NEUTRON DIFFRACTION, MAGNETIZATION AND ESR STUDIES OF PSEUDOCUBIC Nd$_{0.75}$Ba$_{0.25}$MnO$_3$ AND ITS UNUSUAL CRITICAL BEHAVIOR ABOVE $T_C$

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Abstract

Results of structural neutron diffraction study, magnetization and electron spin resonance (ESR) measurements are presented for insulating Nd$_{1-x}$Ba$_x$MnO$_3$ ($x = 0.25$) with the Curie temperature $T_C \approx 129$ K. Its pseudocubic structure reveals the definite distortions to a lower symmetry. Detailed analysis of the data is performed by using $Pbnm$ space group in a temperature range $4.2 \div 300$ K. The compound is found to exhibit the Jahn-Teller (JT) transition at $T_{JT} \approx 250$ K. Character of the coherent JT distortions and their temperature evolution differ from those of the $x = 0.23$ manganite. The field cooled magnetization data are in a reasonable agreement with the predictions for a 3D isotropic ferromagnet above $T_C$. These measurements, however, reveal a difference between the field cooled and zero field cooled data in the paramagnetic region. The ESR results are also in a correspondence with behavior of a 3D isotropic ferromagnet above $T^* \approx 143$ K ($\tau^* \approx 0.12 \leq \tau < 1, \tau = (T - T_C)/T_C$). It is shown that an anisotropic exchange coupling of the Mn and Nd magnetic moments can give a substantial contribution in ESR linewidth masking its critical enhancement. The different temperature treatments (slow/fast cooling/heating, with/without external magnetic field) of the sample reveal a temperature hysteresis of the ESR spectra below $T^*$ indicating an anomalous response in the paramagnetic region. The study of the magnetic phase transition in the $x = 0.23$ and 0.25 manganites suggests change in its character from the second to first order at $T^*$. The conventional free energy including the magnetization and magnetic field is not found to describe this first order transition. This suggests that the charge, orbital and JT phonon degrees of freedom, in addition to magnetization, may be the critical variables, the unusual character of the transition being determined by their coupling. The unconventional critical behavior is attributed to an orbital liquid metallic phase that begins to coexist with the initial orbital ordered phase below $T^*$. 
Doped manganites \( \text{L}_{1-x}\text{A}_x\text{MnO}_3 \) (\( \text{L} \) and \( \text{A} \) are rare earth and alkaline earth ions) usually exhibits the ferromagnetic (F) ground state in a region \( x_0 \leq x \leq 0.5 \). In \( \text{Nd}_{1-x}\text{Ba}_x\text{MnO}_3 \), this state develops below \( T_C \sim 120 \text{ K} \) for \( 0.2 \leq x \leq 0.35 \) [1]. For \( x < 0.3 \), these compounds show insulating (I) behavior in the ordered state, whereas for \( x \geq 0.3 \) the F metallic (M) state is observed below \( T_C \). A colossal magnetoresistance occurs for all \( x \) near \( T_C \). The unexpected coexistence of ferromagnetic and insulating behavior cannot be explained by the double exchange mechanism and remains to be poor understood. A possible reason of the charge localization can be a long-range orbital ordering due to Jahn-Teller (JT) effect. This source of the localization was suggested to account for the FI ground state in \( \text{La}_{1-x}\text{Ca}_x\text{MnO}_3 \) for \( 0.12 < x < 0.2 \) [2]. The FI state can also be consequence of a spontaneous orbital ordering, which develops in a cubic manganite, due to a coupling of the spin and orbital degrees of freedom arising from the electron correlations [3]. \( \text{La}_{0.88}\text{Sr}_{0.12}\text{MnO}_3 \) was found to exhibit the FI state of such nature [4]. Note that a new explanation of the insulator to metal transition, which occurs in an orbital disordered state, has been suggested recently [5]. This approach permits the existence an orbital liquid insulating state. As far as we know, however, such a state has not been found so far.

An important aspect of this problem is a nature of the paramagnetic (P) – FI transition which can differ from that of a 3D isotropic ferromagnet. A few works have only concentrated on behavior at this critical point. An unconventional P-FI transition was observed in insulating \( \text{La}_{0.8}\text{Ca}_{0.2}\text{MnO}_3 \) where a magnetic correlation length increased only from 10 to 35 \( \text{Å} \) as \( T \rightarrow T_C \) [6]. To investigate this issue in detail, we have used \( \text{Nd}_{1-x}\text{Ba}_x\text{MnO}_3 \) compounds, which are closed to the cubic perovskites due to the small structural distortions [7]. Also of interest in this system is the effect of a Mn-Nd interaction on its properties. In \( \text{Nd}_{0.7}\text{Ba}_{0.3}\text{MnO}_3 \), the ordering of the Nd moments, that develops below 20 K [7], changed the magnetic and transport properties [8]. As study of the \( \text{Nd}_{1-x}\text{Sr}_x\text{MnO}_3 \) (0.1 \( \leq x \leq 0.5 \)) showed, the Nd magnetic subsystem came also into play at the low temperatures due to its ordering [9]. At the same time, no effects of the Mn-Nd coupling were found near \( T_C \) in the investigations of critical static behavior of \( \text{Nd}_{0.6}\text{Pb}_{0.4}\text{MnO}_3 \) [10] and critical dynamics of \( \text{Nd}_{1-x}\text{Sr}_x\text{MnO}_3 \) (\( x \approx 0.5 \)) [11]. Our studies have included the measurements of the linear and nonlinear susceptibilities as well as ESR. The neutron diffraction investigations have been also performed to elucidate the effects of the lattice distortions and orbital ordering. The study of the critical magnetic properties of the \( x = 0.23 \) (\( T_C \approx 124 \text{ K} \)) and \( x = 0.25 \) (\( T_C \approx 129 \text{ K} \)) manganites revealed a more complicated scenario than that expected for the 3D isotropic ferromagnets [12, 13, 14, 15]. The conventional critical behavior was observed to proceed for \( T^* < T < 2T_C \) where \( T^* \approx 147 \text{ K} \) (\( x = 0.23 \)) and \( T^* \approx 143 \text{ K} \) (\( x = 0.25 \)). In contrast, the data on nonlinear response of the second order clearly indicated the occurrence of an anomalous critical behavior for \( T_C < T < T^* \) that suggested the coexistence of the two magnetic phases. Although the \( x = 0.23 \) compound exhibited an orbital ordering due to the JT effect below \( T_{JT} \sim 350\text{K} \), the neutron diffraction study did not reveal a two phase structure in the anomalous \( T \)–range [14] that would be a natural explanation of the phase separated (PS) magnetic state. In addition, according to the ESR measurements, the transition did not become hysteretic in temperature and clearly first order.

