Ferromagnetic manganites: Half-metals versus polaronic conductors

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The spin-lattice and spin-spin NMR relaxation times T_1 and T_2 of ^{55}Mn and ^{139}La were measured as a function of temperature in several (LaNa)MnO_3 perovskites, which are ferromagnetic and exhibit metallic conductivity. Exponential dependence was observed for both T_1^{-1} and T_2^{-1}. In the critical region the relaxation rates are further enhanced. The NMR relaxation may be explained by fluctuations of hyperfine fields caused by the hopping of the e_g electrons of Mn ions. The spin dynamics is discussed in terms of the half-metal and polaronic models of the manganites. [S0163-1829(99)07413-5]

I. INTRODUCTION

Perovskite manganites with mixed Mn^{3+},Mn^{4+} valence have attracted a great deal of attention recently, due to the ''colossal'' magnetoresistance observed in ferromagnetic samples near the Curie temperature. The basis for the understanding of simultaneous existence of ferromagnetism and metallic conductivity in these compounds is the concept of the double exchange, which is associated with the hopping of electron holes between Mn ions.[1] However, in order to explain their anomalous properties, strong electron correlations and/or strong electron-lattice interaction must also be considered. Corresponding theoretical models include the Fermi-liquid to polaron crossover,[2] composite polaron mechanism,[3] electron localization due to the spin disorder,[4] nonmagnetic randomness,[5] etc. First-principle calculations of the electron structure of $La_{0.7}Ca_{0.3}MnO_3$ (Ref. 6) indicated that the system in question is effectively half-metallic, i.e., at low temperatures it is metallic for the majority spin channel, while the minority spin electrons exhibit the Anderson localization.

An important feature of the systems in question is the spin dynamics, as it is intimately connected with the colossal magnetoresistance phenomena. Experimental studies of the spin dynamics can be used to test various theoretical approaches as it is sensitive to the model used. Several recent experiments indicate unusual dynamics in the manganites. Using muon spin relaxation, Heffner *et al.*[7] observed anomalously slow spin fluctuations in $La_{0.67}Ca_{0.33}MnO_3$. Mössbauer spectroscopy of the same system doped with Fe and Sn was reported by Simopoulos *et al.*[8] From rather scarce data obtained on ^{57}Fe and ^{119}Sn these authors deduced that the relaxation rates increase exponentially with the temperature. A rapid increase of ^{139}La spin-lattice relaxation rate with the increasing temperature in nonstoichiometric $LaMnO_3$ was found in Ref. 9 using NMR in high magnetic field. Although the results of the above experimental studies, provide valuable information on the spin dynamics, they do not cover directly the most important item — the spin fluctuations on the manganese ions themselves. In our recent papers[10,11] the temperature dependence of the NMR on ^{55}Mn nuclei in several ferromagnetic manganites was studied. A fast increase of the spin-spin relaxation rate with increasing T was found in (Pr,Ba) and (Pr,Ca,Sr) manganites.[10] In the (LaNa) system the relaxation rate in the region close to T_C was studied to clarify the order of the magnetic phase transition.[11]

In this paper we present an extensive study of the temperature dependence of the NMR relaxation in the $La_{1-x}Na_xMnO_3$ system for three concentrations of Na ($x=0.1$, 0.15, and 0.2) between 60 K and T_C. In the system in question Na^{1+} ion is substituted for trivalent La^{3+}. The concentration of Mn^{4+} ions induced by this substitution is twice that obtained in systems with the divalent substitutions and thus it covers the region of ferromagnetism and metallic conductivity (20–40 % of Mn^{4+}). In all three compounds spin-lattice and spin-spin relaxation times T_1,T_2 for ^{55}Mn nuclei were determined. In addition for $x=0.2$ the relaxation of ^{139}La nuclei was also studied.

