

# Paramagnet-spin-glass phase transition in dilute ferrimagnetic oxides

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(Submitted 23 February 1989; resubmitted after revision 29 November 1989)

Zh. Eksp. Teor. Fiz. 97, 1208–1217 (April 1990)

For the case of the system of dilute ferrimagnetic oxides  $\text{Li}_{0.5}\text{Fe}_{2.5-x}\text{Ga}_x\text{O}_4$  ( $x = 1.5-2.0$ ), for which, at low temperatures, a number of properties characteristic of the spin-glass state are observed, the question of the existence and features of a paramagnet-spin-glass phase transition in Heisenberg systems with short-range exchange interaction and random anisotropy is considered. The critical behavior in a magnetic field is investigated and the one-valley order parameter  $q_{EA}$  is calculated.

## 1. INTRODUCTION

The spin-glass (SG) state has now been intensively studied for more than fifteen years. Despite this, such questions as the existence and character of the phase transition at the freezing temperature  $T_f$  and the structure of the low-temperature phase at  $T < T_f$  have not lost their urgency.<sup>1-3</sup> Less studied at this level have been Heisenberg systems with short-range interaction. The results presently available are very contradictory. On the one hand, computer modeling and theoretical calculations, using various approaches, give a value greater than three for the lower critical dimensionality,<sup>2,3</sup> whereas experimentally in systems of this kind typical spin-glass properties are observed in a nonzero temperature range from 0 K to  $T_f$ , the temperature  $T_f$  being clearly displayed in static and dynamic experiments and also in neutron-diffraction studies.<sup>1-4</sup> This contradiction is usually explained by the action of random anisotropy of the SG, as a result of which Heisenberg systems behave like Ising systems;<sup>2,3,5</sup> this, strictly speaking, has been demonstrated in detail only for systems with an infinite interaction range.

Although attempts have been made recently to construct a theory for SG systems with short-range interaction (e.g., the droplet model of Ref. 6), the most developed theory is the mean-field (MF) theory with infinite-range interaction, in the framework of which many characteristic properties and distinctive features of the SG state have been successfully described.<sup>1,2,7</sup> According to the results of the MF theory, the principal distinguishing feature of the SG state is its degeneracy and nonergodicity: The energy spectrum of an SG is a set of valleys (sets of states) separated by infinite barriers. It is because of this that the typical SG properties (magnetic irreversibility and long-time viscosity) arise: For  $T \lesssim T_f$  there exist equilibrium ( $\chi_{\text{eq}}$ ) and nonequilibrium ( $\chi_{\text{neq}}$ ) susceptibilities (magnetizations), which are identified, respectively, with the experimentally observable quantities  $\chi_{FC}$  (preliminary cooling of the sample with  $H \neq 0$ ) and  $\chi_{ZFC}$  (cooling without the field); the nonequilibrium magnetization  $\sigma_{ZFC}(H, T = \text{const})$  depends on the observation time either logarithmically or by a power law with a small power exponent, and the spectrum of relaxation times is very broad.<sup>1,2,8</sup>

In the mean-field model the transition to the SG state from the paramagnetic (PM) phase is characterized by breaking of the replica symmetry at  $T = T_f$  ( $T \rightarrow T_f^+$ ) and by the appearance of a nonzero order parameter  $q(T) = \langle \langle \mathbf{S}_i \rangle_T^2 \rangle_J$ , where  $\langle \dots \rangle_T$  denotes thermodynamic averaging and  $\langle \dots \rangle_J$  denotes configurational averaging; for  $T > T_f$

we have  $q(T) = 0$  (Refs. 1, 2, 7). The transition is preserved in the presence of a magnetic field  $H \neq 0$ , with  $T_f(0) \neq T_f(H)$ . For Ising spin glasses the critical line [the de Almeida-Thouless (AT) line] is described by the dependence  $\tau' \propto H^{2/3}$ , where  $\tau' = 1 - T_f(H)/T_f(0)$ , while for Heisenberg spin glasses the critical line [the Gabay-Toulouse (GT) line] is described by  $\tau' \propto H^2$  (Refs. 9, 10). A substantial influence on the form of the critical lines  $T_f(H)$  in Heisenberg spin glasses is exerted by random anisotropy: In small fields, instead of the GT line a relation corresponding to a line of the AT type is obeyed.<sup>11-13</sup>

In contrast to Ising systems, in vector spin glasses the description of the phase transition requires the introduction of multi-component order parameters;<sup>10</sup> however, because of the action of random anisotropy the PM-SG phase transition in Heisenberg spin glasses can acquire features that are characteristic of Ising systems and can be described by a single one-component order parameter  $q(T)$  (Ref. 13).