In this paper we present the neutron diffraction, magnetization and ESR investigations of \( x = 0.25 \) manganite. The \( T_{JT} \) is reduced on doping, becoming \( T_{JT} \approx 250 \text{ K} \). A character of the JT distortions and orbital ordering differs from that of the \( x = 0.23 \) compound. \( T^* \)}
The evolution of the structural parameters does not show any anomalies that can be definitely attributed to formation of a structural PS state in the anomalous $T$-range. The magnetization, $M(H)$, measurements are performed in the field cooled (FC) and zero field cooled (ZFC) regimes at $H = 1$ kOe in a range $4.2 \div 300$ K. The FC data agree reasonably with prediction for a 3D isotropic ferromagnet in the paramagnetic critical region from $2T_C$ down to $T_C$. At the same time, a difference between the FC and ZFC results is unexpectedly found to proceed up to 300 K. ESR measurements are carried out above $T_C$. A relaxation rate of uniform magnetization $\Gamma$ and $g$-factor show nearly the same dependencies above $T_C$ as for the $x = 0.23$ manganite. The $\Gamma(\tau)$ does not exhibit a critical enhancement as it occurs in the conventional cubic ferromagnets CdCr$_2$S$_4$ and CdCr$_2$Se$_4$ [16], as well as in the near to cubic manganites La$_{1-x}$Ca$_x$MnO$_3$ ($x = 0.18 \div 0.22$ [17]. An anisotropic exchange Mn-Nd coupling is shown can give a substantial contribution in $\Gamma$, masking the critical enhancement. In order to examine character of the phase transition, effect of the different temperature treatments of the sample on the ESR signals is elucidated. These measurements and $M(H)$ data support the existence of the magnetic PS state near $T_C$. The possible reasons of origination of the heterophase magnetic state are discussed.

II. SAMPLES AND METHODS

The polycrystalline samples were prepared by the standard technology [1], an oxygen content being controlled by a thermogravimetric analysis. A 48-counter powder neutron diffractometer of PNPI with a wave length 1.83 Å of an incident neutron beam was used in the structural investigations. The measurements were performed in a cryostat at $4.2 \div 300$ K. Program FULLPROF was employed for the structure refinement.

The magnetization measurements were carried out using a SQUID magnetometer in a temperature range $4.2 \div 300$ K and a magnetic field 1 kOe. A single crystal with a mass $m = 91.8$ mg was used that came from the same growth batch as the crystal of work [15]. In ESR study, the small part of single crystal investigated in work [15] with a mass $m \approx 1.6$ mg was used. We employed a special X-band ESR spectrometer ($f = 8.37$ GHz) described earlier [18, 19]. It registers a component of magnetization of a sample which is proportional to an off-diagonal part of its magnetic susceptibility ($M_y(\omega) = \chi_{yx}(\omega) h_x(\omega)$) when a linear polarized exciting ac-field $\mathbf{h} \parallel Ox$ and a steady field $\mathbf{H} \parallel Oz$.

Below $T^* \approx 143$ K the nonlinear response of the compound reveals features of the PS magnetic state that can signal the first order transition [15]. It is natural, therefore, to expect a temperature hysteresis of the ESR signal. Besides, a state forming at the PS usually depends on a process that is used to transfer a system in the PS regime. Therefore, the several types of the temperature scan without/with an external magnetic field were performed.

1. The sample was slowly cooled from room temperature down to $128$ K below $T_C$ and then slowly heated back to room temperature. In the chosen temperature points ($T-$points) the ESR spectra were registered. A temporal stabilization time before recording a signal was about 200-300 seconds at each $T-$points.

2. The same temperature scan as above was applied but with $H = 4$ kOe during cooling between $T-$points. This procedure also tests a possible PS state because the magnetic field can change a balance between the phase fractions.
3. A fast cooling of the sample was used down to 128 K in zero $H$ and then the sample was slow heated up to room temperature.

III. STRUCTURAL DATA

A neutron diffraction pattern at room temperature is plotted in Fig.1. Similar to Nd$_{0.77}$Ba$_{0.23}$MnO$_3$ crystal [14], pseudocubic structure of Nd$_{0.75}$Ba$_{0.25}$MnO$_3$ exhibits distortions to a monoclinic symmetry, as the characteristic peaks at the low angles of scattering evidence (see the insert in Fig.1). They unambiguously originate from a lattice deformation because the X-ray diffraction patterns also show them at room temperature. A width of the peaks is larger than that of the main Bragg reflections. It indicates that the regions of the relatively small sizes possess the monoclinic distortions. The peaks exhibit no temperature variations and have a small amplitude. The presence of small amount of such a monoclinic phase is expected to have a minimal effect on the $T$–dependent properties of this compound.

The measured diffraction profiles are found to be well fitted using the orthorhombic Pbnm space group. This fit gives only the slightly worse description than that of the monoclinic group $P2_1/m$. Therefore, the further analysis will be based on the Pbnm setting. Fig.2 displays the $T$–variations of structural parameters. The orthorhombic distortions are small in range of the measurements as in the $x = 0.23$ manganite [14]. However, there are essential differences in their character.