II. EXPERIMENTAL PROCEDURE AND RESULTS

The polycrystalline samples used in the present study were described in Ref. 11. Curie temperature was determined from the magnetic and NMR measurements to be 266, 315, and 325 K for $x=0.1$, 0.15, and 0.2, respectively.[11] The spin-echo measurements were performed in zero external magnetic field using a noncoherent spectrometer with frequency sweep and boxcar detector signal averaging. As the NMR spectra of both ^{55}Mn and ^{139}La are broad, short pulses $\sim 0.5-1$ μs were used. We start with the description of the measurements of the ^{55}Mn relaxation. The spin-spin nuclear relaxation time T_2 was determined from the decay of the two-pulse spin echo. We have found that the decay of the amplitude of the spin echo is well described by the sum of two exponentials, moreover the decay depends on the radio-frequency (rf) field (Fig. 1). For sufficiently high rf field this dependence saturates. Shown in Fig. 2 are the relaxation times (a) and the amplitudes (b) of the two components vs the rf field, obtained by decomposing the decay. We attribute the faster component to the contribution of domain walls and the slower component to the nuclei in the domains. The char-

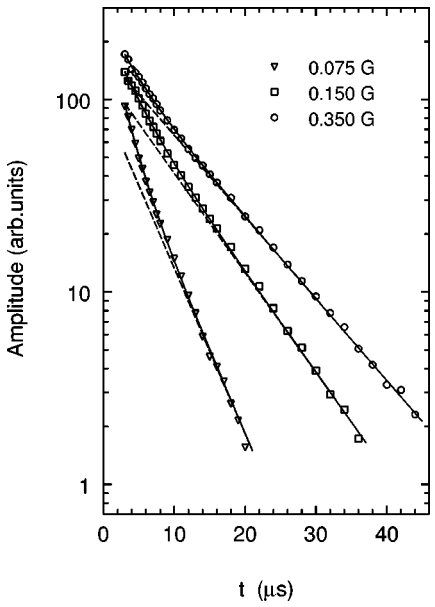

FIG. 1. $La_{0.8}Na_{0.2}MnO_3$. Spin-echo decays at $T=77$ K and frequency 373 MHz for three different rf fields. Full curves correspond to the fit with two exponentials, while the dashed lines correspond to the slower component.

acter of the dependence of the corresponding amplitudes on the rf field [Fig. 2(b)] supports this conclusion. The inhomogeneity of the system studied leads to an inequivalency of ^{55}Mn nuclei as far as NMR is concerned. Even for nuclei in the domains a distribution of T_2 as well as a distribution of optimal resonance conditions exist. This is a probable reason for the dependence of the decay on the rf field we observed.

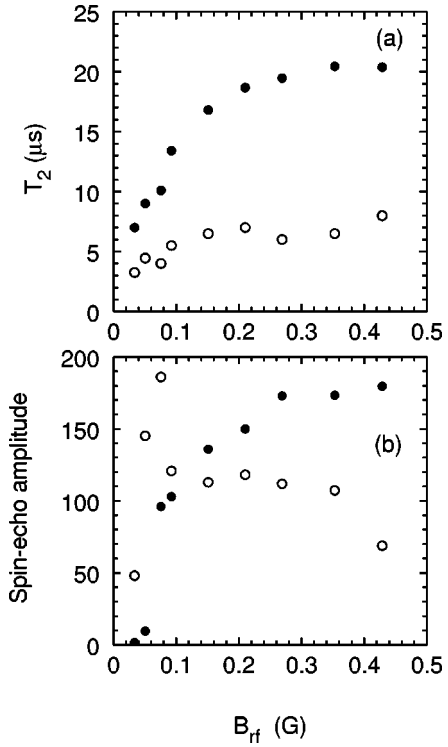

FIG. 2. $La_{0.8}Na_{0.2}MnO_3$. T_2 (a) and amplitude (b) for the fast (○) and slow(●) components of the spin-echo decays as functions of the rf field. The experimental conditions as in Fig. 1.