Order parameters that take explicit account of the degeneracy and nonergodicity of a spin glass were proposed in the theory of Parisi<sup>14</sup> and in the dynamical SG theory of Sompolinsky:<sup>11,15</sup>

$$q_P(T) = \int_0^1 q(x) dx, \quad (1)$$

$$q_S(T) = q(x=1) - \Delta(x=0). \quad (2)$$

In Parisi's theory the role of the order parameter is played by the function  $q(x)$ , whose argument  $x$  ( $x = 0-1$ ) has the meaning of the probability of realizing states that arise from the overlap of valleys, while  $q(x=1)$  is the one-valley order parameter. The Sompolinsky function  $\Delta(x)$  is a measure of the nonergodicity of the system:  $\Delta(x=1) = 0$ , and  $\Delta(x=0) = \chi_{\text{eq}} - \chi_{\text{neq}}$ . The explicit form of the functions  $q(x)$  and  $\Delta(x)$  for  $T < T_f$  is undetermined, but near  $T_f$  ( $T \rightarrow T_f^-$ ), in the region of weak nonergodicity,  $q(T) \approx q(x=1)$ . We note that the one-valley order parameter  $q(x=1)$  coincides with the Edwards-Anderson (EA) parameter

$$q_{EA}(T) = \lim_{t \rightarrow \infty} \langle \langle \mathbf{S}_i(0) \mathbf{S}_i(t) \rangle_T \rangle_J,$$

where  $\mathbf{S}_i(0)$  and  $\mathbf{S}_i(t)$  are the values of the spin at site  $i$  at times 0 and  $t$ , respectively.<sup>2,11,14,15</sup>

Thus, the existence of a PM-SG transition in the EA order parameter  $q_{EA}$  at the temperature  $T_f$ , taken together with the existence at  $T < T_f$  of typical SG properties asso-

ciated with the nonergodicity and degeneracy of the SG, implies in practice the existence of a transition to the SG state in the dynamical sense (i.e., in the sense of Sompolinsky) or in the Parisi order parameter. The parameter  $q_{EA}$  can be calculated in the entire range of temperatures  $T \leq T_f$ , and near the instability line  $T_f(H)$  it can be represented in the form of a series in powers of the reduced temperature  $\tau = 1 - T/T_f$  (Refs. 1, 15):

$$q_{EA}(T) = \tau + \tau^2 - \tau^3 + \dots \quad (3)$$

In addition, it is necessary to note that the existence of the PM-SG transition in the order parameter  $q_{EA}$  is also assumed in the droplet model of an SG, but, according to the results of Ref. 6, the AT or GT instability lines are absent.

## 2. THE OBJECTS OF THE INVESTIGATION: FORMULATION OF THE PROBLEM

In the present paper we present the results of an investigation of the PM-SG transition in the system of dilute ferrimagnetic oxides  $\text{Li}_{0.5}\text{Fe}_{2.5-x}\text{Ga}_x\text{O}_4$ , in which, according to the  $x$ - $T$  diagram of Ref. 16, such a transition occurs for  $x > 1.5$ . The class of compounds under consideration is unusual in the physics of spin glasses, but is of undoubted interest from the standpoint of the questions discussed above, by virtue of specific features of the principal interactions.

In ferrimagnetic oxides the dominant interaction giving rise to the spin ordering is the short-range indirect exchange interaction of magnetically active cations via an oxygen anion. A prerequisite for a transition to the disordered states of the SG type that are realized upon diamagnetic dilution is competition of the inter- and intra-sublattice antiferromagnetic interactions.<sup>16-18</sup> Thus, disordered ferrimagnetic oxides are Heisenberg systems with short-range interaction.