The data on the Mn-O bond lengths (Fig.2c) indicate that the JT transition from the high temperature orbital disordered O phase to the O' phase possessing the cooperative JT distortions occurs at $T_{JT} \sim 250$ K. This transition involves the strong abrupt changes of the Mn-O-Mn angles between 220°/290 K when the curves of monotonic $T$–variation for the Mn-O1-Mn and Mn-O2-Mn angles cross one another (Fig.2d). This transformation is related to rotation of the MnO$_6$ octahedra around the pseudocubic [111] direction [20]. Thus, the OO' transition is driven by both the JT distortions that underlie an orbital ordering and a steric effect (the oxygen ion displacements connected to the octahedrons tilting). The latter leads to peculiarity of the transition when a relationship between the lattice parameters ($a > b \approx c/\sqrt{2}$, Fig.2a) conserves above and below $T_{JT}$, the splitting $a - b$ becoming even smaller on cooling below $T_{JT}$. The OO' transition is not usually accompanied by the octahedral tilting. In this case $c/\sqrt{2} \approx a \approx b > T_{JT}$ and $c/\sqrt{2} < a, b$ below it, as it is found in LaSr [20], LaCa [21] and $x = 0.23$ NdBa [14] manganites. The $T$–variations of the lattice parameters and unit-cell volume (Fig.2a,b) are observed above $T_C$. They mainly reflect compression of the lattice on cooling which is related to reduction of the lattice parameter. This process completes at $T_C$. The Mn-O bond lengths (Fig.2c) coincide above $T_{JT} \approx 250$ K and their splitting remains nearly temperature independent below 230 K, so that the JT phase forms in the narrow temperature interval. The splitting corresponds the orbital ordering closed to $3y^2 - r^2/3x^2 - r^2$ type which develops in the $a - b$ plane because $d$Mn-O1 $\approx d$Mn-O22 $> d$Mn-O21 [20]. In La$_{1-x}$Ca$_x$MnO$_3$ ($0.12 < x \leq 0.21$) with the FI ground state, the JT-distortions monotonically increases on cooling over a large temperature interval [2]. At $x = 0.2$, this behavior is observed from $T \approx 300$ K $> T_C \approx$ down to 100 K. It was interpreted as the coexistence of the O and O' phases i.e. as the structural PS state. Although our measurements of the dMn-O are not a very high accuracy, they contradict the coexistence of the O and O' phase with the comparable volume fractions even at 220 K, and support formation of the nearly structural uniform phase below 230 K. The $T$–evolution of the structural parameters does not reveal any peculiarities in the anomalous $T$–range ($T^* \approx$
measurements at HIV. MAGNETIZATION

The same values of \( \gamma \approx 150 \text{ K} \) (\(|dMn-O21 - dMn-O1| \approx dMn-O1 - dMnO22\), where it depends weakly on temperature [14], differs from that of the \( x = 0.25 \) manganite. Thus, the FI state and the anomalous critical behavior can occur at the different types of these distortions.

Temperature dependence of the ferromagnetic moment (Fig. 2b) shows the usual behavior for a ferromagnet with \( T_C \approx 129 \text{ K} \). The magnetic moment in the ground state \( 3.0(1) \mu \) (\( \mu \) is the Bohr magneton) is less than that of the Mn sublattice \( 3.75 \mu \) expected for this composition. This is mainly due to antiparallel alignment of the Nd magnetic moment \( \approx 0.5 \mu \) that gives the additive contribution in measured ferromagnetic moment of the sample.

IV. MAGNETIZATION

Fig. 3 displays the temperature dependences of \( M \) that are obtained in the FC and ZFC measurements at \( H = 1 \text{ kOe} \). We consider first the FC data above \( T_C \). To determine a scaling behavior of susceptibility of the material \( \chi \), it is convenient to write inverse susceptibility of the sample \( \chi^{-1}_{\text{ext}} = H/M \) as

\[
\chi^{-1}_{\text{ext}} = \chi^{-1} + 4\pi N. \tag{1}
\]

Here \( \chi = C\chi[S(S+1)/(3kT_C)]((g\mu)^2/V_0)\tau^{-\gamma} \) (the conventional units), \( k \) is the Boltzmann constant, \( C \chi \) is the numerical factor, \( g \) is the \( g \)-factor, \( V_0 \approx 41.45 \text{ Å}^3 \) is the volume of the unit magnetic cell and \( N \) is the demagnetization factor. A fit of the data for \( 1 > \tau > 0.093 \) (\( 258 \text{ K} > T > 141 \text{ K} \)) with \( T_C \approx 129 \text{ K} \) and \( N = 1/3 \) gives \( \gamma = 1.39(1) \), \( C \chi = 2.95(3) \) for \( S = 1.875 \), these values being independent on \( N \) for \( 0.25 < N < 0.4 \) (see inset(1) in Fig. 3).

The same values of \( \gamma \) and \( C \chi \) are obtained by employing an interval \( 1 > \tau > 0.256 \) whereas the fit becomes worse below 141 K. This shows that \( H = 1 \text{ kOe} \) affects the \( \chi(\tau) \) dependence only at \( \tau < 0.093 \). The ac linear response [13] and ESR measurements (see Fig. 5) give a slightly less value of \( \gamma \approx 1.32 \) that is due to the demagnetization effects because a fit of the \( M/H = \chi_{\text{ext}} \) data yields also a close magnitude of \( \gamma \).

Since the nonlinear response reveals the anomalous critical behavior in the low magnetic fields below \( T^* \approx 143 \text{ K} \), it is important to check whether the critical \( M(\tau) \) dependence at \( H = 1 \text{ kOe} \) is the conventional one or not in the same \( T \)-range. Our \( M(\tau) \) data in this region are related mainly to an intermediate \( H \)-regime between the weak (\( T > 143 \text{ K} \)) and strong (\( T = T_C \)) magnetic fields. As can be easily verified, a modified Arrot plot scheme frequently employed for analysis of the critical \( M(\tau, H) \) dependences [22, 23] has the incorrect analytic properties in the weak fields. Therefore we use another approximate equation of state [24] with a correct behavior in the weak and strong fields. It can be presented in a convenient form as [25]

\[
\frac{M}{H_{\text{int}}} = C_0 \left( \frac{\varphi(x)}{\tau} \right)^\gamma \left\{ 1 - a(1 - \varphi(x)) \right\}^{-1}, \tag{2}
\]

\[
x = h_{\text{int}}/\tau^{\beta_H} = A(1 - \varphi(x))^{1/2} \left\{ 1 - a(1 - \varphi(x)) \right\} \varphi(x)^{-\beta_H}. \tag{3}
\]

