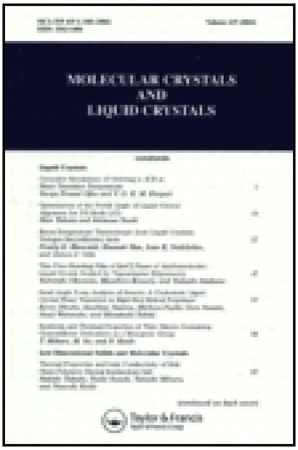

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Magnetic Double Exchange Interaction as a Driving Force of the Coexistence of the Negative Giant Magnetoresistance and the Spin Glass State in the Selected Re-Manganites and the Spinels with Chromium

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Magnetic Double Exchange Interaction as a Driving Force of the Coexistence of the Negative Giant Magnetoresistance and the Spin Glass State in the Selected Re-Manganites and the Spinels with Chromium

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In this article is pointed out the particularly important role of the double exchange magnetic interaction in the appearance of the phenomenon of the coexistence of the negative giant magnetoresistance and the spin glass state in the selected RE-manganites and the spinels with chromium. To explain this phenomenon, its statistical background and the types of magnetic interactions in the compounds under study are taken into account. The particular importance of the double exchange magnetic interaction "cooperating" with the strong external magnetic induction results here from that: i. its coupling constant is several times greater than the superexchange one, ii. the spin orientation of the hopping electrons is conserved (always ferromagnetic coupling!) and the charge transfer makes the p-type metallic conductance.

Keywords: double exchange; magnetic interactions; magnetoresistance; RE-manganites; spin glass; spinels with chromium

1. INTRODUCTION

The magnetization, dc and ac magnetic susceptibilities, resistance and magnetoresistance of the $Sm_{1-x}Sr_xMnO_3$ ($x = 0.33$, 0.45) and

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$RE_{0.55}Sr_{0.45}MnO_3$($RE_{0.55} = Eu_{0.397}Nd_{0.153}$, $Tb_{0.25}Nd_{0.30}$) manganites [1–7] as well as the $CuCr_2X_4$ (X = S, Se) spinels doped with Sb [8–15] have been studied at the high magnetic stationary fields up to 14 T, at the low magnetic alternating fields up to 0.1 T and in the temperature range of 4.2 ÷ 300 K. It turned out that the saturation magnetization has been reached in the manganites under study below the temperature of liquid nitrogen at the magnetic fields less than 3 T. Moreover in these compounds metamagnetic phase transitions occur, e.g. spin-flip and spin-flop in $Eu_{0.40}Nd_{0.15}Sr_{0.45}MnO_3$ and spin-flip in $Tb_{0.25}Nd_{0.30}Sr_{0.45}MnO_3$ [7]. On the other hand the spinels under study magnetize very hard and do not reach the magnetic saturation even at 14 T and 4.2 K, though at room temperature they exhibit a spontaneous magnetization. Also here the metamagnetic phase transitions take place, e.g. in $CuCr_{1.6}Sb_{0.4}S_4$ spin-flop phase transitions occur with several changes of the speed of the spin-flop process [9]. For both the manganites and the spinels under study a difference between ZFC and FC dc susceptibilities takes place [4,9,10,13]. Moreover, the real part of the 2nd harmonic of the ac susceptibility is negative above the spin freezing temperature and below the T_C [e.g. 10]. For all the manganites under study metal-semiconductor phase transitions and the negative giant magnetoresistances down to -100% have been observed [6,7]. In the case of the spinels under study containing sulphur, which are stoichiometric, the giant magnetoresistance is always negative and reaches even -74% at the 38 T for the antimony concentration of 0.4 [9]. On the other hand for the spinels under study containing selenium, which are nonstoichiometric, the magnetoresistance reveals much more dramatic behavior. Especially in the case $Cu_{0.99}Cr_{1.73}Sb_{0.36}Se_{4.05}$ [14] the magnetoresistance changes both the sign and its colossal values from -4433% at 153.2 K up to $+980\%$ at 154.2 K [14]. From what was written above and in the references cited it follows that both the behavior of magnetization and of susceptibility point to the coexistence of the giant magnetoresistance and the spin glass states.