Because of the short-range character of the exchange, an inherent property of dilute ferrimagnetic oxides is clusterization—the formation, upon diamagnetic dilution, of regions of short-range order (ferrimagnetic or antiferromagnetic) that are stable in time.<sup>16,17,19</sup> Although the question of the presence of clusters in SG systems (including the classical, strongly diluted RKKY alloys) has been discussed repeatedly,<sup>1,20,21</sup> the problem of the structure of the low-temperature SG phase remains, as before, unsolved.<sup>1,2,6</sup> In the framework of classical ideas, i.e., in the MF model, as the structural unit of the SG state one always considers an individual spin, while in the droplet model one considers a spin cluster. The important point is that in such approaches, as already noted, different critical behaviors in a magnetic field are predicted. In addition, there are experimental data on the existence of disordered systems of the cluster type in which, despite the presence of certain similar SG properties, there is no phase transition to the SG state at  $T_f > 0$  K. The freezing of the magnetic moments of the clusters occurs in the proper anisotropy field in a broad range of temperatures (down to 0 K).<sup>22</sup>

Thus, an experimental study of the presence in dilute ferrimagnetic oxides (disordered cluster Heisenberg systems) of the PM-SG phase transition at  $T_f > 0$  K and of the critical behavior in an external magnetic field can certainly give information useful for the elucidation of a number of fundamental questions in the theory of spin glasses.

## 3. EXPERIMENTAL TECHNIQUE AND SAMPLES

In the present work, as in Ref. 16, single-phase polycrystalline samples with a  $\text{Ga}^{3+}$ -ion content  $x = 1.5$ – $1.9$  were used.

The measurements of the SG properties such as the equilibrium magnetization  $\sigma_{FC}$  (the sample was first cooled from high temperatures ( $T > T_f$ ) to  $T = 4.2$  K with  $H \neq 0$ ) and the nonequilibrium magnetization  $\sigma_{ZFC}$ , and their temperature dependence  $\sigma_H(T)$  in small constant magnetic fields and their time dependence  $\sigma_{T,H}(t)$ , were carried out using a ballistic magnetometer. The sensitivity of the apparatus was  $10^{-10}$  T·m<sup>3</sup>/kg and the relative error was  $\pm 2\%$ . A magnetic field of intensity up to 7 kA/m was produced by a superconducting solenoid. The same apparatus was used to investigate the susceptibility in the paramagnetic region, i.e., in the range of temperatures from  $T_f$  to 220 K. Magnetic fields of up to 20 kA/m were produced by the superconducting solenoid there also.

## 4. EXPERIMENTAL RESULTS AND DISCUSSION

### 4.1. The magnetic viscosity; lines of critical behavior

For a more accurate determination of  $T_f$  than that in Ref. 16 (which used the position of the maximum on the curves of the dependences  $\sigma_{ZFC}(T)$  in small fields) in this work we studied the magnetic viscosity, i.e., the dependences of  $\sigma_{ZFC}$  and  $\sigma_{FC}$  on time for constant values of  $H$  and  $T$ . Such investigations are appropriate because sharp criteria for the determination of  $T_f$  are absent from the behavior of the dependences  $\sigma_{ZFC}(T)$  and  $\sigma_{FC}(T)$  in real spin glasses.<sup>23</sup> In Ref. 24, for example, it was shown that the positions of the maxima of the dependences  $\sigma_{ZFC}(T)$  may not correspond to  $T_f$ . The character of the irreversibility of real spin glasses, like that of the materials considered in the present paper, often differs from that predicted in MF theory.

As shown in Ref. 25, the appearance of long-time logarithmic viscosity at  $T = T_f$  can serve as a weightier indicator of a transition to the spin-glass state than the phenomenon of irreversibility. The magnetic viscosity observed in static experiments in the temperature range from 0 K to  $T_f$  corresponds to the predicted (in the dynamical theory of spin glasses) critical slowing down of the relaxation of the magnetic moments as  $T \rightarrow T_f^+$ , which exists in the entire low-temperature region  $T < T_f$  (Ref. 8). Thus, the appearance of long-time relaxation processes (magnetic viscosity) at a certain temperature  $T$  that depends on the concentration  $x$  of nonmagnetic ions and on the magnetic-field strength  $H$  is evidence of the presence of the PM-SG phase transition predicted by the dynamical mean-field theory.

