LA-UR-96- 2785

CONF-9608119--5

Title:

Unusual relaxation dynamics in $La_{1-x}Ca_xMn0_3$ (x = 0.33 and 0.67)

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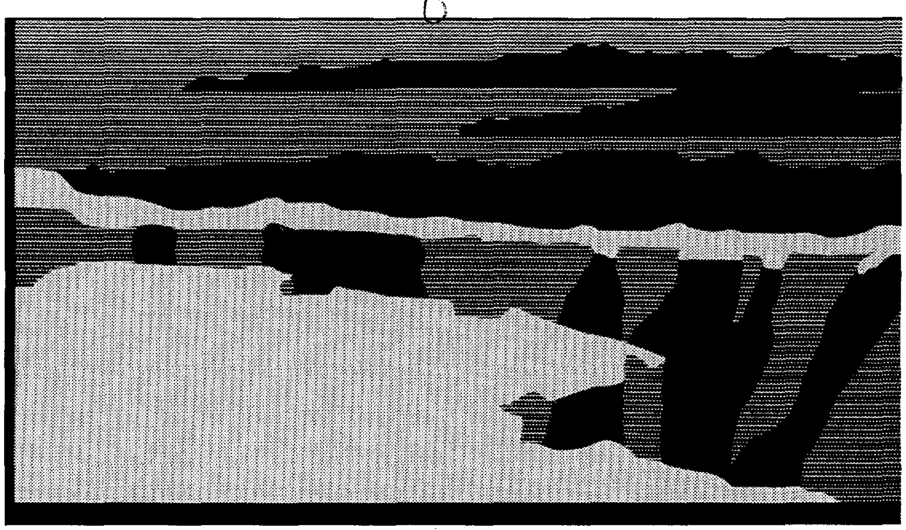
Submitted to:

International Conference on SCES 96
Zurich Switzerland
August 19-22, 1996
FULL

MASTER



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Version date: 19 July, 1996

Unusual relaxational dynamics in $La_{1-x}Ca_xMnO_3$ (x = 0.33 and 0.77)

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Muon spin relaxation studies on $La_{1-x}Ca_xMnO_3$ (x = 0.33 and 0.67) powders show that the dynamic relaxation function changes from exponential above the magnetic transition temperature to stretched exponential below the critical temperature (T_C = 274 K for x = 0.33, T_N = 150 K for x = 0.67). This suggests a spatially inhomogeneous distribution of very slow Mn-ion correlation times characteristic of diffusive relaxational dynamics. An anomalously small applied field destroys the dynamical signature of the (ferro)magnetic transition in x = 0.33, but not the (antiferro)magnetic transition in x = 0.67. The x = 0.67 system also shows evidence for a change in the magnetic phase in zero field near T = 75 K.

Keywords: muon spin relaxation, magnetoresistive oxides, spin polarons

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$LaMnO_3$ is an insulating antiferromagnet (AFM) with a perovskite structure [1]. As Ca^{2+} is substituted for La^{3+}, charge conservation requires Mn^{3+} conversion to Mn^{4+}, resulting in FM correlations for $0.2 \leq x \leq 0.5$. Above T_C the system is insulating but below T_C the system becomes metallic [2]. There is growing evidence that polaron formation in these materials plays an important role in the charge and heat transport [3], but the precise nature of these polarons has yet to be determined. In the $x = 0.67$ material, for example, both charge ordering and magnetic ordering have been observed [4]. Present interest [5] in these systems therefore stems from their interplay between magnetism and electronic degrees of freedom. Here we report on the Mn-ion dynamics in two systems.

Time-differential μ^+SR experiments were carried out using the surface muon M15 channel at TRIUMF in Vancouver, Canada, and the General Purpose Spectrometer at the Paul Scherrer Institute in Villigen, Switzerland. The samples were pressed pellets of polycrystalline $La_{1-x}Ca_xMnO_3$. Sample quality was investigated using electron microprobe analysis to search for possible atomic clustering; no such evidence was found at the level of 5 %.

The zero- and longitudinal-field μSR data are in general described by a relaxation function given by:

$$G(t) = A_1 \exp[-\lambda t] \cos(2\pi\nu_\mu(T)t + \varphi) + A_2 \exp[-(\Lambda t)^K], \qquad (1)$$

where $A_1 + A_2 = 1$. Here $\nu_\mu(T)$ is the muon precession frequency, proportional to the sublattice magnetization below T_C, λ is the inhomogeneous linewidth, and Λ is the dynamic spin-lattice relaxation rate for a stretched-exponential relaxation function. Unlike previous μSR studies on orthoferrites [6], we found no evidence for either multiple stopping sites or muon diffusion in the temperature range studied, probably due to disorder-induced localization. We were able to observe zero-field muon precession below T_C for $x = 0.33$, but not below the antiferromagnetic transition in $x = 0.67$, most likely because of a very large inhomogeneous linewidth λ in the latter compound.