The aim of this article is to show the particularly important role of the double exchange magnetic interaction in the compounds under study.

2. TYPES OF MAGNETIC INTERACTIONS IN THE COMPOUNDS UNDER STUDY

The interrelation of the chemical bonds, of either the localization or delocalization of the magnetic moments and of the types of the electrical conductivity resulting from them is presented for all the magnetic compounds in the Herring's phase diagram [16]. The areas marked in this diagram in the form of circles correspond to different magnetic couplings following from this interrelation. The compounds under study can be placed in the Herring's diagram in the areas containing the direct exchange, the superexchange and the RKKY exchange. The direct exchange presents the Heisenberg-type interaction between the nearest magnetic cations, whereas the magnetic coupling taking place via the nonmagnetic anions is called superexchange. The superexchange has two mechanisms of the magnetic interaction: 1. the Kramers-Anderson superexchange consisting in the overlaps of the magnetic cation orbitals via the nonmagnetic anion orbitals (between the latter nonmagnetic cation orbitals can also appear), 2. the double exchange consisting in the hopping of electrons between the magnetic cations with the different valence. The possible magnetic interaction paths in the case of the Kramers-Anderson superexchange are the following: B-X-B, B-X-X-B and B-X-A-X-B, where X are anions whereas A and B are cations in the tetra- and octahedral positions, respectively. The magnetic interactions along these paths are related to the overlap of wave functions of both p-orbitals of anions and 3d orbitals of cations. The different types of the orbital overlaps can here occur [17]: i. axial overlap creating so-called σ band, ii. nonaxial overlap creating so-called π band and iii. anion overlap creating so-called Δ band. Along the paths mentioned above the following couplings can appear: $\sigma-\sigma$, $\pi-\pi$, $\sigma-\pi$, $\sigma-\Delta-\sigma$, $\pi-\Delta-\pi$, $\sigma-\Delta-\pi$. These couplings can lead to either ferro- or antiferromagnetic orderings, the latter "choice" of Nature being described by the Goodenough-Kanamori rules [18].

One has to point out that the double exchange can also take place along the possible magnetic interaction paths mentioned above. The necessary condition for this interaction to occur is the appearance in these paths of the chemically identical magnetic ions with the mixed valence. One can distinguish two types of the double exchange interactions, namely

a) the first-order process invoked by Zener [19–21]:

$$\begin{array}{ccccccc} B^{4+} & X^{2-} & B^{3+} & \rightarrow & B^{3+} & X^{2-} & B^{4+} \\ & 2\uparrow, 3\downarrow & 1\uparrow & & 2\uparrow & 1\uparrow, 3\downarrow & \end{array}$$

b) the second-order process of Anderson and Hasegawa [20–22]:

$$B^{4+}\quad X^{2-}\quad B^{3+}\ \rightarrow\ B^{3+}\quad X^{4-}\quad B^{2+}\ \rightarrow\ B^{3+}\quad X^{2-}\quad B^{4+}$$

$$2\uparrow, 3\downarrow\quad 1\uparrow\qquad 2\uparrow\quad 3\downarrow\quad 1\uparrow\qquad 2\uparrow\quad 1\uparrow, 3\downarrow$$

$$B^{4+}\quad X^{2-}\quad B^{3+}\ \rightarrow\ B^{3+}\quad X^{4-}\quad B^{2+}\ \rightarrow\ B^{3+}\quad X^{2-}\quad B^{4+}$$

$$2\uparrow, 3\downarrow\ 1\uparrow\qquad 2\uparrow\qquad 1\uparrow, 3\downarrow\qquad 2\uparrow\ 1\uparrow, 3\downarrow$$