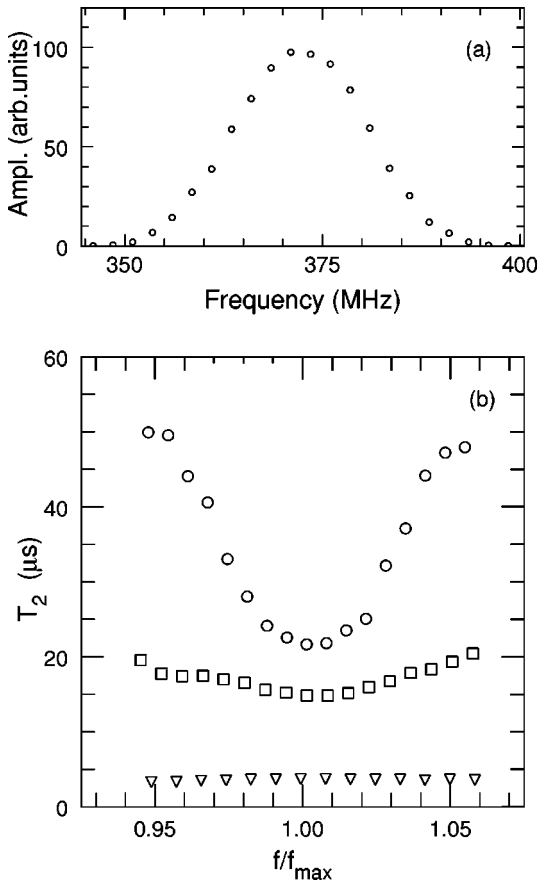

FIG. 3. $La_{0.8}Na_{0.2}MnO_3$. (a) NMR spectrum of ^{55}Mn at $T=77$ K corrected for the relaxation. (b) Spin-spin relaxation time T_2 as a function of reduced frequency at $T=77$ K (○), $T=140$ K (□), and $T=240$ K (▽).

Taking this into account, T_2 of the slower component in the saturated region [Fig. 2(a)] is considered in the following discussion.

The actual rf field acting on the nuclei consists of the external rf field B_{rf} and the field produced by the oscillating magnetic moments of electrons. The presence of the electronic magnetic moments thus enhances the effect of B_{rf}. Corresponding enhancement factors may be estimated from the values B_{rf}^m of the external rf field, at which the amplitude of the spin echo attains its maximum. From the Fig. 2(b) we determine $B_{rf}^m=0.4$ G (0.06 G) for the nuclei in domains (domain walls). Corresponding enhancement factors are $\sim$3000 for the nuclei in the domains and $\sim$20 000 for the nuclei in the domain walls.

Still another complication when studying relaxation in these system is the frequency dependence of the relaxation time T_2 across the NMR spectrum. This is illustrated in Figs. 3(a), 3(b). Analogous behavior was observed by Leung and Morrish[12] in $(LaPb)MnO_3$ at $T=4.2$ K. The change is substantial at low temperatures and decreases with increasing temperature. In Fig. 4 two sets of data are displayed — faster and slower rates correspond to the center and the wing of the line, respectively. In Ref. 12 this behavior was ascribed to the Suhl-Nakamura interaction in which the nuclear spins interact via virtual electronic spin waves. The minimum in T_2 occurs in the center of the line because majority of the nuclear spins precess at or near this frequency. In what fol-

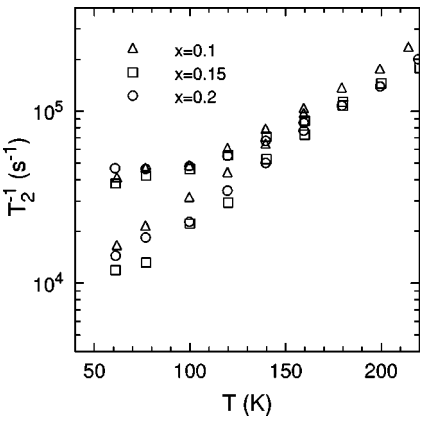

FIG. 4. $La_{1-x}Na_xMnO_3$. Temperature dependence of the relaxation rate T_2^{-1} of ^{55}Mn. Two sets of data correspond to faster and slower relaxation in the center and the wing of the resonance line, respectively.

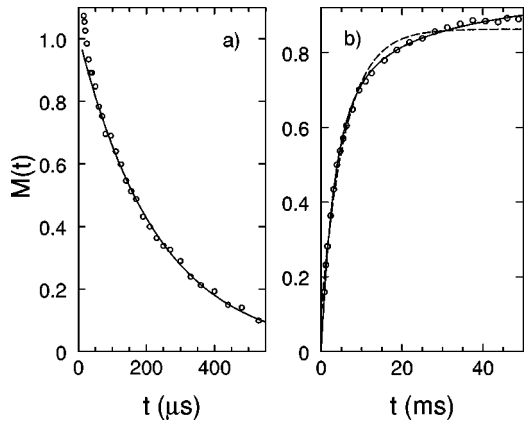

FIG. 5. $La_{0.8}Na_{0.2}MnO_3$, $T=77$ K. (a) Decay of the transversal component of ^{139}La nuclear magnetization *M*. Solid curve is a fit by a single exponential. (b) Decay of the longitudinal component of *M*. Dashed curve is a single exponential fit while the solid curve corresponds to Eq. (1).

lows T_2^{-1} corresponding to the wing of the resonance line is considered, as this is only weakly influenced by the extra relaxation connected with the Suhl-Nakamura interaction.