To process the resulting experimental data we used the relation<sup>8</sup>

$$\sigma_{ZFC}(t) = \sigma_0 + S \ln(t/t_0), \quad (4)$$

where  $\sigma_{ZFC}(t)$  is the magnitude of the nonequilibrium magnetization at any time  $t$  after the magnetic field is switched off,  $\sigma_0 = \sigma_{ZFC}(t_0)$ ,  $t_0 \sim 10$  sec,  $t > t_0$ , and  $S$  is the magnetic-viscosity (magnetic-aftereffect coefficient):  $S = 0$  for  $T \geq T_f$ , and  $S > 0$  for  $T < T_f$ . The vanishing of the coefficient  $S$  as  $T \rightarrow T_f^-$  was used as a criterion for the determination of  $T_f$ .

A logarithmic magnetic viscosity, described by the law (4) and appearing at a sharply defined temperature  $T_f$  that

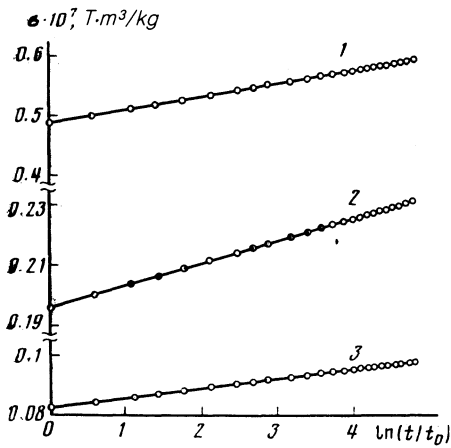


FIG. 1. Time dependence of the magnetization (on a semilogarithmic scale) in a field  $H = 4$  kA/m at temperature  $T = 0.4 T_f(H)$  for samples of  $\text{Li}_{0.5}\text{Fe}_{2.5-x}\text{Ga}_x\text{O}_4$  with: 1)  $x = 1.5$ , 2)  $x = 1.6$ , 3)  $x = 1.7$ .

depends on  $x$  and  $H$ , was observed for concentrations  $x$  of nonmagnetic  $\text{Ga}^{3+}$  ions in the range 1.5–1.9. Figure 1 shows how  $\sigma_{ZFC}$  depends on  $\ln(t/t_0)$  for samples with  $x = 1.5$ –1.7.

In the entire temperature range  $T \geq 4.2$  K samples with  $x > 1.9$  have superparamagnetic or paramagnetic properties; there is no magnetic viscosity in this case. Such concentration behavior agrees fully with the results of percolation theory for a cubic two-sublattice ferrimagnet with one kind of magnetically active ion, when diamagnetic dilution occurs in both sublattices.<sup>17</sup>

In the conditions realized in the experiments, for samples with  $x = 1.5$ –1.9 no changes of the magnetization  $\sigma_{FC}$  with time were observed, i.e., for the time scale under consideration  $\sigma_{FC}$  may be assumed to be the equilibrium magnetization, although here, in contrast to the results of MF theory,  $\sigma_{FC}$  for  $T < T_f$  depends on the temperature.<sup>16</sup>

The regular features of the behavior of the viscosity coefficient  $S$  with variation of the field and temperature, i.e., the functions  $S_{II}(T)$  and  $S_T(H)$ , are analogous to those for

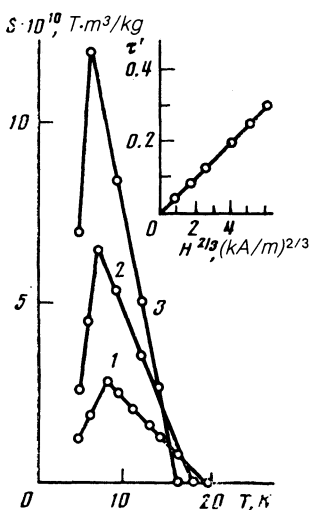


FIG. 2. Temperature dependence of the magnetic-viscosity coefficient  $S_{II}$  for a sample of  $\text{Li}_{0.5}\text{Fe}_{0.9}\text{Ga}_{1.0}\text{O}_4$  in fields  $H$  of: 1) 1.6 kA/m, 2) 4 kA/m, 3) 8 kA/m. Insert: field dependence of the reduced temperature  $\tau' = 1 - T_f(H)/T_f(0)$  for a sample of  $\text{Li}_{0.5}\text{Fe}_{0.9}\text{Ga}_{1.0}\text{O}_4$ .