3. SPIN GLASS STATE AND GIANT MAGNETORESISTANCE

The key feature of all the compounds under study is the coexistence of the spin glass state and the giant magnetoresistance. This effect is illustrated for selected compounds in Figures 1–7, namely: for $Tb_{0.25}Nd_{0.30}Sr_{0.45}MnO_3$ in Figures 1 and 2, for $CuCr_{1.6}Sb_{0.4}S_4$ in Figures 3 and 4, for $Cu_{0.99}Cr_{1.73}Sb_{0.36}Se_{4.05}$ in Figures 5 to 7. Figure 1 presents the temperature dependence of the ZFC and FC magnetization. The ZFC&FC hysteresis below 100 K testifies to the existence of the spin glass state; the data for this diagram are taken from [4]. Figure 2 presents two isotherms of the magnetoresistance vs. external

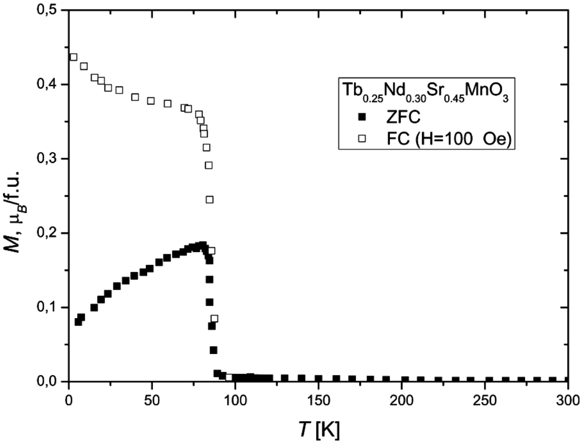

FIGURE 1 ZFC and FC temperature dependence of magnetization for $Tb_{0.25}Nd_{0.30}Sr_{0.45}MnO_3$. The data for this diagram are taken from [4].

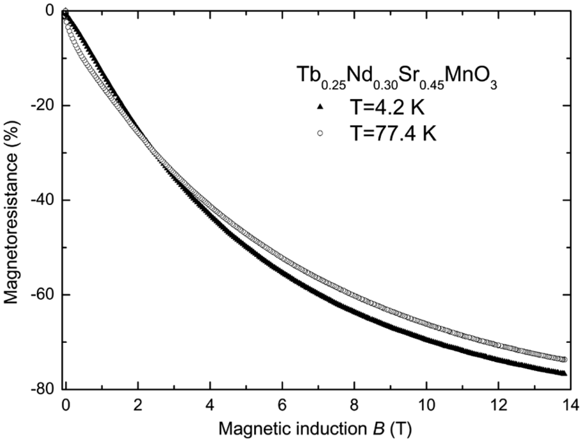

FIGURE 2 Magnetoresistance versus magnetic induction at 4.2 K and 77.4 K for the sample of $Tb_{0.25}Nd_{0.30}Sr_{0.45}MnO_3$. The data for this diagram are taken from [7].

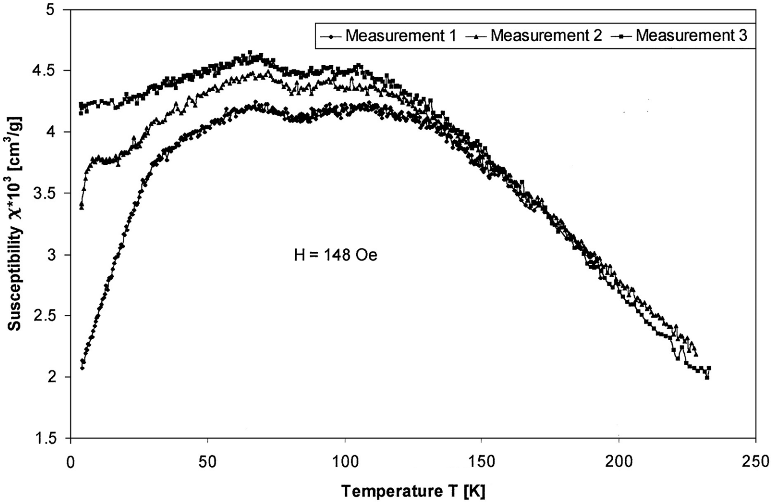

FIGURE 3 Temperature dependence of the magnetic susceptibility taken at 148 Oe for $CuCr_{1.6}Sb_{0.4}S_4$. All three measurements were carried out after one another and when heating. For measurement 1 the heating speed was 0.25 K/min, whereas for measurements 2 and 3 the heating speed was 3 K/min. The cooling after measurements 1 and 2 was carried out at the zero magnetic field [9].