The spin-lattice relaxation time T_1 was determined from the recovery of spin-echo intensity after the inversion of nuclear spins by a 180° rf pulse (method IR) and/or from the saturation of the spin-echo signal (method S). We have found that within the experimental error IR and S methods lead to identical results.

^{55}Mn nuclei has nuclear spin $I=5/2$ and it may thus experience the quadrupole interaction. If the corresponding quadrupole splitting exceeds the excitation bandwidth (≈ 1 MHz) a multiexponential behavior of the nuclear magnetization is expected. The T_1 data we obtained can be well described by a single exponential, however. This indicates that the quadrupole splitting is small, i.e., the octahedrons of Mn ions are only slightly distorted. Similar behavior was observed by Allodi *et al.*[13] Only small (if any) distortion of the octahedra in the ferromagnetic, metalliclike manganites was also found by the EXAFS spectroscopy.[14,15]

For ^{139}La nuclei the decay of the two pulse spin echo may be described by a single exponential, except for the data obtained using the shortest pulse delays [Fig. 5(a)]. No change of T_2 across the NMR spectrum was observed. In contrast to ^{55}Mn, the longitudinal component of ^{139}La nuclear magnetization probed by the IR method does not exhibit the single exponential behavior [Fig. 5(b)]. This indicates that quadrupole interaction of ^{139}La cannot be neglected. Analogous conclusion was made by Allodi *et al.*[9] Assuming that the spin-lattice relaxation mechanism is predominantly magnetic and only the central $-1/2 \rightarrow 1/2$ transition is saturated, the recovery is described by[9]

$$M(t)=M_0\left(1-\frac{4}{21}e^{-2Wt}-\frac{2}{11}e^{-12Wt}-\frac{20}{91}e^{-30Wt}-\frac{175}{429}e^{-56Wt}\right), \quad (1)$$

where the transition probability $2W=T_1^{-1}$.

The relaxation rates obtained as discussed above are shown as functions of *T* in Figs. 6,7. The temperature dependence of the relaxation rates of ^{55}Mn is similar in all compounds studied. The form of T_1^{-1} and T_2^{-1} vs *T* dependence is almost the same, but T_2^{-1} is by an order of magnitude larger. A similar striking result was obtained for all Na concentrations — the spin relaxation phenomena exhibit a rate, which increases exponentially with increasing temperature in a broad temperature interval. We have observed similar behavior of the nuclear relaxation in other manganites.[10,16] Moreover, results obtained by Leung and Morrish[12] in $La_{0.69}Pb_{0.31}MnO_3$ at $T=4.2$ and 77 K also fall on the $T_2^{-1}(T)$ dependence (Fig. 6). It seems therefore that the exponential temperature dependence of the relaxation rates is a common feature of the ferromagnetic metalliclike manganites for temperatures down to 4.2 K. When approaching the critical region ($T \rightarrow T_C$), further enhancement of the relaxation rates is observed.

Relaxation of ^{139}La (Fig. 7) is much slower compared to ^{55}Mn. T_1^{-1} exhibits again an exponential temperature depen-

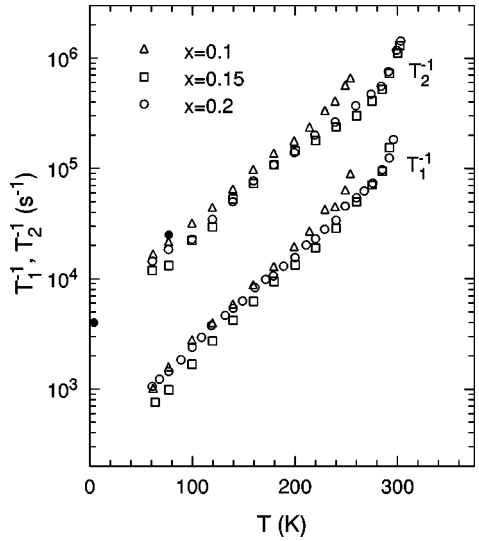

FIG. 6. $La_{1-x}Na_xMnO_3$. Temperature dependence of the relaxation rates T_2^{-1} and T_1^{-1} of ^{55}Mn. The full circles is the spin-spin relaxation in $La_{0.69}Pb_{0.31}MnO_3$ (Ref. 12).