Here \( A > 0, 1 > a > 0 \) are the coefficients, \( C_0 \) is the factor determining the amplitude of \( \chi \), \( \beta_H = \gamma + \beta \) is the magnetic field index, \( \beta \) is the index of the spontaneous magnetization, \( h_{\text{int}} = g\mu H_{\text{int}}/(kT_C) \) and \( H_{\text{int}} = H - 4\pi N M \) is the internal field. The function \( \varphi(x) \) decreases
monotonically from 1 down to 0 as \( x \) increases from 0 up to \( \infty \). The \( \varphi(x) \) and, consequently, \( M/H_{\text{int}} \) are the even functions of \( x \). They can be expended in the power series in \( x^2 \) for \( x^2 \ll 1 \) because Eq.(3) can be presented as \( x^2 = [\text{the right part}]^2 \). Strongly speaking, \( a = \gamma/\beta_H \) in this approximation. Nevertheless, the parameters \( A, a \) and \( \beta_H \) (or some of them) can be treated as the free ones at an approximate fit (\( \gamma \) has been determined earlier). Insert (2) in Fig.3a displays the best fit of \( M/H_{\text{int}} \) at \( \gamma \approx 1.39, \beta_H \approx 5/3 \) (the value for a 3D isotropic ferromagnet) and \( N = 1/3 \) for our sample closed to a cube. It gives \( a \approx 0.98 \) that is not far from the theoretical value \( a = \gamma/\beta_H \approx 0.81 \). We tried also to find \( \beta_H \) using \( a = \gamma/\beta_H \). This yields \( \beta_H \approx 1.87 \) (\( a \approx 0.74 \)), the fit being worse. These results show that the \( M(\tau, H) \) dependence is comparably well described by the conventional scaling static theory in the anomalous region (\( T_C \leq T < T^* \)) at \( H_{\text{int}} \approx H = 1 \) kOe. Thus, the anomalous nonlinear critical behavior, that is detected by the \( M_z \) measurements in the low magnetic fields (\( H < 100 \) Oe), is suppressed by the stronger field. This means also that the anomalous phase occupies a relative small volume (an upper border does not presumably exceed 1/5).

According to the FC and ZFC data, \( M(H, T) \) exhibits the \( T \)-hysteresis (see Fig.3a and inset in Fig.3b). This phenomenon is usually observed in the manganites below \( T_C \), whereas its presence above \( T_C \) up to 300 K is the unexpected result. The \( \delta M(T) = 2(M_{\text{ZFC}} - M_{\text{FC}})/(M_{\text{ZFC}} + M_{\text{FC}}) \) exhibits the maximum (\( T_m \approx 290 \) K, see Fig.3b) at high temperatures between 220 K and 300 K which can be attributed to the structural transition occurring in this \( T \)-range. Below 220 K, \( \delta M \) increases monotonically at \( T \to T_C + 0 \). This means that difference \( M_{\text{ZFC}}(T) - M_{\text{FC}}(T) \) is not reduced to a \( T \)-independent distinguish in the parameters accounting for the \( M_{\text{ZFC}}(T) \) and \( M_{\text{FC}}(T) \) dependences. One may expect that observation of \( \delta M \) here is related to the two-phase coexistence. The \( M_z \)-measurements reveal the two-phase state below \( T^* \approx 143 \) K as the anomalous nonlinear response becomes larger than the conventional one. Here we use another way for disturbance of the system and analyze another quantity, so that the possible inhomogeneous magnetic state may be observed above \( T^* \). For instance, the increasing of \( \delta M \) on cooling below 220 K may reflect the differences in the \( T \)-dependences of the volume fractions and/or mutual arrangement of the phases. In addition, \( T^* \) seems to be also seen in \( \delta M(T) \) data as a temperature where \( \partial \delta M(T)/\partial T \) has a maximum. At last, \( \delta M(T) \) exhibits the maximum at \( T_C \). This corresponds to a conclusion based on the \( M_z \)-data [15] that the ordering of both phases occurs simultaneously at \( T_C \). These observations agree with an assumption that formation of the two-phase state takes place independently of the structural transition and does not reduce to the coexistence of the \( O' \) and a small amount of the initial \( O \) phases.

Note some peculiarities of the \( T \)-hysteresis below \( T_C \). According to Fig.3a, difference between the FC and ZFC data increases for \( T < 90 \) K. This agrees with \( T \)-dependence of the hysteresis loop found in the \( \text{Re}M_z(H, T) \) measurements. The modification seems to reflect change in a domain formation. Below 20 K the \( M(T) \) exhibits the sharp reduction, indicating onset of ordering of the Nd ions whose magnetization directed antiparallel to that of the Mn moments.

V. ESR

Fig.4 shows the typical spectra for some temperatures obtained at different \( T \)-scans. The similar spectra are also observed for other temperatures above \( T_C \). The spectra recorded at the different treatments well coincide for temperatures above approximately \( T^* \), and are
fitted by a Lorentzian line shape

\[
\chi_{as}(\omega, H) = \frac{1}{2} \chi \left\{ \frac{\omega \Gamma}{(\omega - g \mu H)^2 + \Gamma^2} - \frac{\omega \Gamma}{(\omega + g \mu H)^2 + \Gamma^2} \right\} \cos \varphi_s - \left\{ \frac{\omega (\omega - g \mu H)}{(\omega - g \mu H)^2 + \Gamma^2} - \frac{\omega (\omega + g \mu H)}{(\omega + g \mu H)^2 + \Gamma^2} \right\} \sin \varphi_s \right\}.
\]

(4)

Here \(\chi_{as}\) is the antisymmetric component of the dynamic susceptibility of the sample, \(\chi\) is the static susceptibility, \(\omega\) is the frequency of \(ac\)-field, \(\Gamma\) is the relaxation rate of a uniform magnetization, and \(\varphi_s\) is the phase of the signal. Since \(\Gamma\) is a rather large, the both circularly polarized components of \(ac\) field are taken into account in Eq.4. On this reason, a control sample (a polycrystalline stable nitroxyl radical with \(g = 2.055\) and \(\Gamma \approx 60\) Oe) is used for tuning a phase of \(ac\) field which is the reference point for \(\varphi_s\). The fitting parameters in Eq.4 are \(\Gamma, g, \varphi_s\) and the amplitude of the signal \(A_{as}(T) \propto \chi\).