classical spin glasses<sup>26</sup> and to those given in Ref. 27 for a sample with  $x = 1.5$ . For illustration, Fig. 2 shows  $S_{II}(T)$  in fields  $H = 1.6, 4$ , and 8 kA/m for a sample with  $x = 1.6$ . For all samples of the system investigated, and in the entire range of fields in which viscosity exists the maximum values of  $S_{II}$  are observed at temperatures  $T = 0.4 T_f(H)$ . In the table we give data obtained in a field  $H = 4$  kA/m. It can be seen that with increasing  $x$  the values of  $S$  are lowered, but even in the SG region ( $x > 1.5$ ) they are an order of magnitude greater than the corresponding values for classical spin glasses.<sup>26</sup> This fact can be regarded as support for existing ideas that in SG systems with short-range interaction the height of the energy barriers separating the valleys is finite.<sup>1,2</sup>

In the presence of a magnetic field the freezing temperatures  $T_f$  of all the samples examined decrease with increase of the field. The upper critical lines  $T_f(H)$  have three sharply defined parts with the same field boundaries. In weak fields ( $H \lesssim 24$  kA/m) the transition to the spin-glass state occurs along a line of the AT type ( $\tau' \propto H^{2/3}$ ), while in strong fields ( $H \gtrsim 40$  kA/m) it occurs along a line of the GT type ( $\tau' \propto H^2$ ); the region of intermediate fields corresponds to crossover. Behavior of this type is predicted in MF theory for Heisenberg spin glasses with random anisotropy.<sup>13</sup> The insert in Fig. 2 shows the low-field part of the critical line ( $\tau' \propto H^{2/3}$ ) for a sample with  $x = 1.6$ ; the question of the critical behavior and of the sources of the random anisotropy in the materials under investigation was considered in Ref. 28. The values  $T_f(0)$  of the freezing temperatures in the absence of a field were determined from the low-field parts of the critical lines, of the type shown in the insert in Fig. 2. The results obtained, which are listed in the table, coincide to within  $\pm 1$  K with the data in the  $x$ - $T$  diagram in Ref. 16, in which the values of  $T_f$  were determined from the positions of the maxima of the dependences  $\sigma_{ZFC}(T)$ .

Thus, investigation of the magnetic viscosity has shown that the dilute ferrimagnetic oxides  $\text{Li}_{0.5}\text{Fe}_{2.5-x}\text{Ga}_x\text{O}_4$  with  $x = 1.5$ –1.9 in the limit  $T \rightarrow T_f^+$  display critical behavior with the features predicted by MF theory for Heisenberg spin glasses with random anisotropy: There exists a PM-SG phase transition (in the dynamical sense) that is preserved in the presence of a magnetic field. In accordance with the theoretical conclusions described above,<sup>13</sup> this transition should be described by the one-component EA order parameter  $q_{EA}$ .

#### 4.2. Determination of the EA order parameter

To calculate the order parameter  $q_{EA}$  and its temperature dependence we used the relation<sup>22</sup>

$$q_{EA}(T) = 1 - T [C \chi_{ZFC}^{-1}(T) + \Theta]^{-1}. \quad (5)$$

The values of the Curie constant  $C$  and the paramagnetic Curie temperature  $\Theta$  were determined by the standard method from the temperature dependences of the paramagnetic susceptibility, which are presented in Fig. 3. The concentration dependences of  $C$  and  $\Theta$  are given in Fig. 4, and the values of the temperatures  $T^*(x)$  above which the Curie-Weiss law is fulfilled are given in the table.

The dependence of the order parameter  $q_{EA}$  on the reduced temperature  $T/T_f$  is presented in Fig. 5. It can be seen that for all the investigated samples exhibiting SG behavior, except the samples with  $x = 1.5$  (this composition corre-

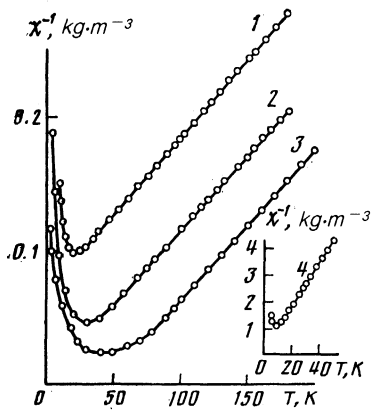


FIG. 3. Temperature dependence of the inverse paramagnetic susceptibility of samples of  $\text{Li}_{0.5}\text{Fe}_{2.5-x}\text{Ga}_x\text{O}_4$  with: 1)  $x = 1.6$ , 2)  $x = 1.55$ , 3)  $x = 1.5$ , 4)  $x = 1.9$ .