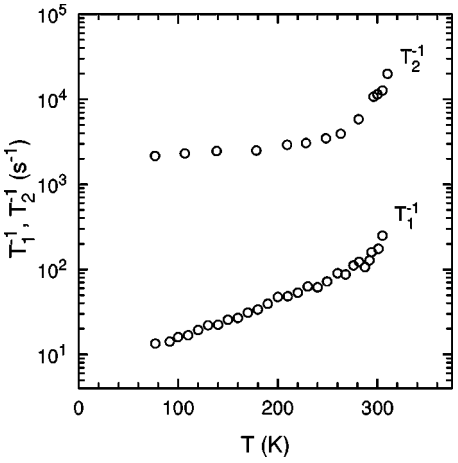

FIG. 7. $La_{0.8}Na_{0.2}MnO_3$. Temperature dependence of the relaxation rates T_2^{-1} and T_1^{-1} of ^{139}La.

dence, while T_2^{-1} remains almost constant outside the critical region. It is interesting that, except for the critical region, our results in zero external magnetic field resemble those obtained by Allodi *et al.*[9] in high field.

III. DISCUSSION

Let us examine the contributions of possible mechanisms to the nuclear spin-lattice relaxation in the system studied. In conventional ferromagnetic metals the spin-latice relaxation is dominated by a Korringa process in which the relaxing nuclear spin flips an electronic spin down. Corresponding relaxation rate is proportional to T:

$$T_1^{-1} \propto T N_\uparrow(E_F) N_\downarrow(E_F), \tag{2}$$

where $N_\uparrow(E_F), N_\downarrow(E_F)$ are the densities of states of up and down spin electrons at the Fermi energy. As our results clearly show an exponential dependence, we have to conclude that despite the metalliclike conductivity the system in question is not a conventional metal. In an ideal half-metal $N_\downarrow(E_F)=0$ at $T=0$, the Korringa process is therefore ineffective. A weak, two-magnon process[17] will then determine the relaxation at low temperatures. As T_C is approached, transition to the metallic Korringa-like behavior should occur, leading to an anomaly in the $T_1(T)$ dependence. Our results do not support this model either. We note that the case of an ''effective'' half-metal is different and it will be discussed below.

Another source of the relaxation is the fluctuation of hyperfine fields caused by the migrating electron holes — an analogous mechanism was used to explain the nuclear magnetic relaxation in magnetite[18] and in CuO:Li.[19] The general formulas[20] relating the relaxation rates T_1^{-1}, T_2^{-1} to the fluctuating local field $\delta\vec{H}$ are

$$T_1^{-1} = \frac{\gamma_N^2}{2} \int_{-\infty}^{\infty} dt \cos(\omega_0 t) \langle \{ \delta H_+(t)\, \delta H_-(0) \} \rangle, \tag{3}$$

$$T_2^{-1} = \frac{1}{2} T_1^{-1} + \gamma_N^2 \int_{-\infty}^{\infty} dt \langle \{ \delta H_z(t)\, \delta H_z(0) \} \rangle, \tag{4}$$

where γ_N is the nuclear gyromagnetic ratio and ω_0 is the nuclear magnetic resonance frequency. If the time-correlation function of $\delta\vec{H}$ is characterized by a single correlation time τ, i.e.,

$$\langle \{ \delta\vec{H}(t)\, \delta\vec{H}(0) \} \rangle \propto \exp(-t/\tau) \tag{5}$$

then from Eqs. (3),(5) follows

$$T_1^{-1} \propto \frac{\tau}{1+(\omega_0\tau)^2}. \tag{6}$$

The correlation time τ corresponding to the hopping of the electron holes varies with temperature as

$$\tau = \tau_\infty \exp(E/k_B T), \tag{7}$$

where E is the activation energy.

In the ''effective'' half-metal model of Picket and Singh[6] the minority spin electrons are localized at low temperatures. As the temperature is increased they can migrate and cause the relaxation which attains a maximum for $\omega_0\tau=1$. There exist at least two reasons, however, which make this scenario improbable.

