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PHASE TRANSITION SPIN GLASS–LONG MAGNETIC ORDER IN NEW THIOSPINELS CONTAINING Cu, Cr AND Sb

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The reentrant spin-glass transition was found in the samples of solid solutions $CuCr_{1.5+0.5x}Sb_{0.5-0.5x}S_4$ with $x = 0.34$ and 0.4. The transition temperature to the spin-glass state T_f, determined from the bend on the temperature dependence of the initial susceptibility in an ac magnetic field, depends on the measurement frequency. It was shown that the dependence $T_f(\omega)$ obeys the power law $1/\omega = (1/\omega_0)[T_f/(T_f - T^*)]^{z\nu}$ assuming existence of the phase transition at T^*. The maximum of the temperature dependence of electroresistivity was found for the sample with $x = 0.34$ in T^* region. These facts show that considered spin-glass–long magnetic order transition is the phase transition.

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As is known, the question of the existence of phase transitions from spin glass (SG) to paramagnetic (PM) and from SG to the long magnetic order (LMO) in real materials is one of main problems for SG physics. A great number of papers was devoted to the investigation of the SG–PM transition and rather few concerned SG–LMO transition. In the present paper the experimental evidences of existence of the SG–LMO phase transition in semiconductor spinels $CuCr_{1.5+0.5x}Sb_{0.5-0.5x}S_4$ ($x = 0.34$ and 0.4) are given.

The preparation and basic characteristics of the new system of solid solutions $CuCr_{1.5+0.5x}Sb_{0.5-0.5x}S_4$ ($0 \leq x \leq 0.6$) have been described in [1]. The system features the dilution of the recently discovered antiferromagnetic semiconducting mineral florensovite $CuCr_{1.5}Sb_{0.5}S_4$ with ferromagnetic metallic $CuCr_2S_4$. The samples with $0 \leq x \leq 0.6$ are semiconductors. Magnetic properties of the samples with $0 \leq x \leq 0.2$ are typical of antiferromagnets, namely: the linear dependence of magnetization σ on a field H and the susceptibility maximum at the Néel temperature are observed. The dependence of the low-temperature magnetization on a field for the samples with $x = 0.34$ and 0.4 has a nonlinear character, while the magnetization isotherms are far from saturation up to the maximum field of 30 kOe, in which measurements were performed.

The temperature dependence of initial susceptibility χ for the samples with $x = 0.34$ and 0.4 was measured in an ac magnetic field whose frequency was being changed from 0.25 kHz to 4 kHz. As an example, for the sample with $x = 0.34$, the curve $\chi(T)$ ($\omega = 1$ kHz, $H_\sim = 0.3$ Oe) is represented in Fig. 1. It can be seen from Fig. 1 that, with further temperature decrease, a sharp fall of χ at $T_f = 38$ K takes place. The value of T_f increases from 37.5 K to 39.5 K with frequency changing from 0.25 kHz to 4 kHz. Similar dependence χ on T and ω was observed for the

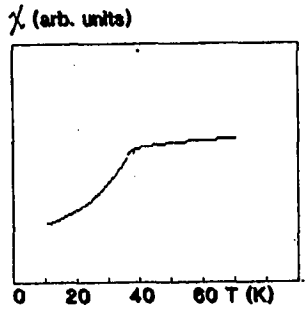

Fig. 1. Temperature dependence of the initial magnetic susceptibility χ in an ac magnetic field $H_\sim = 1$ kHz of sample $CuCr_{1.67}Sb_{0.33}S_4$.

sample with $x = 0.4$. As has been known, such temperature dependence of the initial susceptibility is characteristic of reentrant SG behaviour. Spin-glass state at $T < T_f$ is confirmed by dependence of magnetic properties on the sample thermomagnetic prehistory. Therefore, at $T < T_f$ the field-cooled magnetization is higher than the zero-field magnetization in $H = 57.5$ Oe; and thermoremanent magnetization is higher than isothermal remanent magnetization; this difference disappears at $T \geq T_f$ (Fig. 2a). The displaced hysteresis loops on a H axis of the samples cooled in a weak field are observed for both samples; at the same time a displacement of the loop is not registered for the zero-field cooled samples. The magnetization isotherms of the samples with $x = 0.34$, 0.4 and 0.6 are saturated in temperature region above 77 K in a $H \approx 3$ kOe field. The Curie temperatures of these samples were determined by the Arrott–Belov method. With increasing x from 0.34 to 0.6 the Curie temperature rises from 168 to 334 K.

