

# Critical behavior of semiconductor spin glasses $x\text{CuCr}_2\text{Se}_4-(1-x)\text{Cu}_{0.5}\text{Me}_{0.5}\text{Cr}_2\text{Se}_4$ (Me = In, Ga; $0 \leq x < 0.1$ )

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An experimental study was made of the behavior of the magnetic susceptibility in weak static and alternating magnetic fields in the region of the spin glass–paramagnet transition in  $x\text{CuCr}_2\text{Se}_4-(1-x)\text{Cu}_{0.5}\text{Me}_{0.5}\text{Cr}_2\text{Se}_4$  (Me = In, Ga) systems. The critical exponents  $\gamma$ ,  $\delta$ , and  $\beta$  of the compositions with  $x = 0, 0.01$ , and  $0.03$  (Me = In), deduced from the nonlinear static susceptibility  $\chi_{nl}$  in the region of the freezing temperature  $T_f$  corresponding to the maximum susceptibility, were close to the values predicted by the mean-field theory. The experimental values of  $\chi_{nl}$  fitted well a scaling function in the critical range  $H \leq 30$  Oe and  $(T - T_f)/T_f \leq 0.4$ . The freezing temperature  $T_f$  of spin glasses with compositions in the range  $0 \leq x \leq 0.05$  (Me = In, Ga), deduced from the maximum of the initial susceptibility in an alternating field, was a function of the measurement frequency in accordance with the Vogel–Fulcher law. The constant  $T_0$  in the Vogel–Fulcher law was close to  $T_f$ . The statistical scaling relationships and the Vogel–Fulcher law obeyed by  $T_f$  indicated that these spin glasses exhibited a phase transition to the paramagnetic state.

Many experimental investigations of the critical exponents in spin glasses have been reported recently.<sup>1–20</sup> The agreement between the experimental results and the predictions of the scaling theory have been frequently regarded as proof of the existence of a spin glass–paramagnet phase transition. The critical behavior of spin glasses has been found to be more complex than that of ordinary magnetic materials. A divergence of the linear magnetic susceptibility has been observed at the magnetic ordering temperature. The Edwards–Anderson model of a spin glass at the phase transition temperature predicts a divergence of  $\chi_{EA}$ , which is the susceptibility associated with the Edwards–Anderson parameter  $q_{EA}$  for which the external field is the mean square field  $\bar{h}^2$ . However, it is difficult to determine  $\chi_{EA}$  experimentally. Chalupa<sup>21</sup> and Suzuki<sup>22</sup> showed that  $\chi_{EA}$  is proportional to the nonlinear static susceptibility  $\chi_{nl}$  and that the critical behavior of  $\chi_{EA}$  can be investigated by an analysis of the measured values of  $\chi_{nl}$ . Katsura<sup>23</sup> was the first to show, by applying the Bethe approximation in the Ising model with random bonds, that  $\chi_{nl}$  diverges at the phase transition point where its value is negative.

This relationship between  $q_{EA}$  and  $\chi_{nl}$  makes it possible to establish the following scaling relationships for  $T \gtrsim T_f$  (Refs. 8 and 21):

$$\chi_{nl} = \chi_0 - \chi = t^\beta g(H^2/t^{\beta+1}) = t^\beta g(X), \quad (1)$$

$$\chi_{nl} = H^{2/\delta} f(t/H^{2/\Phi}) = H^{2/\delta} f(Y). \quad (2)$$

Here,  $t$  is the reduced temperature defined by

$$t = (T - T_f)/T_f,$$

$H$  is a static magnetic field, and  $\beta$ ,  $\delta$ , and  $\Phi$  represent the following critical exponents:  $\beta$  is the critical exponent of the order parameter  $q$  such that

$$q \propto (T_f - T)^\beta \text{ for } H=0 \text{ and } T \lesssim T_f; \quad (3)$$