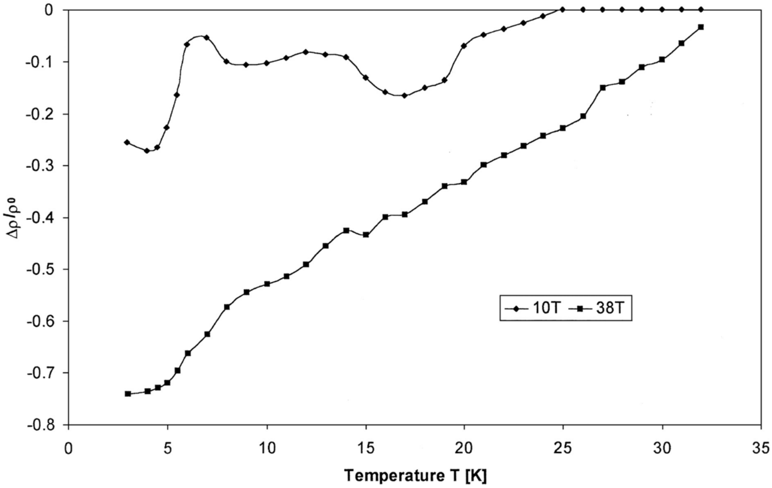

FIGURE 4 Temperature dependence of the magnetoresistance for two different values of magnetic induction for $CuCr_{1.6}Sb_{0.4}S_4$ [9].

magnetic induction taken at 4.2 K and 77.4 K (which is below 100 K!). From these isotherms it follows that these magnetoresistances are giant for higher values of the magnetic induction reaching their greatest absolute values of about 75% at 14 T; the data for this figure are

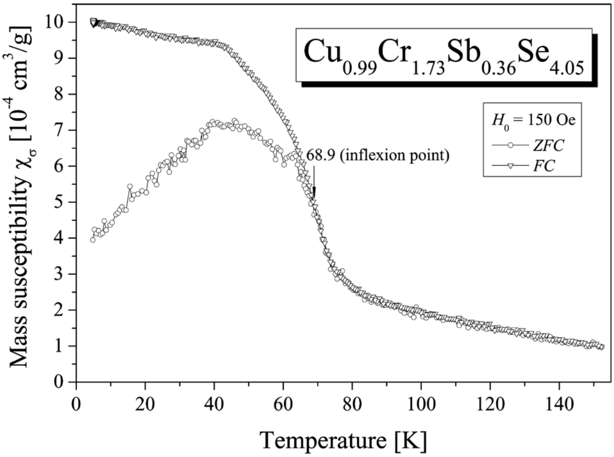

FIGURE 5 ZFC and FC temperature dependence of dc susceptibility of $Cu_{0.99}Cr_{1.73}Sb_{0.36}Se_{4.05}$.