sponds to a multicritical point of the  $x$ - $T$  phase diagram<sup>16</sup>), the behavior  $q_{EA}(T)$  agrees with the theoretical predictions:  $q_{EA} = 0$  for  $T > T_f$ , and  $q_{EA} \rightarrow 1$  as  $T \rightarrow 0$  K.

The existence, for the materials under consideration, of an AT instability line ( $\tau' \propto H^{2/3}$ ) has made it possible, with the use of the relation (3), to analyze the critical behavior of  $q_{EA}(\tau)$ . For all cases the dependence  $q_{EA}(\tau)$  is linear up to  $\tau = 0.2$ , while for  $\tau > 0.2$  the main contribution to (5) is given by the quadratic term. The critical behavior of  $q_{EA}(\tau)$  is illustrated by the insert in Fig. 5, in which the behavior of the dependence  $q_{EA}(\tau)$  is shown on a log-log scale for a sample with  $x = 1.7$ . Analogous results were obtained in Ref. 22 in an analysis of the dynamics of concentrated metallic spin glasses and for classical RKKY spin glasses in the static regime.<sup>29</sup>

It is necessary to note that the  $EA$  order parameter  $q_{EA}(T)$  for the materials under consideration depends on time through the measurable quantity  $\chi_{ZFC}$ , as it should in accordance with the physical meaning of this quantity. The value of  $q_{EA}(T)$  decreases slowly with time, and  $T_f$  is lowered correspondingly. This result agrees with the conclusions of Ref. 30, reached on the basis of computer modeling for Ising spin glasses with short-range interaction.

Thus, the entire set of experimental results obtained is clear evidence of the existence in dilute ferrimagnetic oxides of a PM-SG phase transition in the  $EA$  order parameter. The question of the extent to which this is due to the presence of random anisotropy remains open. All that can be asserted is that, in the given case, we are certainly dealing with anisotropic Heisenberg spin glasses with critical behavior (in an external magnetic field) similar to the behavior of Ising spin glasses.

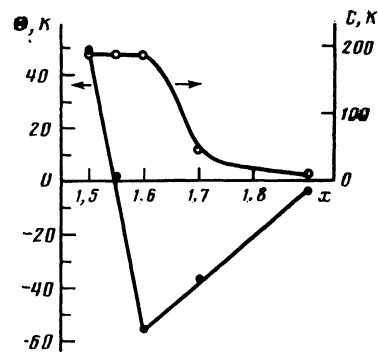


FIG. 4. Concentration dependences of the Curie constant  $C$  and paramagnetic Curie temperature  $\Theta$  of samples of  $\text{Li}_{0.5}\text{Fe}_{2.5-x}\text{Ga}_x\text{O}_4$ .

### 4.3. Structure of the SG state in dilute ferrimagnetic oxides

The existence, for finite observation times, of a PM-SG phase transition in the  $EA$  order parameter is assumed in all known SG models, which, at the same time, are based on different ideas about the structure of the SG state and predict both the general and the distinguishing features of the behavior.

The investigations performed have shown that dilute ferrimagnetic oxides are typical anisotropic Heisenberg SG systems with short-range interaction, which, together with general properties (magnetic viscosity and magnetic irreversibility), display certain properties that are specific for different models. As already noted, the critical behavior in a magnetic field (AT lines in small fields and GT lines in large fields), and also the possibility of describing the PM-SG transition by means of the one-component  $EA$  order parameter are completely consistent with the conclusions of MF theory for anisotropic vector spin glasses with an infinite-range interaction in which an individual spin is considered as the structural unit.<sup>13</sup> At the same time, the presence of temperature dependence of the equilibrium magnetization  $\sigma_{FC}$  and the character of the occurrence of the irreversible phenomena do not agree with the MF model. The latter, in particular, may be due to the cluster structure of the SG in dilute ferrimagnetic oxides<sup>16,17,27</sup> In addition, a number of other experimental data point unambiguously to the presence of clusters in the SG structure and to their real role in the formation of the properties of the spin-glass phase.