$\delta$  is the field critical exponent of  $\chi_{nl}$  at the phase transition temperature  $T_f$ ;  $\Phi$  is the critical exponent for the tempera-

ture–field crossover ( $\Phi = \beta + \gamma$ ). The scaling functions  $g(X)$  and  $f(Y)$  satisfy the following relationships:

$$g(X) \rightarrow g_0 - g_1 X \text{ for } X \rightarrow 0; \quad f(Y) \rightarrow g_3 \text{ for } Y \rightarrow 0, \quad (4)$$

$$g(X) \rightarrow -g_2 X^{1/\delta} \text{ for } X \rightarrow \infty; \quad f(Y) \rightarrow g_4 Y^{-1} \text{ for } Y \rightarrow \infty,$$

where  $g_0, g_1, g_2, g_3$ , and  $g_4$  are constants.

Expansion of the susceptibility described by Eq. (1) in powers of  $H$  near  $T_f$  gives

$$\chi_{nl} = a_2 t^{-1} H^2 + a_4 t^{-(2\gamma+\beta)} H^4 + \dots \quad (5)$$

and it makes it possible to determine directly the values of the critical exponents  $\gamma$  and  $\beta$ . It is usual to determine accurately the magnetization  $M$  in the region of  $T_f$  in weak fields and then differentiate the  $M(H)$  curves in respect of  $H$  and thus obtain a family of curves described by the equation

$$\chi = \chi_0 - \chi_{nl}. \quad (6)$$

We investigated the critical behavior of semiconductor spin glasses in the specific case of solid solutions with compositions described by  $x\text{CuCr}_2\text{Se}_4-(1-x)\text{Cu}_{0.5}\text{Me}_{0.5}\text{Cr}_2\text{Se}_4$  (Me = In, Ga;  $0 \leq x < 0.1$ ), which had been investigated earlier.<sup>24</sup> In the region of the freezing temperature, defined on the basis of the maximum of the initial magnetic susceptibility in a static field, we determined the magnetization of spin glass samples in fields from 10 to 50 Oe. The magnetization measurements were made by a ballistic method. During such measurements a sample was pulled out from a system of two measuring coils wound in opposition and having different diameters. The difference between the number of turns in these coils was  $1 \times 10^3$ , which ensured a high sensitivity (the weak-field magnetization was determined to within 5%, whereas in higher fields the error decreased to 2.5%). A constant magnetic field was created by a single-layer superconducting solenoid. The terrestrial magnetic field was compensated for. In the absence of a current through the solenoid a sample cooled in zero field

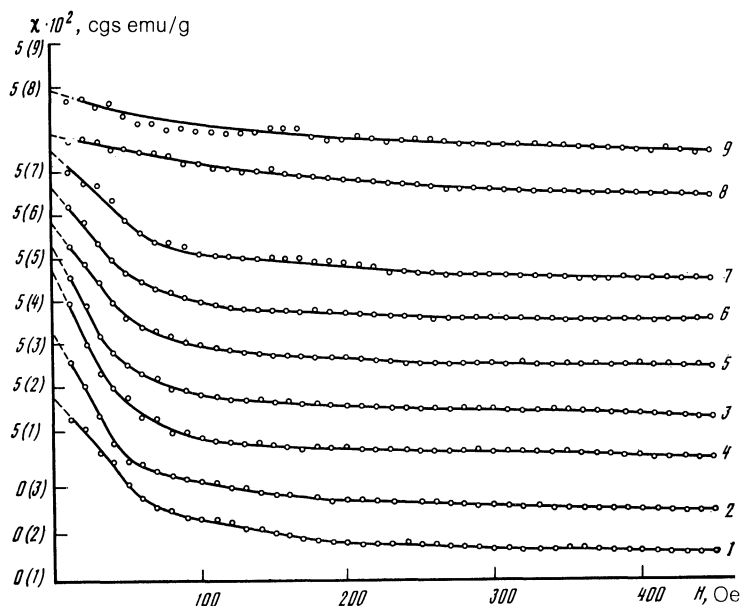


FIG. 1. Dependences of the magnetic susceptibility  $\chi$  of a sample of  $\text{Cu}_{0.5}\text{In}_{0.5}\text{Cr}_2\text{Se}_4$  on  $H$  in the region of  $T_f$  [the dashed parts of the curves represent extrapolation of the dependences  $\chi(H)$  to the field  $H = 0$ ]. The scale is the same for all the curves; temperature  $T$  (K): 1) 4.2; 2) 4.6; 3) 5.0; 4) 5.3; 5) 5.5; 6) 5.7; 7) 6.0; 8) 8.4; 9) 10.8.

showed no magnetic moment, demonstrating that the residual magnetic field of the solenoid did not affect the measurements (because of its smallness). The dependence  $\chi(H)$  was determined by differentiating the  $M(H)$  curves with respect to the field.