In a previous work [7] we showed that the order parameter $\nu_\mu(T)$ for $x = 0.33$ is well described by the function $\nu_\mu(T) = (1 - T/T_C)^\beta$, where $\beta = 0.345 \pm 0.015$ and $T_C = 274.3 \pm 1.4$ K. Theoretical values of the critical exponent for 3D Heisenberg, XY and Ising systems are 0.38, 0.33, and 0.31, respectively [8], while a value of 1/2 corresponds to a mean-field transition. The measured value of β is thus close to the theoretical critical value expected for a 3D spin system.

Figure 1 shows the dynamical relaxation rate Λ as a function of temperature and longitudinal (along the muon spin) magnetic field for $x = 0.33$. One observes a peak in $\Lambda(T)$ for zero applied field corresponding to the critical slowing down of the local field fluctuations for $T \geq T_C$. Below T_C Λ decreases because of the decreasing amplitude of the local-field fluctuations as the Mn spin system freezes. The dynamical fraction of the relaxation function (with amplitude A_2 in Eqn. (1)) changes from an exponential ($K = 1$) near 300K to a 'root-exponential' ($K = 1/2$) at $T \approx T_C$ (see inset to Fig. 1), and is well described using $K = 1/2$ for $150\ \text{K} < T < T_C$. Below about 150 K the relaxation rate is too small to distinguish unambiguously between the exponential and non-exponential forms. Most ferromagnetic materials studied by μSR show an exponential relaxation function above and below T_C, with a rate which approaches divergence at T_C [9,10]. Furthermore, in standard ferromagnets like $GdNi_5$ [9] both the magnitude of the relaxation rate and its temperature dependence at low temperatures ($T^2\ln(T)$, $T << T_C$) are consistent with those expected [11] for a two-magnon scattering process. Similar behavior has also been observed in the random ferromagnet *Pd*Mn (2 at. %) alloy, with $T_C = 5.8$ K [10].

As described previously [7], the measured relaxation rate Λ for x = 0.33 is 3-4 orders of magnitude larger than the two-magnon process expected [11] for an ordinary ferromagnet. Furthermore, the observation of a non-exponential relaxation function for 150 K < $T \leq T_C$ suggests that the spin dynamics are 'glassy' , i.e., cannot be characterized by a single Mn-ion correlation time. These anomalously slow spin fluctuations and the non-exponential relaxation below T_C imply diffusive spin motion [7], which is to be contrasted with the ordinary spin waves observed in more typical ferromagnets.

The relaxation rate Λ for x = 0.33 is strongly suppressed in very modest applied fields; as seen in Fig. 1, a field of only 3 kOe completely destroys the peak in Λ at T_C. For comparison, μSR experiments [10] in *Pd*Mn (2 at. %), a random ferromagnet with a giant Mn moment $\mu_{eff} \approx 8\mu_B$, show a suppression of the peak in $\Lambda(T)$ near T_C ($\approx$ 5 K) in $H_{app} = 5$ kOe applied field. In this case $\mu_{eff}H_{app} \approx (1/2)k_BT_C$, so that the suppression of the ferromagnetic fluctuations for H_{app} = 5 kOe is not surprising. However, in $La_{0.67}Ca_{0.33}MnO_3$, where $\mu_{eff} \approx 3.7$ μ_B/Mn-atom, one has $\mu_{eff}H_{app} \approx (1/360)k_BT_C$ for H_{app} = 3 kOe. Thus even if there are large ferromagnetic clusters (so that $\mu_{eff} >> 3.7$ μ_B), this extreme sensitivity to small fields is indeed very unconventional.

We now turn to the experiments on the x = 0.67 compound. Figure 2 shows the zero-field relaxation rate Λ as a function of temperature, with the inset showing the behavior of K(T). The temperature dependence of $\Lambda(T)$ indicates a magnetic transition near 150 K (denoted by a small peak at this temperature), followed by evidence for another possible magnetic transition near 75 K, where $\Lambda(T)$ again goes through a maximum. As for x = 0.33 one sees a change from an exponential relaxation function (K $\approx$ 1) at elevated temperatures to a stretched exponential function (K $\approx$ 1/2) near and below 150 K. Specific heat, transport and diffraction data [4] in this material give evidence for a charge-ordering transition at about 265 K, followed by a broad magnetic transition at about 150 K. Thus our relaxation rate data confirm the magnetic transition at about 150 K, but also show a change in the Mn-ion dynamics at lower temperatures, giving evidence for an additional magnetic transition.

Figure 2 also shows the behavior of $\Lambda(T)$ in an applied field of 3.5 kOe for x = 0.67. Remarkably, the spin lattice relaxation rates between 50 - 150 K *increase* with applied field, in stark contrast to the field-induced suppression of the transition for the x = 0.33 material. The origin of this behavior is under investigation.