From the concentration dependence of the Curie constant (Fig. 4) it can be seen that only for the sample with  $x = 1.9$  does the value of  $C$  approach the theoretical value (equal to 4.38), while for  $x = 1.5$ -1.7 it is much greater. Thus, only for comparatively large diamagnetic dilution (80% or more of nonmagnetic ions) does the structure of the SG in the materials investigated approach a uniform

TABLE I. Dependence of parameters of samples of the system  $\text{Li}_{0.5}\text{Fe}_{2.5-x}\text{Ga}_x\text{O}_4$  on  $x$ .

Parameters	$x$				
	1.5	1.55	1.6	1.7	1.9
$S \cdot 10^{19}, \text{T} \cdot \text{m}^3/\text{kg}$	26	-	6.4	2.4	1.0
$T_f(0), \text{K}$	$37 \pm 1$	$32 \pm 1$	$22 \pm 1$	$14 \pm 1$	$7 \pm 1$
$T^*, \text{K}$	$80 \pm 2$	$40 \pm 2$	$26 \pm 2$	$20 \pm 2$	$12 \pm 2$

Note: The first row gives values of  $S_{\text{max}}$  for  $T = 0.4T_f(H)$  in a field  $H = 4 \text{ kA/m}$ .

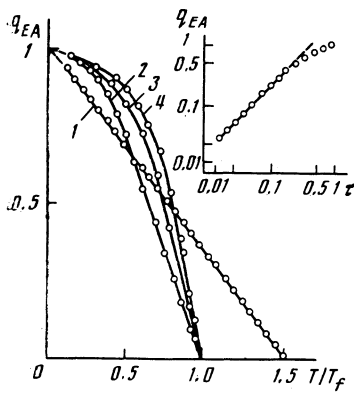


FIG. 5. Dependence of the order parameter  $q_{EA}$  on the reduced temperature  $T/T_f$  for samples of  $\text{Li}_{0.5}\text{Fe}_{2.5-x}\text{Ga}_x\text{O}_4$  with: 1)  $x = 1.5$ , 2)  $x = 1.55$ , 3)  $x = 1.6$ , 4)  $x = 1.7$  and 1.9. Inset: dependence of  $q_{EA}$  on  $\tau = 1 - T/T_f$  (on a double logarithmic scale) for a sample of  $\text{Li}_{0.5}\text{Fe}_{0.8}\text{Ga}_{1.7}\text{O}_4$ .

structure in which the structural unit is an individual spin.

On the basis of conclusions reached in the framework of the percolation model of Ref. 17, and of the results of neutron-diffraction investigations of compounds similar in nature to those under consideration,<sup>19,20</sup> we can characterize the SG state realized in dilute ferrimagnetic oxides as a non-uniform (cluster) state. It can be represented in the form of an SG matrix of the classical type, containing clusters that are uncorrelated in the direction of the frozen magnetic moments. The entire system (matrix and clusters) freezes at the same temperature  $T_f$ , whereas the ordering in the clusters is destroyed at much higher temperatures. The existence of exchange correlation between the clusters, as a result of which their magnetic moments freeze in arbitrary directions at  $T = T_f$ , is explicitly indicated by the high values of the paramagnetic Curie temperature  $\Theta$  (Fig. 4). Taking into account that the values of  $\Theta$ , like those of  $C$ , were obtained in the paramagnetic region comparatively close to  $T_f$  and in measurements in small fields, we arrive at the conclusion that they characterize a cluster subsystem. Since these "cluster characteristics" were also used in the calculation of  $q_{EA}(T)$ , it should be recognized that here both the individual spins and spin clusters appear as the structural units. Such a structure of the SG state, which follows directly from the experimental results, is practically entirely analogous to that proposed in the droplet model of Ref. 6, in which it was found that a number of spin-glass properties are due to the excitation of large clusters with low energy.