Figure 1 shows, by way of example, the dependence  $\chi(H)$  for a sample of the  $\text{Cu}_{0.5}\text{In}_{0.5}\text{Cr}_2\text{Se}_4$  composition in the region of the freezing temperature (5.8 K), deduced from the maximum of the initial magnetic susceptibility in a static magnetic field  $\chi_n$ . Extrapolation of these curves to zero field was used to determine  $\chi_0$  at different temperatures, which made it possible to separate  $\chi_{nl}$  using Eq. (6). The temperature dependence of  $\chi_{nl}$  was determined for the same composition in different magnetic fields (Fig. 2). Clearly, all the curves exhibited a maximum at  $T_f = 5$  K. In accordance with the theory developed in Refs. 21–23, this was the temperature of the spin glass–paramagnet phase transition. The critical exponent  $\gamma$  was determined as fol-

lows. We plotted the  $\chi_{nl}(H^2)$  dependence in the range  $T > T_f$ . Clearly, the slope of the initial linear region of these curves yielded, in accordance with Eq. (5), the value of  $a = a_2 t^{-\gamma}$ ; the slopes were used to find  $a$ . Then, the  $a(t)$  dependence was plotted in a log–log scale. It was found that for the investigated compositions this dependence was linear in the temperature range  $t < 0.4$  and its slope gave the value of  $\gamma$ . We thus found the following values of  $\gamma$  (to within  $\pm 0.1$ ) for the compositions with  $\text{Me} = \text{In}$ :  $\gamma = 1.2$  for  $x = 0$  and  $x = 0.01$ ;  $\gamma = 1.1$  for  $x = 0.03$ .

The critical exponent  $\delta$  was deduced from the field dependence of the nonlinear susceptibility at  $T = T_f$ . In fact, the scaling relationships given by Eqs. (2) and (4) indicated that at  $t = 0$  we should have

$$\chi_{nl} = g_3 H^{2/\delta}. \quad (7)$$

The dependence of  $\chi_{nl}/\chi_0$  on  $H$  is plotted on a double logarithmic scale in Fig. 3 for three compositions with  $x = 0$ ,

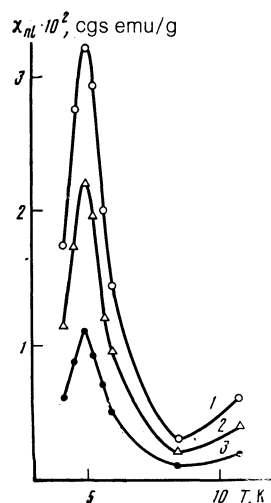


FIG. 2. Temperature dependences of the nonlinear magnetic susceptibility  $\chi_{nl}$  obtained in different magnetic fields (Oe): 1) 30; 2) 20; 3) 10. Composition of the sample  $\text{Cu}_{0.5}\text{In}_{0.5}\text{Cr}_2\text{Se}_4$ .

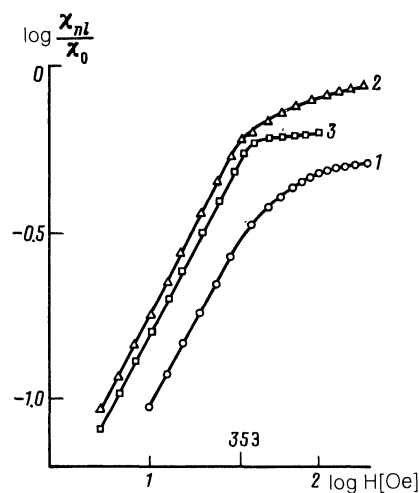


FIG. 3. Field dependence of  $\chi_{nl}/\chi_0$  plotted on a double logarithmic scale at the freezing temperature  $T_f$ , found from the maximum of  $\chi_{nl}$ , for samples with  $\text{Me} = \text{In}$  and  $x = 0$  (1), 0.01 (2), and 0.03 (3).