In conclusion, the relaxational dynamics below the ferromagnetic transition temperature in x = 0.33 are indicative of a very slow, spatially inhomogeneous relaxation process, suggesting diffusive spin motion, unlike ordinary ferromagnets, where spin-wave excitations dominate. These spin fluctuations are unexpectantly damped in very weak applied fields. The zero-field spin dynamics in the x = 0.67 system give evidence of two transitions, an antiferromagnetic transition at 150 K, seen in specific heat data [4], and a previously unreported transition near 75 K.

Work at Los Alamos was performed under the auspices of USDOE; work at UC Riverside and at Columbia University was supported in part by the USNSF (DMR-94-18991 and DMR-95-10454, respectively). Work at the University of Leiden was supported in part by FOM.

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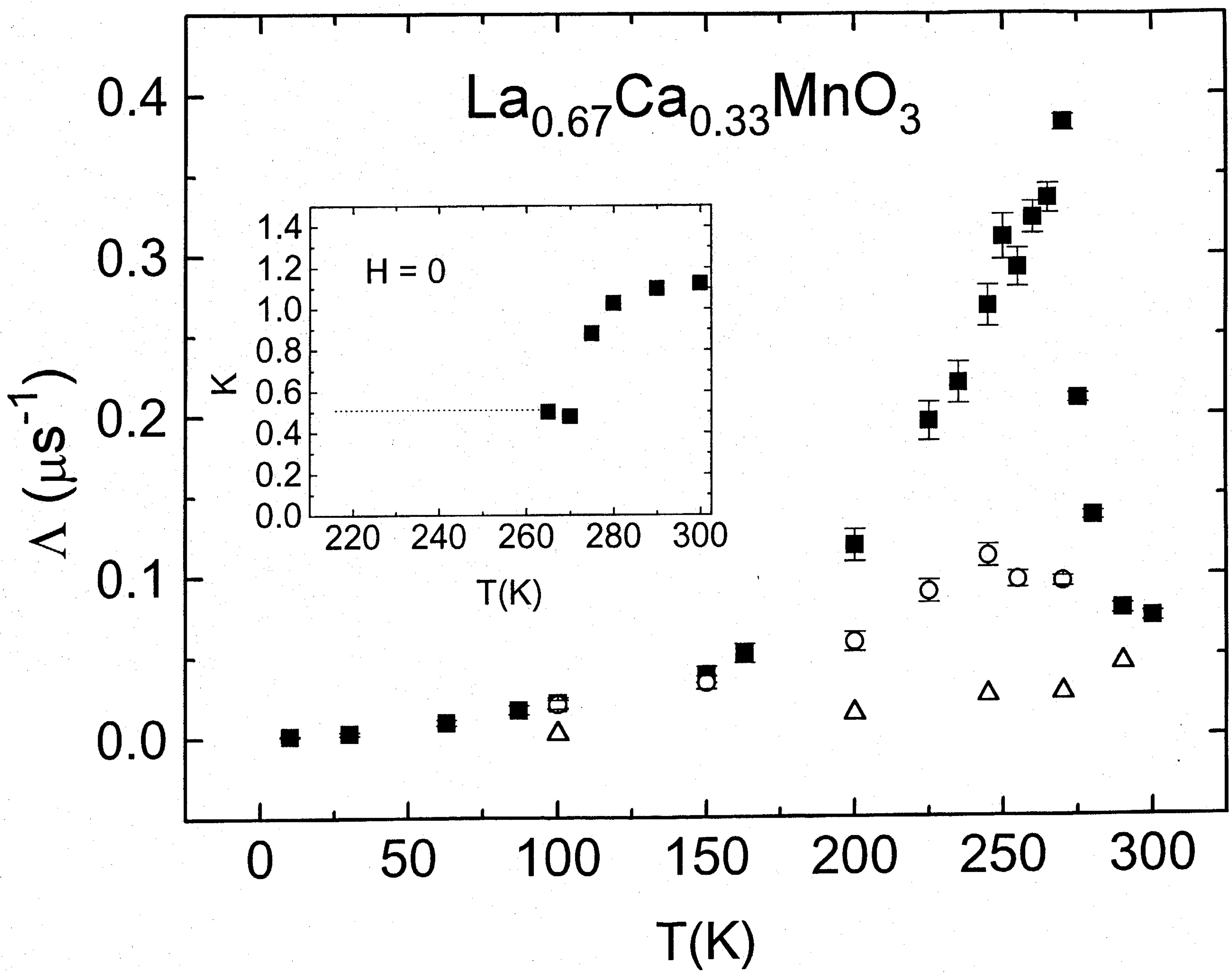
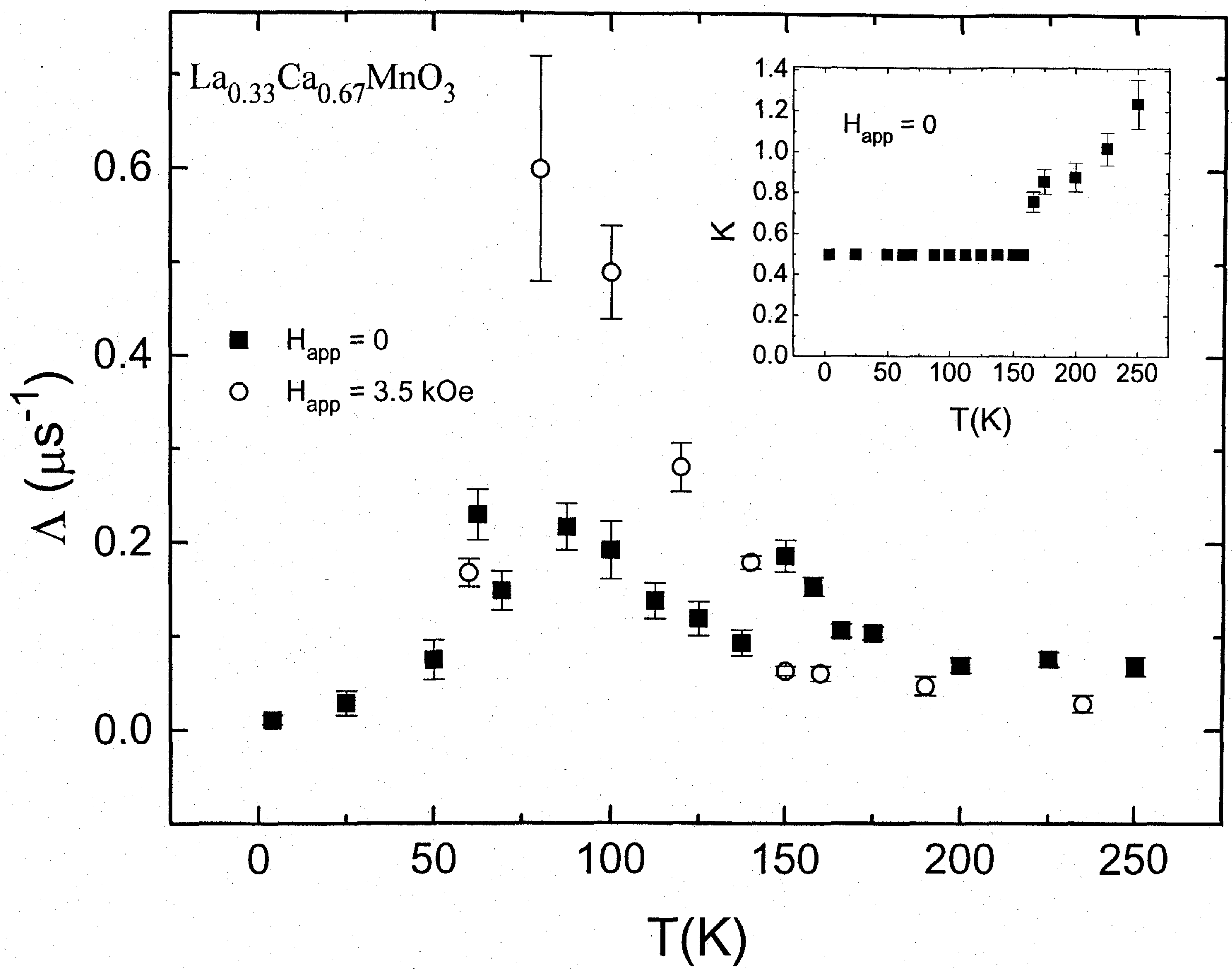
Figure Captions.

Figure 1. Temperature dependence of dynamic μSR spin-lattice-relaxation rate Λ in $La_{0.67}Ca_{0.33}MnO_3$ for zero applied field (filled squares), and longitudinal applied fields of 1 kOe (open circles) and 3 kOe (open triangles). The inset shows the stretched exponent K (see Eqn. 1) in zero field.

Figure 2. Temperature dependence of zero- and longitudinal-field μSR relaxation rate Λ and stretched exponent K (see Eqn. 1) in $La_{0.33}Ca_{0.67}MnO_3$.



$La_{0.67}Ca_{0.33}MnO_3$
Λ (μs⁻¹)
T(K)
H = 0
K
T(K)



$La_{0.33}Ca_{0.67}MnO_3$
$H_{app} = 0$
$H_{app} = 3.5$ kOe
Λ (μs^{-1})
0.0
0.2
0.4
0.6
T(K)
0
50
100
150
200
250
K
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0