In conclusion, we shall discuss certain features of the concentration transition to the SG state that are directly intrinsic to the class of compounds under consideration and that are manifested in the dependence  $\Theta(x)$ . Here it is necessary to take into account that in ferrimagnets, despite the antiferromagnetic character of the inter-sublattice exchange,  $\Theta$  is always positive; in Li-Ga spinels all the  $\text{Fe}^{3+} - \text{O}^{2-} - \text{Fe}^{3+}$  exchange interactions (both inter- and intra-sublattice) are negative.<sup>31</sup> As can be seen from Fig. 4, the destruction of the long-range ferromagnetic order and the transition to the SG state correspond to a sudden change of the sign of  $\Theta$ : For practically equal values of  $x$ ,  $\Theta > 0$  for  $x = 1.5$  (the multicritical point) and  $\Theta < 0$  for  $x = 1.6$  (the spin glass). It is interesting to note that from a number of

indicators,<sup>32</sup> including the determination of the spontaneous magnetization by the kink-point method, it follows that for  $x \gtrsim 1.5$  we have  $\sigma_s = 0$ . Nevertheless, from the data obtained for  $\Theta$  it can be seen that here, i.e., for  $x = 1.5$  and 1.55, the distinctive "memory" of the long-range ferrimagnetic order is still preserved ( $\Theta > 0$ ), i.e., for concentrations very close to the multicritical point (on the SG side) a certain disordered state intermediate between a ferrimagnetic spin glass ( $x < 1.5$ ) and a cluster spin glass ( $x > 1.5$ ) is realized. In this region we still have comparatively high positive values of  $\Theta$  for  $\sigma_s = 0$ .

## 5. CONCLUSION

Our investigations have shown that in the system of dilute ferrimagnetic oxides  $\text{Li}_{0.5}\text{Fe}_{2.5-x}\text{Ga}_x\text{O}_4$  with short-range antiferromagnetic indirect exchange a PM-SG phase transition in the EA order parameter  $q_{EA}$  exists for  $1.5 < x \leq 1.9$  and is preserved in the presence of a magnetic field. Although the materials under consideration are Heisenberg spin glasses, on account of the presence of random anisotropy they exhibit features of Ising spin glasses: The upper critical line in small fields has the form of an AT line ( $\tau' \propto H^{2/3}$ ) while in large fields it has the form of a GT line ( $\tau' \propto H^2$ ), and the PM-SG transition is described by a one-component order parameter displaying critical behavior near the AT instability line:

$$q_{EA}(T) = 1 - T/T_f.$$

The transition to the SG state is manifested in the onset of long-time logarithmic viscosity at the freezing temperature  $T_f$  ( $T \rightarrow T_f^+$ ) and in sharply pronounced irreversible phenomena at  $T < T_f$ . Taken together, all these facts point to the existence of a PM-SG phase transition in the dynamical sense and agree with the ideas developed in MF theory concerning features of this transition as a transition to a degenerate nonergodic state whose energy spectrum has the form of valleys separated by barriers. In view of this, it may be considered that the EA parameter  $q_{EA}$  determined experimentally in this work has the meaning of the one-valley order parameter. At the same time, inasmuch as the nonergodicity is small near  $T_f$  ( $T \rightarrow T_f^-$ ), the presence of a PM-SG transition in the parameter  $q_{EA}$  indicates, in essence, the possibility of a PM-SG transition in the Parisi order parameter or Sompolinsky order parameter.

Despite the fact that the results obtained agree quite fully with the conclusions of MF theory with an infinite-range interaction, in which the structural unit of the SG is taken to be an individual spin, a number of experimental data clearly demonstrate the role of short-range exchange interaction in the formation of the SG state and point to the realization of an SG state of the cluster type predicted by the droplet model. Here, the role of the structural units is played by both individual spins and spin clusters, and the entire system, consisting of the matrix (the set of individual spins) and the clusters, freezes at the same temperature  $T_f$ .

Thus, in the dilute ferrimagnetic oxides  $\text{Li}_{0.5}\text{Fe}_{2.5-x}\text{Ga}_x\text{O}_4$  ( $x = 1.5-1.9$ ), i.e., disordered Heisenberg systems with short-range exchange and random anisotropy, at low temperatures a cluster-SG state with an Ising type of critical behavior in a magnetic field is realized; the PM-SG transition is a transition to a degenerate nonergodic

state, and the one-valley order parameter  $q_{EA}$  depends on time and, at temperatures up to  $0.2T_f$ , displays critical behavior.

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Translated by P. J. Shepherd