0.01, and 0.03; clearly, the initial parts are linear and then begin to bend. The correct values of  $2/\delta$  are obviously those obtained in lower fields. We can see from Fig. 3 that the dependence of  $\log(\chi_{nl}/\chi_0)$  on  $\log H$  becomes nonlinear in fields from 30 Oe (for  $x = 0$ ) to 35 Oe (for  $x = 0.01$  and 0.03), i.e., the scaling relationships are obeyed in the range of fields where the dependence is linear. In this case in a field of  $\sim 30$  Oe the change in the nonlinear susceptibility is  $\sim 0.5$  of  $\chi_0$ , i.e., it is approximately as large as in the case of the magnetically concentrated spin glass  $\text{CsNiFeF}_6$  (Ref. 9). The critical exponent  $\delta$ , deduced from the slope of the initial part of the dependence of  $\log(\chi_{nl}/\chi_0)$  on  $\log H$  with an error not exceeding  $\pm 0.1$ , was found to be 2.1, 2.0, and 2.0 for the three compositions mentioned above. The critical exponent  $\beta$  was deduced from the scaling relationship

$$\beta = \frac{\gamma}{\delta - 1} \quad (8)$$

and was 1.1, 1.2, and 1.1 (accurate to within  $\pm 0.2$ ) for the same compositions.

The scaling function  $g(X)$  occurring in Eq. (1) is plotted in Fig. 4 in a log-log scale for the composition  $\text{Cu}_{0.5}\text{In}_{0.5}\text{Cr}_2\text{Se}_4$ . The experimental points in the range of temperature  $t < 0.4$  and fields up to 30 Oe fit well a single curve. The  $g(X)$  curve is plotted on the assumption that  $T_f = 5$  K,  $\beta = 1.1$ , and  $\gamma = 1.2$ . At higher values of  $X$  the scaling function should tend to the limit  $X^{1/\delta}$  [see Eq. (4)]; we can see from Fig. 4 that  $1/\delta \sim 1/2$ , i.e., that  $\delta \sim 2$ , in good agreement with the above value of 2.1 deduced from Eq. (7). Similar scaling functions were plotted also for the compositions with  $x = 0.01$  and 0.03 ( $\text{Me} = \text{In}$ ); the limits for  $t$  and  $H$  for these compositions were approximately the same as for the composition with  $x = 0$  ( $\text{Me} = \text{In}$ ).

The values of the critical exponents obtained by us were close to those predicted by the mean field theory<sup>22,26-29</sup> ( $\gamma = 1, \beta = 1, \delta = 2$ ) also in the case of spin glasses **Ag Mn** ( $\gamma = 1$ ) (Ref. 2) and **Au Fe** (Ref. 7). On the other hand, they differ greatly from the values obtained for other spin