As has been noted above, in these samples the freezing temperature T_f, determined from a bend of the curve $\chi(T)$, depends on measurement frequency. Apparently, according to which law the dependence $T_f(\omega)$ holds, it may be concluded whether the transition at T_f is a phase transition. It is known that in SG materials the relaxation time in the freezing temperature region is excessive in comparison with Arrhenius law, typical of superparamagnets, and, in a number of cases, it obeys the Vogel–Fulcher empiric law [2]

$$\tau = \tau_0 \exp\left[E/k(T - T_0)\right], \tag{1}$$

where τ_0 is a frequency factor and E is a magnitude with energy dimension. Since the relaxation time τ is diverged at $T = T_0$, on this ground in a number of papers it was supposed that T_0 is SG–PM phase transition temperature. At the same time

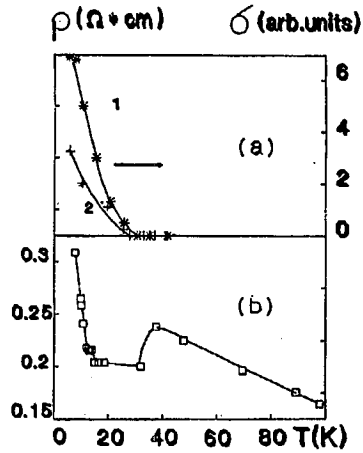

Fig. 2. The sample $CuCr_{1.67}Sb_{0.33}S_4$. (a) Temperature dependence of thermoremanent magnetization TRM (curve 1) and isothermal remanent magnetization IRM (curve 2); (b) temperature dependence of electroresistivity ρ.

it is known that if a phase transition takes place at a certain temperature T^*, the relaxation time of magnetization fluctuations passes the critical slowing-down at T^* and τ obeys the power law [3]:

$$\tau = \tau_0[T/(T - T^*)]^{z\nu}. \tag{2}$$

Here z is the dynamic exponent and ν is the critical exponent of the Edwards–Anderson correlation length. The numerical calculation with the Monte Carlo technique made by Ogielski for the Ising SG with $T^* \neq 0$ ($\pm J$-model, 3D case) gave the value $z\nu = 7.2 \pm 1$.

Binder and Young suggested the alternative hypothesis in which a zero temperature phase transition had been assumed [4]. They obtained the following relation for τ based on the generalized Arrhenius law:

$$\ln \frac{\tau}{\tau_0} = T^{-z\nu}. \tag{3}$$

The numerical calculation with the Monte Carlo technique of the Ising system ($\pm J$-model) made by Binder and Young gave the value $z\nu = 2$ for 2D case and $z\nu = 4$ for 3D case. In the present paper the $T_f(\omega)$ dependence was made to fit the three laws given above for samples with $x = 0.34$ and 0.4. For this purpose the experimental dependences (1)–(3) were plotted on a log–log scale and besides τ was substituted for $1/\omega$ and T for T_f in power and Vogel–Fulcher laws. T_0 in Eq. (1), T^* in Eq. (2) and τ_0 in Eq. (3) were the fitting values. From the plotted dependences we selected those whose experimental points lie satisfactorily on the straight lines. These lines are used to estimate the values of τ_0 and E in the Vogel–Fulcher law, of τ_0 and $z\nu$ in the power law and of $z\nu$ in the generalized Arrhenius law. It was found that the power law with $T = 33.5$ K, $z\nu = 7.7$, $\tau_0 \approx 9.1 \times 10^{-11}$ s is the most suitable one for samples with $x = 0.34$. The Vogel–Fulcher and generalized Arrhenius laws give the strongly excessive values of $\tau_0 \approx 10^{-8}$ s and the Arrhenius

law, on the contrary, gives the underestimated value of $\tau_0 \approx 9.1 \times 10^{-26}$ s. The power law for the sample with $x = 0.4$ also is the most suitable.

The temperature dependence of electroresistivity ρ was also measured in T_f region (Fig. 2b). The maximum on $\rho(T)$ curve in 38 K region a little above of $T = 33.5$ K is typical of magnetic phase transitions. Thus, execution of power law for $T_f(\omega)$ and existence of maximum of ρ in SG–LMO transition region proves that this transition is a phase transition.

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