We consider first a range \(T^* \div 2T_C\) where the critical behavior follows to that of a 3D isotropic ferromagnet [15], and the structural refinement does not show any structural transformation. Fig.5 presents the \(\Gamma(T)\) and \(A_{as}(T)\) dependencies for the different \(T\)-scans which are obtained between \(T^*\) and \(2T_C\). It is seen that the different treatments of the sample have a little effect on the data. The \(g\)-factor \((=2.00(1))\) is independent of temperature here. \(A_{as}(\tau)\) is expected to obey a scaling law for a 3D isotropic ferromagnet, \(A_{as}(\tau) \propto \tau^{-\gamma}, \gamma \approx 4/3,\) in a weak field regime at \(\tau > \tau_H = (g \mu H/kT_C)^{3/5} \approx 3.1 \cdot 10^{-2}\) for resonance field \(H \approx 3\) kOe. The inset in Fig.5b shows that \(A_{as}(\tau)\) curve follows this prediction down to approximately \(\tau \approx 0.11\) and deviates from it at \(\tau < 0.11\). This behavior is in a reasonable correspondence with that of \(\chi/\tau\) found in the \(M(H)\) (see above) and low frequency measurements of the linear response on a weak \(ac\) field [15].

Let us go to \(\Gamma(\tau)\). In the weakly anisotropic cubic ferromagnets, it is usually given by [14, 26, 27]

\[\Gamma(\tau) = \Gamma_c \tau^{-1} + \Gamma_{unc} \tau^{4/3}.\]

(5)

Here \(\Gamma_c\) is controlled by the dipolar forces, a single ion anisotropy \((\propto S^\alpha S^\beta)\) and anisotropy of the exchange interaction (the two latter appear in the \(Pbnm\) symmetry) [22, 28]. The term with \(\Gamma_{unc}\) describes the uncritical contribution including a spin-lattice relaxation rate. The Dzyaloshinskij-Moria interaction also presents in this space group [22, 28]. Following to an analysis given in Ref. [29], one can show that this interaction gives contribution in \(\Gamma_{unc}\) only since it connects fluctuations of the magnetization (critical) and staggered magnetization (uncritical) whose characteristic energy \(\sim T_C\). The \(\Gamma(\tau)\) exhibits a critical enhancement at \(\tau \to 0\) below a minimal value at \(\tau_m\). This behavior is observed in the traditional cubic ferromagnets [16] and in the near to cubic manganites La\(_{1-x}\)Ca\(_{x}\)MnO\(_3\) \((x = 0.18 \div 0.22)\) with \(\tau_m \sim 0.25\) [17]. In our compound \(\Gamma(\tau)\) reduces monotonically at \(\tau \to 0\). This dependence cannot be described by the single term with \(\Gamma_{unc}\) and suggests the presence of a new relaxation mechanism. Since Nd\(^{3+}\) ion has a magnetic moment and this magnetic subsystem is not presented in the indicated above compounds, a Mn-Nd coupling is the most likely to be additional source of the relaxation. The ground state of Nd\(^{3+}\) ion (total moment \(J = 9/2, L = 6\) and \(S = 3/2\)) is split by a low symmetric (lower than cubic) crystal field into the five doublets described by the effective spins \(S = 1/2\). It is expected that a Mn-Nd exchange interaction \(\hat{J}_{12}\) is larger than that of the Nd-Nd [4, 23]. We will show that the weak anisotropic coupling \(\hat{J}_{12}\) can give the needed contribution in \(\Gamma(\tau)\). The main effect of \(\hat{J}_{12}\) can be explained by taking into account the single ground doublet of the Nd\(^{3+}\) ion.
and neglecting the Nd-Nd interactions. We consider first a result of a perturbation theory for $\Gamma$ with $H_{\text{int}} = \sum_{i,k} S_{1i} \hat{J}_{12ik} S_{2k}$, where $S_{1i}$ and $S_{2i}$ are spins of the Mn and Nd ($S_2 = 1/2$) ions, respectively. A relaxation rate $\Gamma_{\text{MnNd}}$ of total spin of the system $S = S_1 + S_2$ is determined by an anisotropy of $H_{\text{int}}$. To get $\Gamma_{\text{MnNd}}$, a pair spin Green function of the operators $\partial S^\alpha/\partial t$ is analyzed. This includes a decoupling of a four-spin Green function, which appears as a result of a commutating $[S^\alpha, H_{\text{int}}]$ into product of the two-spin functions. In our case the latter are the Green functions of the Mn $(G_1)$ and Nd $(G_2)$ subsystems, $G_1$ having the properties of a 3D isotropic ferromagnet and $G_2$ corresponding to the free Nd spins. The $\Gamma_{\text{MnNd}}$ is also determined by an uniform static Green function of the system $G^{(0)} = G^{(0)}_1 + G^{(0)}_2$ $(\Gamma_{\text{MnNd}} \propto (G^{(0)})^{-1}, G^{(0)} \approx G^{(0)}_1$ at a small $\tau$ because $G^{(0)}_1 \sim T^{-1} \tau^{-4/3}$ and $G^{(0)}_2 \sim T^{-1})$. As a result, we find $\Gamma_{\text{MnNd}}(\tau) \propto \tau^{1/3}$. This answer differs from the critical term $(\propto \tau^{-1}, \text{Eq.}(2))$ in a factor $\tau^{4/3}$ since the critical contribution contains product of the two critical functions $(G^2)$ whereas $\Gamma_{\text{MnNd}} \propto G^{(0)}_1 G^{(0)}_2$ that gives $G^{(0)}_2 / G^{(0)}_1 \propto \tau^{4/3}$. The magnitude of $\Gamma_{\text{MnNd}}$ is determined by anisotropic part of the $\hat{J}_{12}(q)$ at $q = 0$, where $\hat{J}_{12}(q)$ is the Fourier transform of $\hat{J}_{12ik}$, $\hat{J}_{12}(0) = Z_{12} \hat{J}_{12} \equiv \hat{U}_{12}, Z_{12} \approx 8$ is the coordination number of the Mn-Nd sublattice. This part is $(U^{\text{an}}_{12})^{\alpha\beta} = U^{\alpha\beta}_{12} - \delta_{\alpha\beta} \hat{U}_{12}$, where $\hat{U}_{12} = \text{Sp} \hat{U}_{12}/3$ is the mean exchange coupling. Generally speaking, one can find the different relaxation rates along the axes of the crystal. However, the anisotropy of $\Gamma$ is not observed. It can be due to a twinning of the crystal in the $(a, b)$ plane and an averaging of the relaxation rates by the circular polarized components of $h(t)$. Therefore, we introduce an effective exchange anisotropy $(U^{\text{an}}_{12})^{\alpha\beta} \to U_{12}^{\text{an}}$ and write

$$\Gamma_{\text{MnNd}} = \Gamma_{\text{MnNd}}^{\text{an}} \tau^{1/3}, \quad \Gamma_{\text{MnNd}}^{\text{an}} \sim (U_{12}^{\text{an}})^2 / T_C. \tag{6}$$