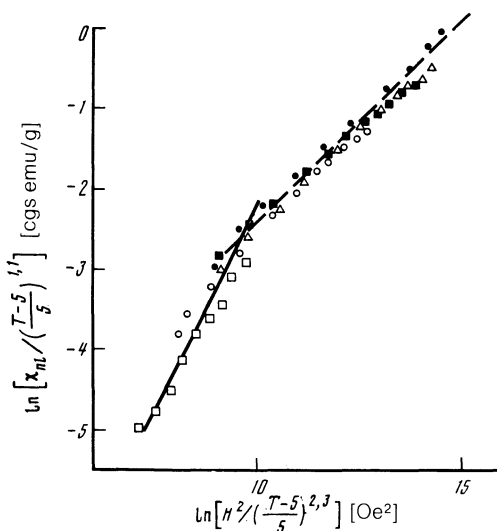


FIG. 4. Scaling function  $g(X) = \chi_{nl}/t^{\beta+\gamma}$  plotted as a function of  $X = H^2/t^{\beta+\gamma}$  on a double logarithmic scale for  $T_f = 5$  K,  $\beta = 1.1$ , and  $\gamma = 1.2$  and the composition  $\text{Cu}_{0.5}\text{In}_{0.5}\text{Cr}_2\text{Se}_4$ . The continuous straight line indicates that the initial slope of the function  $g(X)$  is 1; the dashed line is an asymptote of this function in the limit  $X \rightarrow \infty$  and its slope is  $1/2$ .

glasses<sup>1,8-12</sup> including **Ag Mn**, for which data are reported in Refs. 3-6. The values of the critical exponents reported in these papers lie in the ranges  $2.1 \leq \gamma \leq 4.6$ ,  $0.38 \leq \beta \leq 1.4$ , and  $2 \leq \delta \leq 10$ . Numerical modeling of a three-dimensional Ising system by the Monte Carlo method ( $\pm J$  model with a Gaussian distribution)<sup>30-32</sup> yielded critical exponents very different from those obtained by us:  $2.9 \leq \gamma \leq 3.12$ ;  $0.4 \leq \beta \leq 0.5$ .

In contrast to materials with a long-range magnetic order, such as ferromagnets, the critical exponents of spin glasses vary from one compound to another. One of the possible reasons for the discrepancies between the published values is the difference between the ranges of temperatures and fields used in the experiments. For example, Malozemoff *et al.*<sup>8</sup> estimated the critical region up to  $t = 0.4$  and  $H \leq 15$  kOe, but in calculation of the critical exponents they used the experimental points up to  $t = 2$ . The limiting values of  $t$  were not exceeded in Ref. 10. Bouchiat<sup>3</sup> reported a value of  $\gamma$  in the range  $10^{-2} \leq t \leq 10^{-1}$  and pointed out that  $\gamma$  changed at higher values of  $t$ . According to Barbara and Malozemoff,<sup>11</sup> the discrepancy between the experimentally determined critical exponents and those predicted by the mean field theory is due to the fact that in the case of spin glasses we cannot neglect fluctuations of the local fields because their average values are small as a result of competition between the exchange interactions. In their opinion, another reason for the discrepancy is the large size of the critical region, particularly in the percolation model of spin glasses.<sup>1</sup>

In our opinion, determination of the phase transition temperature is a very important aspect of determination of the critical exponents. It is known that the freezing temperature of spin glasses depends on the measurement frequency and there is as yet no agreement on the occurrence of the phase transition itself in spin glasses.<sup>1</sup> The temperature  $T_f$  was determined in the cited investigations by a variety of methods in the presence of a magnetic field and the measurement time was finite.

In contrast to earlier investigations, we determined the freezing temperature from the maximum of  $\chi_{nl}$  as recommended in the mean field theory treatments.<sup>21-23</sup> Moreover, we did this in a very narrow critical range  $t \leq 0.4$  and  $H \leq 35$  Oe. Clearly, this was the reason why we obtained values of the critical exponents  $\gamma$ ,  $\delta$ , and  $\beta$  and the scaling function  $g(X)$  close to those predicted by the mean-field theory, which can be regarded as a confirmation of the existence of a phase transition in the investigated spin glasses.

In our earlier investigation<sup>24</sup> we determined the freezing temperature  $T'_f$  from the maximum of the initial susceptibility in an alternating magnetic field of frequency  $\omega \leq 8$  kHz and observed that  $T'_f$  was a function of the frequency. The relative change in  $T'_f$  with the frequency  $\omega$  (i.e., the value of  $\Delta T'_f/T'_f \Delta \log \omega$ ) was found to be fairly large and approximately the same as for  $\text{Eu}_x\text{Sr}_{1-x}\text{S}$  (Ref. 33). The frequency dependence of  $T'_f$  reported in Ref. 24 was described only approximately by the Arrhenius law

$$\tau = \tau_0 \exp(E_a/kT'_f), \quad (9)$$

where  $\tau = 1/\omega$ ,  $E_a$  is the energy of the potential barrier, and  $\tau_0$  is the frequency factor. Although the experimental points fitted well the linear dependence of  $\log \omega$  on  $1/T'_f$ , the value

of  $\tau_0$  varied within wide limits from  $10^{-9}$  to  $10^{-22}$  s.