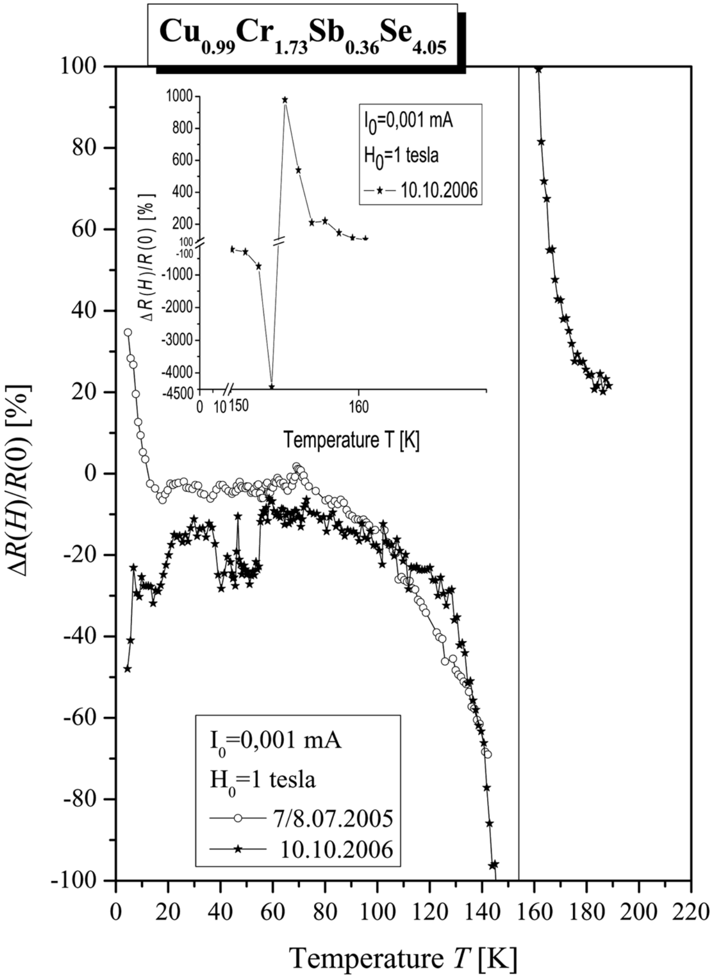

FIGURE 6 $Cu_{0.99}Cr_{1.73}Sb_{0.36}Se_{4.05}$. Temperature dependence of magnetoresistance at 10 kOe [14].

taken from Figures 5 and 6 in [7]. Figure 3 presents the temperature dependence of the magnetic susceptibility taken at 148 Oe. All the three measurements presented in this figure were carried out after one another and when heating. For the measurement 1 the heating speed was 0.25 K/min, whereas for the measurements 2 and 3 the heating speed was 3 K/min. The cooling after the measurements 1 and 2 was carried out at the zero magnetic field [9]. One can easy see in this figure that instead of the sharp peak a wide maximum

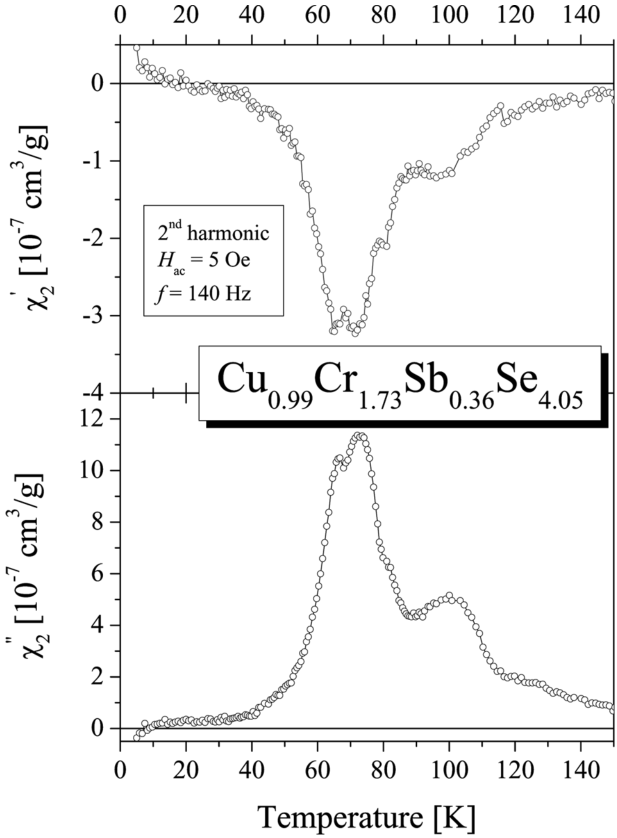

FIGURE 7 Real and imaginary parts of the second harmonic of the ac magnetic susceptibility for the $Cu_{0.99}Cr_{1.73}Sb_{0.36}Se_{4.05}$.