No maximum is observed in $T^{-1}(T)$ dependence. The temperature of the maximum might be eventually outside the interval of our measurement, i.e., higher than T_C. This implies, however, $\omega_0\tau>1$ and Eq. (6) then predicts thermally activated behavior of the relaxation rate $[T_1^{-1} \propto \exp(-E/k_B T)]$ which can be hardly reconciled with our results. The disagreement remains unchanged even if a distribution of τ instead of a single correlation time is assumed.

If such slowly moving $(\omega_0\tau>1)$ carriers exist, distinct lines in the NMR spectrum should appear.[10] Indeed, the line corresponding to the concentration $c\sim 0.4\%$ of localized Mn^{4+} was observed at $f_{\rm res}\sim 330$ MHz in the spectra of $x=0.2$ sample . At the same time, for $x=0.1$ and $x=0.15$ samples, no evidence for such line was found $(c<0.02\%)$. It seems improbable that so different c will lead to a similar relaxation rate in the three compounds (Fig. 6).

As the most likely source of the relaxation we therefore propose the fluctuation of hyperfine fields caused by the rapidly hopping $(\omega_0\tau \ll 1)$ electron holes. The hopping leads to fluctuation of the Mn valency. The magnetic part of the electron-nuclear hyperfine coupling is

$$\hat{H}_{\rm hf} = \vec{S}\hat{A}\vec{I}. \tag{8}$$

The Mn^{4+} ion has the spin $S=3/2$ and the hyperfine coupling tensor $\hat{A}$ is nearly isotropic, while for the Mn^{3+} ion $(S=2)$ $\hat{A}$ is strongly anisotropic.[21,22] Fluctuation of the spin and the magnitude of $\hat{A}$ leads to the fluctuation of the longitudinal component of the local field and it is the cause of T_2^{-1} [Eq. (4)]. The mechanism responsible for T_1^{-1} arises again from the fluctuation of the spin and $\hat{A}$, but this time due to the anisotropy of the hyperfine coupling when there is an angle between the axes of quantization of $\vec{S}$ and $\vec{I}$. In the limit $\omega\tau \ll 1$, both rates are proportional to τ. Usually the hyperfine coupling tensor changes only slightly with temperature, similar form of the temperature dependence of T_1^{-1} and T_2^{-1} is then expected. The behavior of the ^{55}Mn relax-

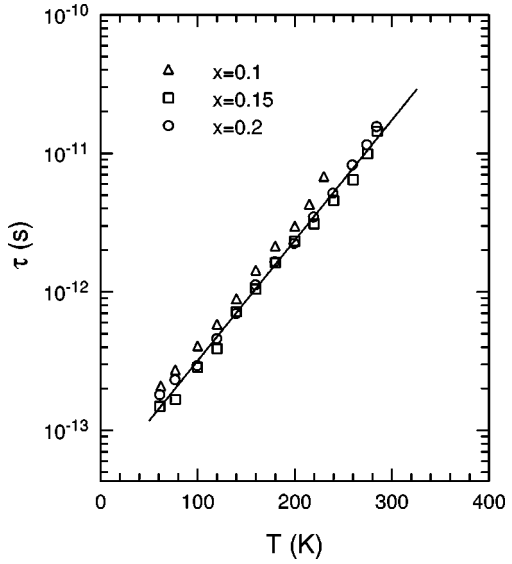

FIG. 8. $La_{1-x}Na_xMnO_3$. Temperature dependence of the correlation time for the e_g hole hopping. The line is the fit of data to $a\exp(bT)$.

ation observed experimentally (Fig. 7) can be explained in this way. There is, on the contrary, no such interconnection between T_1^{-1} and T_2^{-1} of ^{139}La (Fig. 7). This, however, can be understood from the geometry of the perovskites in question. The e_g orbitals of Mn give little contribution to the transferred hyperfine field on the La sites. In this situation the migrating e_g holes cause the fluctuation of $\delta\vec{H}$ on ^{139}La mainly through the fluctuating dipolar field. Straightforward calculation shows that for spin direction close to $\langle 100\rangle$ the fluctuation is perpendicular to the hyperfine field on La and therefore it gives rise to T_1^{-1}. This relaxation channel is relatively weak, however, and other processes might also be important. The experimental results correspond to the situation when another source, which causes weakly temperature dependent relaxation with $T_2^{-1} \gg T_1^{-1}$, adds to the relaxation caused by the migrating e_g holes.