The frequency dependence of  $T'_f$  for several other spin glasses obeyed the Arrhenius law only for unrealistic values of  $\tau_0$ : for example, in the case of Cu Mn it was found that  $\tau_0 = 10^{-200}$  s, which led to the proposal of the empirical Vogel–Fulcher law<sup>34</sup>:

$$\tau = \tau_0 \exp[E_a/k(T - T_0)]. \quad (10)$$

Since the relaxation time  $\tau$  in Eq. (10) diverges at  $T = T_0$ , it was assumed that  $T_0$  is the phase transition point. However, if the phase transition occurs at some temperature  $T^*$ , the relaxation time of fluctuations of the magnetization exhibits a critical increase in  $T^*$ . The relaxation time is related to the Edwards–Anderson correlation length  $\xi_{EA}$  with the aid of a dynamic exponent  $z$  (Ref. 35):

$$\frac{\tau}{\tau_0} \propto \xi_{EA}^z. \quad (11)$$

It is known that if  $H = 0$  and  $T \gtrsim T^*$ , we have

$$\xi_{EA} \propto \left(\frac{T - T^*}{T}\right)^{-\nu}, \quad (12)$$

where  $\nu$  is the critical exponent of  $\xi_{EA}$ . Substituting Eq. (12) into Eq. (11), we obtain the power law for  $\tau$ :

$$\tau = \tau_0 \left(\frac{T}{T - T^*}\right)^{z\nu}. \quad (13)$$

Here,  $\tau_0$  is the characteristic relaxation time, i.e., it is the shortest average relaxation time of the system, amounting to

$$\tau_0 \approx \frac{\hbar}{kT^*}. \quad (14)$$

In the case of real spin glasses with the freezing temperature 1–10 K this relaxation time is  $\sim 10^{-12}$  s. Numerical Monte Carlo modeling of an Ising spin glass with  $T^* \neq 0$  ( $\pm J$  model, three-dimensional case) carried out by Ogielski<sup>30</sup> gave  $z\nu = 7.2 \pm 1$ .

Binder and Young<sup>36</sup> proposed an alternative hypothesis in which the temperature of the spin glass–paramagnet phase transition was assumed to be zero. They postulated that in this case we should have

$$\ln \frac{\tau}{\tau_0} \propto \xi_{EA}^z, \quad (15)$$

and for  $H = 0$  they obtained

$$\ln \frac{\tau}{\tau_0} = T^{-z\nu}. \quad (16)$$

The relationship (16) is known as the generalized Arrhenius law.

The numerical Monte Carlo modeling of an Ising system ( $\pm J$  model) carried out by Binder and Young<sup>36</sup> gave  $z\nu = 2$  for the two-dimensional case and  $z\nu = 4$  for the three-dimensional situation.

In several recent investigations the dependence  $T'_f(\omega)$  was compared with three laws: the Vogel–Fulcher law of Eq. (10), the power law of Eq. (13), and the generalized Arrhenius law of Eq. (16). For example, according to Ref. 13 the value of  $\tau$  for the spin glass  $\text{Eu}_{0.4}\text{Sr}_{0.6}\text{S}$  obeys satisfactorily both the power law with  $T^* = 1.5$  K ( $\tau_0 = 2 \times 10^{-7}$  s,  $z\nu = 7.2$ ) and the logarithmic law with  $T^* = 0$  K ( $\tau_0 = 10^{-5}$  s,  $z\nu = 8$ ). An even lower value amounting to