$\Gamma_{\text{MnNd}}^{\text{an}}$ reflects an exchange narrowing that is due to the strong isotropic exchange interaction in the Mn subsystem. This law is the consequence of decay of the uniform magnetization into the critical and uncritical modes which is enforced by the anisotropy, a characteristic energy of the free Nd-spins being obviously equal to zero. Note that the same contribution in $\Gamma$ appears also as an uncritical (or rather intermediate) mode has an uncritical amplitude ($\sim T_C$) and a characteristic energy that is equal that of the critical mode. This occurs in some cubic collinear ferrimagnets.

To determine a region of the validity for this result, the equations of motion for the Green functions $G^{\text{an}}_1(q, \omega)$ and $G^{\text{an}}_2(q, \omega)$ at $q = 0$ should be considered. The $\hat{U}_{12}$ introduces a frequency scale $\Omega_2(\tau)$ in $G^{\text{an}}_2(\omega)$ that gives the main restriction. The $\hat{U}_{12}$ is assumed to be rather anisotropic $| (U_{12}^{\text{an}})^{\alpha\beta} | \sim | \hat{U}_{12} |$. This is not unusual situation for the exchange coupling between the $d$ and $f$ ions in the perovskite lattice and an estimation given below supports this assumption. Therefore, one can use the isotropic component $\hat{U}_{12}$ to get $\Omega_2(\tau)$. The traditional mode coupling theory yields $\Omega_2(\tau) \sim (U_{12}^{\text{an}} T_C)^{-1}$. Result (6) is valid if $\Omega(\tau) = T_C \tau^{5/3} > \Omega_2(\tau)$, i.e. $\tau > \tau_\text{an} \sim (\hat{U}_{12} / T_C)^{3/4}, | \hat{U}_{12} | \sim | (U_{12}^{\text{an}})^{\alpha\beta} |$, where $\Omega(\tau)$ is the characteristic energy of the critical fluctuations for a 3D isotropic ferromagnet. At $\tau < \tau_\text{an}$, a new dynamic regime originates which corresponds to a system with completely destroyed conservation law of the $\mathbf{S}$. This regime develops when the static behavior weakly changes. Employing the usual two sublattice expressions for $G^{(0)}$ and $\hat{U}_{12}\delta_{ab}$ for the estimations, we find for a static anisotropic border $\tau_{\text{an}}^\text{st} \sim \tau_\text{an}^2$. Note that the small anisotropic spin interactions in the Nd subsystem $U_{\text{NdNd}}^\text{an}$ (for instance, the dipolar forces) give an additional contribution in $\Gamma_{\text{unc}}$. They do not effect $\tau_\text{an}$ if $\Omega(\tau_\text{an}) \gg | (U_{\text{NdNd}}^\text{an})^{\alpha\beta} |$.

We have discussed above dynamics of the $\mathbf{S}$ which coincides with that of the total magnetization $\mathbf{M}$ when the $g$-factors of the Mn ($g_1$) and Nd ($g_2$) ions are isotropic and equal to
each other. The anisotropy of $\hat{g}_1$ can be neglected whereas that of $\hat{g}_2$ may be substantial. Consider first a case of isotropic $g_1 \neq g_2$. When $\dot{J}_{12} = 0$, the spectrum reveals the two resonance at $\omega_{1,2} = g_{1,2}\mu H$. With increasing of isotropic part of the Mn-Nd exchange, the lines begin to overlap. In a regime of a strong exchange narrowing ($|\bar{U}_{12}| \gg g_{1,2}\mu H$), the spectrum is dominated by a single resonance determined by an effective $g$–factor $g_{\text{eff}}$.

Contributions of the $g_1$ and $g_2$ in $g_{\text{eff}}$ depend on the relationships between $G_1(0), G_2(0)$ and $|\bar{U}_{12}|$ as well as $S_1$ and $S_2$. When $G_1(0)$ dominates ($G_1(0) \gg G_2(0) ; g_{\text{eff}} \approx g_1$ if $|\bar{U}_{12}|/G_2(0) \leq 1$ and $S_1 \geq S_2$). In virtue of the same reason, the anisotropy of $\hat{g}_{\text{eff}}$ related to that of $\hat{g}_2$ and $\bar{U}_{12}$ is also to be small. The expression for $\Gamma_{\text{MnNd}}$ (6) corresponds to the damping of this resonance. Note, that in our case the admixture of $g_2$ to $g_1$ is very small: $g_{\text{eff}} = g_1 + Cg_2$ [31], where $C \sim [S_2(S_2+1)/S_1(S_1+1)]^2(G_2(0)/G_1(0)) \approx 4 \cdot 10^{-4}$ even at $\tau = 1$ because $G_1(0) = C \chi S_1(S_1+1)/3TC$, $C \chi \approx 2.95$, $S_1 \approx 1.88$ and $G_2(0) = C_{\text{Nd}}S_2(S_2+1)/6TC$, $C_{\text{Nd}} = 0.75$ is the concentration of Nd atoms and $|\bar{U}_{12}|/G_1(0) \ll 1$ for estimated below $|\bar{U}_{12}| \sim 5 \text{ K}$.

The same factor $C$ accounts for the $T$-dependent corrections to $g_2$ related to the Mn-Nd coupling. The closeness $g_1 = 2.00(1)$ to $g_2 \approx 2$ for La$_{1-x}$Ca$_x$MnO$_3$ [17] and its independence from $T$ agree well with the smallness of $C$.