We now turn back to the analysis of ^{55}Mn relaxation. The correlation time τ may be estimated using the Anderson model of the motional narrowing[18,23]

$$T_2^{-1} = \pi^2[f_{Mn^{3+}} - f_{Mn^{4+}}]^2\tau \simeq 8\times 10^{16}[f(T)/f(0)]^2\tau, \tag{9}$$

where T_2^{-1} is in s^{-1} and f is the NMR frequency observed. We took for $T\to 0$ $f_{Mn^{3+}} = 410$ MHz, $f_{Mn^{4+}} = 320$ MHz,[24] and identical temperature dependences of $f_{Mn^{3+}}, f_{Mn^{4+}}$, and f were assumed. Substituting experimental values of T_2^{-1} into Eq. (9), τ as a function of T was obtained. As already noted in Sec. II, the system in question is inhomogeneous, τ determined in the above way must therefore be considered as a mean correlation time. τ as a function of the temperature is displayed in Fig. 8 (the data in the critical region were left out). It is seen that the correlation time decreases with decreasing temperature. This behavior is opposite to the usual case of thermally activated hopping with fixed activation energy. It can be understood, however, if the activation energy rapidly decreases with the decreasing temperature. Such behavior occurs if the hopping e_g holes are considered as spin polarons (for thorough discussion of spin polarons see Ref. 25). In accord with the concept of spin polaron the motion of e_g holes polarizes ferromagnetically the spins of the t_{2g} electrons of the Mn ions and, on the other hand, the existence of the ferromagnetic ordering makes their hopping easier. In the fully spin-ordered state ($T\to 0$) the activation energy of the spin polaron vanishes and it has a maximum in the vicinity of T_c.[25]

There exist other experimental results pointing to the existence of spin polarons in the manganites. In a recent paper[26] the decrease of the activation energy with decreasing temperature was successfully used to explain the dependence of the resistivity on temperature and magnetic field in $Nd_{0.52}Sr_{0.48}MnO_3$. In several manganites the neutron experiments[27,28] indicated the presence of the spin polarons below T_C. We also note that the correlation time deduced from our results is consistent with the width of the quasielastic scattering peak below T_C ascribed to the spin polarons in Refs. 27,28.

For all three samples the temperature dependence of τ is approximatelly exponential (full line in Fig. 8):

$$\tau = a\exp(bT), \quad a = 4.3\times 10^{-14}\ \text{s}, \quad b = 0.02\ \text{K}^{-1} \tag{10}$$

Comparison of Eqs. (10) and (7) implies a quadratic dependence of the activation energy on temperature

$$E = bk_BT^2, \quad \tau_\infty = a. \tag{11}$$

Using Eqs. (10),(11) we obtain for the three manganites in question the activation energy at the Curie temperature $E(T_C) = 0.13 - 0.18$ eV. Above T_c a number of groups report thermally activated conduction with activation energy of the order of 0.1 eV. It is interesting to note the near equality of the two values.

It is clear that from our results we cannot conclude that the only cause of the relaxation in the system are the migrating carriers. Similarly to the effective half-metal model, another type of electron holes might exist, for which the band picture is appropriate. If, however, the holes causing the relaxation are associated with the minority $e_g\downarrow$ carriers in the half-metal, it follows from our results that (i) the minority carriers are never fully localized ($\tau < 10^{-10}$ s), (ii) they move by hopping, and (iii) the barriers are strongly temperature dependent and they increase with the increasing temperature. It is the last item which we think is difficult to comprehend within the effective half-metal model. Coey *et al.*[29] proposed the model in which the strong localization effect due to the spin disorder and nonmagnetic randomness leads to the hopping or tunneling of $e_g\uparrow$ holes between weakly localized wave packets. This or another similar scenario which takes the spin disorder and nonmagnetic randomness into account, could be more relevant to the results we obtained.

IV. CONCLUSIONS

Our study of $(NaLa)MnO_3$ manganites revealed an anomalous temperature dependence of the ^{55}Mn nuclear magnetic relaxation. The exponential dependence observed can be understood in terms of the fluctuating hyperfine field caused by the migration of the e_g electron holes. The de-

crease of the activation energy of the hopping with decreasing T can be associated with the existence of the spin polarons in the broad temperature interval. The explanation of the results within the framework of the effective half-metal would require significant modification of this model.

ACKNOWLEDGMENTS

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