$\tau_0 = 2 \times 10^{-8}$  s ( $z\nu = 10.6$ ) was obtained in Ref. 14 by approximation of the dependence with a power law at  $T^* = 1.54$  K. Similar investigations had been carried out for spin glasses  $\text{CdCr}_{1.7}\text{In}_{0.3}\text{S}_4$  (Refs. 15 and 16),  $\text{Cd}_{0.6}\text{Mn}_{0.4}\text{Te}$  (Ref. 17), and amorphous samples of  $(\text{Fe}_{0.15}\text{Ni}_{0.85})_{75}\text{P}_{16}\text{B}_6\text{Al}_3$  (Ref. 19) and  $\text{Mn}_3\text{Al}_2\text{SiO}_{12}$  (Ref. 20), obeying the power law, and for  $\text{Fe}_2\text{TiO}_5$  (Ref. 18), in which case all three laws could be used.

We compared the frequency dependence of  $T'_f$  for spin glasses belonging to the  $x\text{CuCr}_2\text{Se}_4 - (1-x)\text{Cu}_{0.5}\text{Me}_{0.5}\text{Cr}_2\text{Se}_4$  system using all three laws. The results are presented in Table I. This table includes also the values of the freezing temperature deduced from the maxima of  $\chi_{nl}$  and from the initial susceptibility measured in a static magnetic field ( $\chi_n$ ). The fitting was carried out as follows. For each composition we plotted the dependences (9), (10), (13), and (16) on a logarithmic scale and substituted the values of  $T = T'_f$ . In the case of the power and Vogel–Fulcher laws, we substituted for  $T_0$  and  $T^*$  the freezing temperatures deduced from the maximum of  $\chi_{nl}$  and the maximum of  $\chi_n$ , as well as several other arbitrarily selected temperatures located near the two temperatures just mentioned. From these plotted dependences we selected those for which the experimental points fitted satisfactorily the corresponding straight lines and these were used to find the values of  $\tau_0$ ,  $z\nu$ , and  $E_a$ . In the case of the generalized Arrhenius law the fitting values was  $\tau_0$ . Table I lists the values of  $\tau$ ,  $z\nu$ , and  $E_a$  deduced from the most successful fitting to each law, whereas Fig. 5 gives the most successful fitting curves for the composition  $\text{Cu}_{0.5}\text{In}_{0.5}\text{Cr}_2\text{Se}_4$ .

It is clear from Table I that the Vogel–Fulcher law was satisfied well by the majority of the investigated compositions and for those for which we determined  $\chi_{nl}$ , the value of  $T_0$  was either equal to  $T_f$  deduced from the maximum of  $\chi_{nl}$  ( $\text{Me} = \text{In}$ ,  $x = 0.01$  and  $0.03$ ) or a value close to it but smaller than  $T_f$  deduced from the maximum of  $\chi_n$  ( $\text{Me} = \text{In}$ ,  $x = 0$ ). In the case of compositions with  $x = 0.05$  ( $\text{Me} = \text{In}$ ,  $\text{Ga}$ ) the value of  $T_0$  was less than  $T_f$  deduced from the maximum of  $\chi_n$ . The power law of Eq. (13) was hardly suitable for our spin glasses because in this case we obtained an unrealistic value of  $z\nu$ ; its minimum value 12.9 was obtained for the composition with  $x = 0.03$  ( $\text{Me} = \text{In}$ ); for the other compositions the values were even higher (Table I). On the other hand, Ogielski<sup>30</sup> estimated, as mentioned above, that  $z\nu = 7.2$ . In the case of the generalized Arrhenius law the value of  $z\nu$  was found by us to be less than  $z\nu = 4$  predicted by Binder and Young.<sup>36</sup> The best fit (for  $\text{Me} = \text{In}$ ,  $x = 0.05$ ) was obtained by assuming that  $z\nu = 2.5$ , which was considerably less than 4. In the case of  $x = 0.1$  ( $\text{Me} = \text{In}$ ,  $\text{Ga}$ ) none of these three laws was obeyed, but the best results were obtained employing the Arrhenius law.