Going to the data, we present $\Gamma$ by

$$\Gamma(\tau) = \Gamma_c \tau^{-1} + \Gamma_{\text{MnNd}}^{\text{an}} \tau^{1/3} + \Gamma_{\text{unc}} \tau^{4/3}. \quad (7)$$

Inset in Fig.5a shows the fit of the $\Gamma(\tau)$ for $1 > \tau > 0.13$. The fitting parameters are $\Gamma_c = 9(2) \text{ Oe}$, $\Gamma_{\text{MnNd}}^{\text{an}} = 590(20) \text{ Oe}$ and $\Gamma_{\text{unc}} = 505(24) \text{ Oe}$. The $\Gamma_c$ is controlled by the dipolar forces since the corresponding contribution is estimated as $\Gamma_d \sim 4\pi(g\mu)^2/V_0TC \approx 16 \text{ Oe}$ for $V_0 \approx (3.9)^3 \text{ A}^3$ [16]. It is remarkable that for $\Gamma_{\text{MnNd}}^{\text{an}} = 0$ we obtain $\Gamma \approx 115 \text{ Oe}$ at $\tau \approx 0.25$. This value is very close to $\Gamma = 120 \div 130 \text{ Oe}$ which was found in La$_{1-x}$Ca$_x$MnO$_3$ ($x = 0.18 \div 0.22$ and $T_C = 180 \text{ K} \div 188 \text{ K}$) at $\tau_m \sim 0.25$ [17]. This observation strongly supports the assumption on the leading role that plays the Mn-Nd coupling in the suppression of critical enhancement of the $\Gamma(\tau)$. From Eq.6, we find $|U_{12}^{\text{an}}| \sim 3.2 \text{ K}$. The strength of $\bar{U}_{12}$ is not completely known. We will try to estimate $\bar{U}_{12}$ exploiting $T$-dependence of the small Nd magnetic moment (0.45 $\mu$ at 1.5 K) which was measured by neutron diffraction in Nd$_{0.7}$Ba$_{0.3}$MnO$_3$ for 20 K $T \geq 1.5 \text{ K}$ [7]. This dependence suggests that Nd magnetization is induced by the ordered Mn subsystem, i.e. it is due to the splitting of the Nd ground Kramers doublet by the mean field of the Mn spins. Assuming that $\bar{U}_{12}$ dominates and $g_2 \sim 2$, we find $|\bar{U}_{12}| \sim 5 \text{ K}$ of the same order as $|U_{12}^{\text{an}}|$.

The border of the anisotropic dynamical crossover is $\tau_{\text{an}} \sim \langle |\bar{U}_{12}^{\text{an}}|/TC \rangle^{3/4} \approx 6.3 \cdot 10^{-2}$. The weak anisotropic spin interactions in the Nd subsystem do not affect $\tau_{\text{an}}$ if $T_C \tau_{\text{an}}^{5/3} \approx 1.3 \text{ K} > |(U_{\text{MnNd}}^{\text{an}})_{a\beta}|$. This condition satisfies for the Nd-Nd dipolar interaction whose magnitude does not exceed that of the Mn-Mn ($\sim 0.5 \text{ K}$). The exchange coupling of the Nd spins seems to be an order of 1 K or less [32]. The $\tau_{\text{an}}$ is close to the minimal $\tau \approx 0.13$ which is available for the precise analysis of the $\Gamma(\tau)$. Since $\Gamma(\tau)$ cannot exhibit a critical enhancement in the anisotropic regime, this effect is completely destroyed in our compound that is confirmed by estimation of the line width at $\tau < 0.13$.

The anisotropic static crossover occurs at a very small $\tau_{\text{an}}^{st} \sim \tau_{\text{an}}^{2} \approx 4 \cdot 10^{-3}$ ($T_{\text{an}}^{st} = T_C + 0.5 \text{ K}$). It is not surprising, therefore, that $\chi(\tau)$ dependence of our compound and the static critical properties of Nd$_{0.6}$Pb$_{0.4}$MnO$_3$ ($T_C \sim 156 \text{ K}$) [10] correspond to those of a 3D isotropic ferromagnet.
We have characterized above the Nd$^{3+}$ ion by its single ground doublet. Other states are ineffective if a distance between the ground doublet and a next one is larger than $2T_C$. It can be a realistic situation, since a cubic component, which dominates in the crystal field of the weakly distorted cubic crystal, can produce a large splitting between the ground doublet and the two upper quartets. In a comparably strongly distorted NdCuO$_4$, for example, this splitting is 15 meV whereas the total splitting is 109 meV [33]. If the next several states are relevant they can be simply taken into account since a distance between the neighboring doublets is definitely larger than Mn-Nd exchange. In this case these states can be treated separately by the same manner as the ground state so that they produce the obvious additive effect.

We consider now the ESR data below $T^*$ where a pronounced deviation of the signal from Lorenzian starts to be observed. A reason is that $\text{Im}4\pi\chi(x,x)(\omega,H) \gg 1$ at $\omega \approx g\mu H$ as $4\pi\chi \sim 1$ ($4\pi\chi \approx 0.6$ at $T^*$). This leads to a complex nonuniform distribution of the $ac$-field in the sample because a wave vector $k$ in a medium $k \propto \left\{\mu_0 = 1 + 4\pi\chi_{xx}(\omega,H)\right\}^{1/2}$ for a wave spreading along $H$, where, for simplicity, we neglected the $\chi_{as}(\omega,H)$. As a result, phase $\varphi_s$ of the signal begins to exhibit a strong $H$-dependence that cannot be correctly determined for our sample in the resonator. Therefore below $T^*$, we present directly the signals for the several types of the $T$-scans. The pronounced $T$-hysteresis observed in all the measurements (Fig.4) is the clear indication of the anomalous behavior that develops below $T^*$. Since a precise quantitative analysis of the signal is not feasible here, we cannot determine whether this hysteresis is caused by that of $M$ or it is related also to hysteresis of the $\Gamma$ and $g$–factor. A rough estimation gives approximately $T$-independent values for $\Gamma$ and $g$–factor below $T^*$ down to $T_C$. The same reason does not allow us to elucidate a possible two-phase character of the signal. At the same time, above $T^*$ phase $\varphi_s$ reflects mainly phase shift of the $h(t)$ in the sample which is related to a comparably large $\varepsilon \sim 10$ and conductivity $\sigma$. This effect, being independent of $H$, can be taken into account correctly by Eq.(4). For the largest conductivity ($\sigma \approx 1.7(\Omega\text{cm})^{-1}$) at $T = 2T_C \approx 260$ K, we get $l_{\perp} | k | \sim 0.25$ for $\mu_0 \approx 1$, were $l_{\perp} \approx 0.1$ mm is the thickness of the sample. Since $\sigma$ decreases sharply on cooling ($\sigma \sim 0.3 \cdot 10^{-2}$ (\Omega cm)$^{-1}$ at $T^*$), the $h$-distribution in the sample is closed to uniform between $T^*$ and $2T_C$.

