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Temperature dependence of the muon spin relaxation in $Pr_{1/2}Sr_{1/2}MnO_3$

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Abstract

The temperature dependence of the positive muon spin relaxation rate, $\lambda(T)$, in $Pr_{1/2}Sr_{1/2}MnO_3$, measured for zero and transverse fields, strikingly resembles that of magnetization $M(T)$, showing successive transitions from a paramagnetic insulating to a ferromagnetic metallic (FMM) and subsequently to an antiferromagnetic insulating state. In the FMM regime, between $T_C \cong 259$ K and $T_N \cong 134$ K, maximum shifts in λ and in M seem to scale with applied fields. Dynamic local field variations averaged over the muon lifetime may be responsible for the λ temperature dependence.

Keywords: Manganites; μSR; CMR

In spite of extensive work recently published on perovskites focused on colossal magnetoresistance (CMR) behavior, very few muon spin rotation (μSR) investigations have yet been reported on these materials [1,2]. We present preliminary μSR results on the $Pr_{0.5+y}Sr_{0.5-y}MnO_3$ manganite series which, depending on stoichiometry or on additional doping elements, may undergo two magnetic transitions. Indeed, with decreasing temperature from 300 K, a double transition has been observed [3,4] first from a paramagnetic insulating (PMI) to a ferromagnetic metallic (FMM) and subsequently from a ferro to an antiferromagnetic insulating (AFMI) state, the latter possibly also involving orbital ordering.

Prepared by high-temperature powder sintering, our 5 mm thick $Pr_{1/2}Sr_{1/2}MnO_3$ sample had a slightly truncated cone shape (diameters: 12 and 9 mm) [3]. For the classic transverse field setup (TF) used in the longitudinally spin polarized μE4 beam channel at the Paul Scherrer Institute, such a thickness limited the useful muon momentum, hence the maximum applied field to $B \approx 300$ mT. After earth-field cancellation, the temperature dependence of the muon spin relaxation rate was first measured during zero-field cooling (ZFC), in a Janis helium-flow cryostat, from room temperature to 15 K. Subsequent investigations were performed while increasing the temperature, with TF applied at 15 K after ZFC (i.e., far below the FMM transition) in order to tentatively induce higher magnetic order in the antiferromagnetic state while trying to minimize the influence of possible hysteresis effects due to the ferromagnetic transition. 10 and 300 mT TF-μSR temperature dependences were

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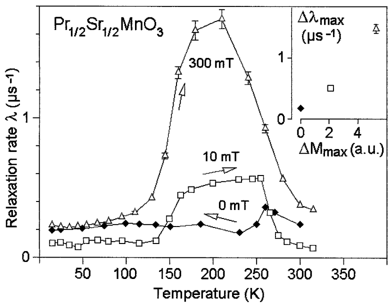

Fig. 1. Temperature dependence of the relaxation rate (λ) in $Pr_{1/2}Sr_{1/2}MnO_3$ for various fields applied after ZFC (arrows refer to measurements during cooling or warming). Inset: maximum changes in λ and in M around T_C for identical fields (except for M at 1 mT).

measured in warming after independent ZFCs (Fig. 1). Additional magnetization measurements were later performed on the same sample after ZFC at equivalent fields (1, 10 and 300 mT), using the CRISMAT-Caen magnetometer (Foner) [3].

The relaxation rate, λ, was extracted from Lorentzian fits to the time-dependent polarization:

$$P_x(t) = P_0 \exp(-\lambda t)\cos(2\pi v_\mu t + \phi),$$

where v_μ and ϕ are the Larmor frequency and phase shift, respectively. During the first ZFC (Fig. 1), $\lambda(T)$ displays a small sharp peak around 260 K indicating the occurrence of a possible local rearrangement of magnetic moments at a temperature where the ferromagnetic transition is expected. However, no oscillations superimposed on the muon decay spectra could be detected over the whole temperature scan, in spite of a reasonably high statistics data collection ($\sim 6.10^6$ events/spectrum), suggesting rather poor long- and short-range magnetic orderings. For both non-zero field values (Fig. 1), the overall temperature dependence of the relaxation rate, $\lambda(T)$, displays a "pulse-shape" behavior similar to that of magnetization curves $M(T)$, observed for both "pure" and Ca doped $Pr_{1/2}Sr_{1/2}MnO_3$ manganites [3,4]. At low-field (10 mT), $\lambda(T)$ exhibits a stronger asymmetrical shape, i.e., its slope associated with the FMM–PMI transition is much steeper than that of the AFMI–FMM transition. While approaching the former transition from the paramagnetic state, the relaxation time (λ^{-1}) seems to follow a Curie-type law with an onset temperature, $T_C = 259 \pm 1$ K, slightly lower than the value quoted by Wolfman et al. [3]. For 300 mT, a Curie transition broadening occurs, making accurate T_C extrapolation more difficult. The Néel temperature (T_N) of the AFMI-FMM transition, observed by Damay et al. [4], corresponds to the appearance of the antiferromagnetic Fmmm phase in the neutron powder diffraction (NPD) pattern, whereas the smooth variation of the muon spin relaxation rate renders the determination of T_N somewhat uncertain. Using the empirical expression: $\Delta\lambda/\Delta\lambda_{max} = 1 - A\exp(-\alpha T)$, where $\Delta\lambda_{max}$ is the maximum change occurring between T_N and T_C and A, α are constants, the λ onset for $B = 10$ mT is at 134 ± 1 K, a temperature in excellent agreement with T_N derived from NPD data. For $B = 300$ mT, the temperature of the $\lambda(T)$ "pulse" onset is far less obvious though only lower by a few degrees.

It is worth noting that, at equivalent fields, $\Delta\lambda_{max}$ scales with ΔM_{max} almost linearly (Fig. 1 inset). Considering the nature of both probes, i.e., global for the magnetization as opposed to local for the muon, we infer that, in this material, long- and short-range magnetic orders are strongly correlated and equally affected by external field. Such scaling of the relaxation rate with field has already been reported for EuPdAs in its paramagnetic valence-fluctuation regime [5]. A dynamic behavior for which local field variations are averaged over the muon lifetime would explain why no spontaneous precession was found, as observed for instance in $LaCrO_3$ at 130 K [6]. In spite of a monotonic temperature dependence exhibited, at first glance, by both electrical resistivity and magnetization in the 15–130 K range [3], particular attention was paid to a slight offset in λ ($0.05 \pm 0.01 \mu s^{-1}$) occurring at 50 K ($\approx 0.38 T_N$) for 10 mT and suppressed at high field. A careful scanning of $M(T)$ indeed reveals a similar discontinuity in the magnetization with similar sensitivity to high field. Such a feature also appears in the $M(T)$ curve for Mn_3O_4, but tends to develop with increasing field. Hence, assigning the origin of the λ(50 K) discontinuity to an impurity effect is somewhat debatable.

Further μSR investigations are needed to determine whether the similarity of both $M(T)$ and $\lambda(T)$ temperature dependences reflects the occurrence of spin ordering and fluctuation or whether it results from a sample-dependent sub-micron domain configuration induced by structural changes [4] and contributing to the widening of the local field distribution.

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