Our results thus demonstrated that the static scaling relationships are obeyed by some semiconductor spin glasses in the form of  $x\text{CuCr}_2\text{Se}_4 - (1-x)\text{Cu}_{0.5}\text{Me}_{0.5}\text{Cr}_2\text{Se}_4$  ( $\text{Me} = \text{In}$ ,  $\text{Ga}$ ) solid solutions. The critical exponents  $\gamma$ ,  $\delta$ , and  $\beta$  were close to those predicted by the mean field theory. The frequency dependence of  $T'_f$  obeyed the Vogel–Fulcher law. The constant  $T_0$  in this law was the temperature close or equal to the temperature of the transition in the static scaling. Hence, we concluded that a spin glass–paramagnet phase transition occurred at this temperature. However, at

TABLE I. Fitting of the dependence  $T_f'(\omega)$  to various laws.\*

Composition		$T_f^{(1)}, \text{K}$	$T_f^{(2)}, \text{K}$	Generalized Arrhenius law		Power law			Vogel-Fulcher law	
				$\tau_0, \text{S}$	$z\nu$	$\tau_0, \text{S}$	$z\nu$	$T^*, \text{K}$	$\tau_0, \text{S}$	$E_a, \text{meV}$
=In,	$x=0$	5	5.8	$10^{-12}$	2.0	$5.4 \cdot 10^{-11}$	46.6	5	<u><math>4.64 \cdot 10^{-13}</math></u>	<u>8.36</u>
	0.01	5.5	6	$10^{-12}$	1.93	$2.56 \cdot 10^{-13}$	27.6	6	<u><math>1.39 \cdot 10^{-12}</math></u>	<u>9.26</u>
	0.03	6.3	8.5	$10^{-12}$	2.18	<u><math>2.57 \cdot 10^{-12}</math></u>	<u>12.9</u>	<u>8.5</u>	<u><math>8.4 \cdot 10^{-12}</math></u>	<u>1.44</u>
	0.05		9.5	$3.45 \cdot 10^{-12}$	2.5	<u><math>3.81 \cdot 10^{-13}</math></u>	<u>14.9</u>	9	<u><math>1.38 \cdot 10^{-12}</math></u>	<u>8.3</u>
	0.1		10.9	$3.85 \cdot 10^{-13}$	0.93	$8.42 \cdot 10^{-10}$	17	9.3	$4.16 \cdot 10^{-9}$	8.92
=Ga,	$x=0.05$		8.6	$10^{-10}$	2.17	$8.4 \cdot 10^{-11}$	13.7	8.6	<u><math>4.18 \cdot 10^{-10}</math></u>	<u>5.43</u>

\* Here,  $T_f^{(1)}$  and  $T_f^{(2)}$  are the freezing temperatures deduced from the maximum of  $\chi_{nl}$  and from the maximum of  $\chi_n$ , respectively. The results of the best fits are underlined.

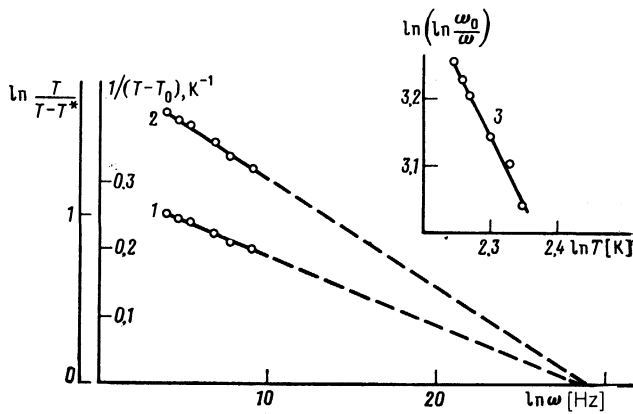


FIG. 5. Relationships represented by Eqs. (10), (13), and (16) (1, 2, and 3, respectively) plotted on a double logarithmic scale for parameters  $T_f$ ,  $\tau_0$ ,  $\nu$ , and  $E_a$  ensuring the best fitting; composition  $\text{Cu}_{0.5}\text{In}_{0.5}\text{Cr}_2\text{Se}_4$ .

higher values, such as  $x = 0.1$ , this phase transition did not occur because the frequency dependence of  $T_f'$  of these compositions obeyed the Arrhenius law.

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