $[\chi' (\nu) - \chi_{\infty}] / \chi_{\infty}$ is close to zero [4] or as the tem-
perature at which $\chi''(\nu) / \chi'(\nu)$ is equal to a very low constant value [5, 6]. We could not use the mentioned criteria as, on one hand, our static measurements were performed applying a quite high field ($H = 400$ Oe) [3] and, on the other hand, the accuracy in the measure of $\chi''(\nu)$ is too low for some frequencies. We have therefore taken $T_f$ as the temperature corresponding to the maximum of $\chi'(\nu)$, even if this is not the best criterion.

The fit of $T_f(\nu)$ to the generalised Arrhenius law $\tau = \tau_0 \exp \left(T^{-z\nu}\right)$, proposed within a model of a $T=0$ transition, gives $10^{-13} < \tau_0 < 10^{-12}$ s and $4 < z\nu < 6$, to be compared to the theoretical value for three dimensions, which is 4 [7].

The use of the power law $\tau = \tau_0 \left[(T - T_0) / T_0\right]^{-z\nu}$, proposed within a $T \neq 0$ transition model, leads to $10^{-13} < \tau_0 < 10^{-12}$ s and $8 < z\nu < 9$, in agreement with the value obtained by computer simulation [8] and reported for many spin-glasses (e.g. Eu$_{0.4}$Sr$_{0.6}$S [5], CdCr$_{1.7}$In$_{0.3}$S$_4$ [6]), for which the existence of a phase transition at finite temperature is confirmed by static measurements.

Finally we have checked the empirical Fulcher law $\tau = \tau_0 \exp \left[E_c / k(T - T_0)\right]$ which also accounts for the data with $\tau_0 = 10^{-13}$ s. The characteristic temperature $T_0$, which should reflect the strength of the magnetic correlations, decreases with increasing $x$ (e.g. $T_0 = 13.6$ for $x = 0.40$; $T_0 = 10.7$ for $x = 0.85$), approaching the antiferromagnetic regime. The $(T_f - T_0) / T_f$ ratio, calculated for $\nu = 110$ Hz, has values between 0.14 and 0.24, which are comparable to those of other insulating spin-glasses, like Eu$_{0.4}$Sr$_{0.6}$S and CdCr$_{1.7}$In$_{0.3}$S$_4$.

The same kind of analysis has been performed for $x = 0.33$, observing similar results. This indicates that also for this composition, where the ferromagnetic interactions are predominant, spin-glass features are observed at low temperature, as confirmed by a maximum of $\chi_c$. However the irreversibility appears at a temperature much higher than $T_f$ [3].

3. Conclusions

AC susceptibility measurements at different frequencies seem to lend support to the existence of a transition at a finite temperature in the investigated samples. Nevertheless, the present results do not allow us to exclude a $T = 0$ transition. The temperature variation of the non linear susceptibility is being measured in order to confirm the existence of a transition at $T \neq 0$.