VI. DISCUSSION AND CONCLUSION

The $T$-hysteresis observed in the magnetization and ESR measurements above $T_C$ evidences at least a weak first order transition. According to the $M_2$-data, a reason of changing regime of the transition at $T^*$ is formation of the new phase with the anomalous strong nonlinear magnetic behavior in the weak fields which is stronger than that of a 3D isotropic ferromagnet near $T_C$. This phase coexists with the normal one at least from $T^*$ down to $T_C$. This PS magnetic state cannot be trivially related to a two-phase structure because, according to the neutron diffraction, the sample remains the structural uniform compound below $T^*$. Note that, first, the anomalous phase coexists with the conventional one possessing the different orbital ordering for the $x = 0.23$ and $x = 0.25$ compounds. Second, it arises in the manganites that are closed to border of the I-M transition ($x \approx 0.3$), where, according to Ref.[7] and our preliminarily data, the orbital ordering disappears. These observations suggest that the anomalous behavior can be attributed to an orbital disordered metallic phase. The lack of observability of the anomalous phase in the structural measurements means that this phase is structurally closed to the conventional one and/or its volume is
small. Fragmentation of the sample in the PS state may be such that the anomalous phase does not form a percolative conductive cluster, and the system remains an insulator. The strong \( H \)-nonlinear behavior of this phase is expected to be a combined effect of its own properties and a magnetic coupling of its fragments through the normal phase with the developed ferromagnetic correlations. It is very important to stress a difference between this transition and a first order one whose character is determined on the base of \( M(H) \) data. In the latter, type of the transition (first/second) is characterized by sign of coefficient \( b \) in an expansion of \( H/M \) in \( M^2 \) above \( T_C \): \( H/M = a + bM^2 + \ldots \) (\( b < 0 \) (first order), \( b > 0 \) (second order)). Such a criterion suggests that the magnetization is the single critical degree of freedom accounting for the transition. This peculiarity is indeed observed in the \( M(H) \) dependence of traditional ferromagnet MnAs \[34\] and a number of the manganites \[35, 36\]. Besides the conventional \( M(H) \) measurements, sign of \( b \) can be found from the \( M_2 \)-data because \( \text{Re}M_2(\omega, H) \propto \partial^2 M/\partial H^2 = -6(b/a^4)H \) for \( H \to 0 \) and a small \( \omega \) when \( \text{Re}M_2 \) dominates in the response \[13\]. Our results on the \( x = 0.23 \) and \( x = 0.25 \) compounds obtained by this method indicate clearly that \( b > 0 \) for both coexisting phases \[13, 15\]. This implies that \( M \) is not the single degree of freedom involved in the transition, and one should consider the charge, orbital and JT-phonon variables (or some of them) as the critical ones as well. It is not the unexpected conclusion for the manganites that are characterized by close interconnection of the magnetization and these degrees of freedom. In our system, the charge and orbital degrees of freedom together with \( M \) are the most likely to be the critical variables. The JT-subsystem seems to be less important because the JT distortions are small.

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Figure captions

Fig. 1. The neutron diffraction pattern, calculated profile and residual curve for Nd$_{0.75}$Ba$_{0.25}$MnO$_3$ at room temperature. The insert shows the wide and weak monoclinic peaks at the small angles.

Fig. 2. Temperature dependences of the structural parameters $a, b, c$ (panel (a)); volume of the unit cell $V$ and magnetic moment of the sample per Mn ion (panel (b)); the Mn-O bond lengths (panel (c)); and the Mn-O-Mn angles (panel (d)).

Fig. 3. Temperature dependence of the magnetization for the cooling and heating regimes under $H = 1$ kOe. Panel (a) shows these dependencies in the full temperature range $6 \div 300$ K. Insert (1) displays fit of the $M^{-1}(T)$ measured in the ZFC regime to the function $\tau\gamma/CH + 4\pi N/H$. The fitting parameters are found to be $1/CH = 3.98(3)$ g/emu and $\gamma = 1.39(1)$ at $N = 1/3$. Insert (2) represents fit of the ZFC $M(\tau)/H_{int}$ to expressions (2), (3) (see the text) in $T$-range $T_C \div 261$ K. Panel (b) shows the relative difference $\delta M$ versus $T$ in the $T$-range $80 \div 300$ K. Insert in (b) displays the $M(T)$ curves registered in the ZFC and FC regimes in $T$-range $140 \div 180$ K.

Fig. 4. The ESR spectra for the different $T$-scans at some close temperatures: cooling (solid line, panels (a-d)), heating after fast cooling (dashed line, panels (a-c)), cooling under $H = 4$ kOe (dotted line, panels (a) and (d)). Panels (a) and (c) represent also fit of the spectra recorded on cooling to a Lorenzian (dash-dotted line).

Fig. 5. Temperature dependencies of parameters of the ESR spectra for different $T$-treatments of the sample: cooling (full symbols) and heating (open symbols) under $H = 0$ (squares) and $H = 4$ kOe (triangles); heating after fast cooling (stars). Panel (a) displays the spin relaxation rate $\Gamma$ versus temperature. Insert shows $\Gamma(\tau)$ dependence on cooling, and its fit described in the text. Panel (b) represents amplitudes $A_{as}(T)$ of the spectra versus $T$. Insert shows fit of $A_{as}(\tau) \propto \chi(\tau)$ to the scaling law.
\( \delta M = 2 (M_{ZFC} - M_{FC}) / (M_{ZFC} + M_{FC}) \)


\[
\begin{align*}
\chi''(10^3 \text{arb. units}) & \quad H (\text{kOe}) \\
T = 130 \text{ K} & \quad T = 138.9 \text{ K} \\
T = 145.6 \text{ K} & \quad T = 240.2 \text{ K}
\end{align*}
\]
(a) Amplitude, $A_{as}$ (arb. units)

(b) Amplitude, $A_{as}$ (arb. units)

$\tau = \frac{(T-T_C)}{T_C}$

$A_{as} = A_0 \tau^{-\gamma}$

$\gamma = 1.32(3)$