appears extended over a large temperature range. Moreover the three curves presented in this figure turned out to be different which means that the susceptibility of this sample is sensitive to its thermic history. One has to admit that also the essential difference of the ZFC and FC magnetization isotherms has been here observed [9]. All these three effects point to a spin glass state adopted by the sample at low temperatures. Figure 4 presents the temperature dependence of magnetoresistance for two different values of magnetic induction [9]. Note that the temperature range of the existence of the giant magnetoresistance revealed in this figure is contained in the temperature range of the existence of the spin glass state shown in Figure 3. Figure 5 shows the temperature dependence of the ZFC and FC dc susceptibility. The ZFC&FC hysteresis below 70 K is easily seen in this figure.

The temperature dependence of magnetoresistance at 10 kOe is shown in Figure 6 [14]. One can see in this figure that indeed below 70 K a giant magnetoresistance exists revealing the hysteresis in the range between +40% and −60%, the latter effect pointing simultaneously by the way to the existence of spin glass state. The primary and the secondary curves of this hysteresis coincide at about 140 K. This hysteresis seems to be related to both the thermal and the time (aging) effects which influence no doubts the reconstruction of the spin glass clusters. This in turn influences the spin glass state under consideration. Above 140 K the unique curve tends to very low values, where – as it was mentioned in the Introduction – a jump-like phase transition was observed changing both the sign and the measured value of the magnetoresistance from −4433% at 153.2 K up to +980% at 154.2 K. In Figure 7 real and imaginary parts are presented of the 2nd harmonic of the ac magnetic susceptibility. Here the real part is negative which testifies very strongly to the existence of the spin glass state [15,23,24] in this sample together with the ZFC&FC thermal hystereses mentioned above.

These effects are related to the double exchange magnetic interaction taking place in these crystals which in turn is related to the mixed valence of the magnetic ions. In the RE-manganites under study it is $\mathrm{Mn}^{3+}-\mathrm{Mn}^{4+}$ mixed valence [1–7] which makes the base of this interaction, whereas in the spinels with chromium this effect concerns the analogous $\mathrm{Cr}^{3+}-\mathrm{Cr}^{4+}$ mixed valence [8–15].

4. STATISTICAL BACKGROUND OF THE COEXISTENCE OF THE SPIN GLASS STATE AND THE NEGATIVE GIANT MAGNETORESISTANCE

Spin glass state model for the case of random distribution of dopants and defects below the percolation threshold in the ferromagnetic matrix has been proposed by the four of us [25].

In this case the Hamiltonian assumes the following form:

$$\widehat{H} = -\sum_{j\neq i}\sum_{i=1}^{N_j-n_j} J_{ij}\widehat{S}_i\cdot\widehat{S}_j - \sum_{j\neq i}\sum_{i=1}^{n_j} J'_{ij}p(i,j)\widehat{S}_i\cdot\widehat{S}_j, \tag{1}$$

where: N_j is a number of magnetic ions surrounding a j-th magnetic ion and interacting with it with a coupling constant J_{ij}, n_j is a number of dopants (or defects) surrounding a j-th ion, J'_{ij}-coupling constant between i-th and j-th ion in the presence of a dopant (or a defect), $p(i,j)$ is probability of appearance of a dopant (or a defect) between i-th and j-th ions [25].

A quantum-mechanical equation for magnetization **M** has been derived and solved after linearization [25]. This equation contains a. o. the following expression:

$$\widetilde{J}(-\mathbf{q}') = \sum_{j\neq i} \sum_{i=1}^{N_j-n_j} J_{ij} e^{i\mathbf{q}'(\mathbf{R}_i-\mathbf{R_j})} + \sum_{j\neq i} \sum_{i=1}^{n_j} J'_{ij} p(i,j) e^{i\mathbf{q}'(\mathbf{R}_i-\mathbf{R_j})} \tag{2}$$

where $\mathbf{R}_i$, $\mathbf{R}_j$ – the radius-vectors of the i-th and j-th magnetic ions, respectively.

Note that the random distribution of either dopants or defects $p(i,j)$ leads to the random distribution of the magnetic coupling constants p(J'_{ij}) and – referring to the Anderson spin glass state model (e.g. [26]) – one comes to the conclusion that the latter effect causes the frustration of the magnetic couplings and the appearance of the spin-glass state in the ferromagnetic system under consideration containing either dopants or defects.

The above reasoning is confirmed by Eq. (2), because it expresses – as a Fourier transform – a kind of a “global” magnetic coupling constant $\widetilde{J}(-\mathbf{q}')$, which turns out to be random, the latter characteristic following at least from the dependence of the second term of Eq. (2) on $p(i,j)$. Moreover, the randomness of $\widetilde{J}(-\mathbf{q}')$ seems to be of a Gaussian type, because the random quantity $\widetilde{J}(-\mathbf{q}')$ is expressed in Eq. (2) as a sum of many different random quantities, the latter effect making typical conditions of the functioning of the central limit theorem of the theory of probability, i.e. the Lyapunov theorem. As it follows from the linearized quantum-mechanical equation for magnetization **M** mentioned above and from Eq. (2), also the magnetization in the spin-glass state becomes a random quantity, which obtains the Gaussian-like distribution, too (the Lyapunov theorem again applies) [25]. In other words introducing either defects or dopants changes the relation of strengths of all the coupling constants acting between the magnetic atoms. In the case of the equalization of strengths of all the different types of the coupling constants acting in the crystal a frustration of the spin orientation appears bringing to the spin glass states. In the compounds under study the energetic equilibrium of the spin glass state consists in the creation of the ferromagnetic and antiferromagnetic clusters in the crystal. On the other hand the electrical conductance in the presence of the external magnetic induction is related to the reconstruction of these clusters in two cases, namely: i. if with the increase of the external magnetic induction the volume of the ferromagnetic clusters increases at the cost of the antiferromagnetic ones, then one deals with the appearance of the negative giant magnetoresistance, ii. in the reverse situation the positive giant magnetoresistance appears [7,9]. Note that ascribing a representative magnetization vector to every cluster one can say – according to what was said above – that the orientation of these magnetization vectors undergoes the Gaussian statistics, too. This effect is accompanied by the significant changes of conductance in the sample leading to either the negative or the positive giant magnetoresistance. The mechanism of these changes concerns the conductance bridges whose number rises in the ferromagnetic clusters and declines in the antiferromagnetic ones, both in the case of the volume increase of the ferromagnetic clusters and the volume decrease of the antiferromagnetic ones. In the reverse situation the insulating bridges tend to dominate. It is obvious that the statistics of both these kinds of bridges makes the replica of the statistics of the orientation of the magnetization vectors mentioned above.

5. PARTICULAR IMPORTANCE OF THE DOUBLE EXCHANGE MAGNETIC INTERACTION IN THE COMPOUNDS UNDER STUDY

As mentioned above, in all the compounds under study the structure of the spin glass states consists of the two types of clusters, namely ferromagnetic with the domination of the double exchange interaction accompanied by the p-type metallic conductance and the antiferromagnetic with the superexchange interaction only accompanied by the insulating properties [9,27,28]. Therefore with the expansion of volume of the ferromagnetic clusters at the cost of antiferromagnetic ones taking place in the strong external magnetic induction the p-type metallic conductance increases significantly up to its percolation to the whole crystal. This effect causes in turn the appearance of the negative giant magnetoresistance and its coexistence with this cluster spin glass state. It is the double exchange interaction which makes the necessary condition for this coexistence to appear.

Thus the particular importance of the double exchange magnetic interaction "cooperating" with the strong external magnetic induction in the compounds under study results from that:

1. Its coupling constant is several times greater than the superexchange one.
2. The spin orientation of the hopping electrons is conserved (always ferromagnetic coupling!).
3. The charge transfer makes the p-type metallic conductance.

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