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Investigation of Rare-Earth Manganates and Manganites by the μ*SR*-Method

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Abstract—The present paper is devoted to studying the multiferroics $HoMnO_3$, $YMnO_3$, $EuMn_2O_5$, and $GdMn_2O_5$ by means of the μ*SR*-method. Determination of the dynamic relaxation parameter λ and the distribution of the local magnetic fields results in a clear phase diagram.

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INTRODUCTION

There has in recent years been the great interest in multiferroics. These are substances in which magnetic and electric orderings coexist [1–3]. On the one hand, creating devices that transform information in the form of magnetization into electric signal and back on the basis of a single material is a very attractive solution of the urgent tasks of sensor technology, magnetic memory, and microelectronics. This is especially true for spintronics, which is aimed at combining the advantages of nonvolatile magnetic memory and fast systems of data processing. On the other hand, the experience accumulated since the first multiferoics were discovered in the early 1960s allows us to produce materials that exhibit strong magneticelectrical properties under normal conditions. We are now seeing a true renaissance in the area of magnetocelectrical (ME) research, expressed in the appearance of sections devoted to multiferroics at symposia on magnetism, the holding of special conferences, and the exponential growth of publications on this topic.

$RMnO_3$ manganites exhibit a wide variety of physical properties depending on rare-earth element R. The compounds with a large ion radius of element R (La, Pr, Nd, Sm, Eu, Gd, and Tb) crystallize in an orthorhombic structure with spatial group *Pnma* [4]. In compounds with a shorter ion radius of element R (Ho, Er, Tm, Yb, Lu, Y, Sc, and In) we can observe a hexagonal crystal structure with spatial group $P6_3cm$ [5]. Hexagonal manganites belong to a class of ferroelectromagnetics, in which the temperature of transition to ferroelectric state T_C ~ 600–900 K is much higher than temperature of antiferromagnetic (AFM) ordering T_N ~ 70–130 K [6].

RMn_2O_5 manganates (R are rare-earth ions from Pr to Lu, Y, or Bi) are magnetoelectrics having simultaneous antiferromagnetic and ferroelectric long-distance order with close Neel and Curie temperatures $T_{N,C}$ ~ 30–40 K (at room temperature with spatial group *Pbam*) [7].

This paper presents the results of investigations of the magnetic properties of multiferroics $HoMnO_3$, $YMnO_3$, $EuMn_2O_5$, and $GdMn_2O_5$ by means of the muon spin rotation method (below, the μ*SR*-method). Determination of the dynamical relaxation parameter λ and the distribution of the local magnetic fields resulted in a clearer phase diagram. The measurements were done on the muon channel of the PNPI synchrocyclotron, using the μ*SR*-installation [8].

EXPERIMENTAL

The experiment involved measuring the time distributions of positrons $N_e(t)$ produced upon μ^+-meson ($\mu^+ \to e^+ + \nu_e + \tilde{\nu}_\mu$) decay and escaping in the direction of the initial muon polarization in time interval $\Delta t \sim 4.5\ \tau_\mu$ after the moment when each muon in the sample stops, where τ_μ is the muon lifetime. The time distribution of the positrons is described by the expression:

$$N_e(t) = [N_0\exp(-t/\tau_\mu)][1 + a_sG_s(t) + a_bG_b(t)] + \Phi, \quad (1)$$

where N_0 is a normalization constant (which is proportional to registered positrons); $\tau_\mu \approx 2.19711 \times 10^{-6}$ s is the muon lifetime; a_s and a_b are the initial decay assymetry of muons stopped in the sample (a_s) and its background component (a_b) from muons stopped in the input windows of the cryostat and start counter of the muon detector; $G_s(t)$ and $G_b(t)$ are the relaxation functions of polarization for the muons stopped in the sample and background sources; and Φ is the background of random coincidences.

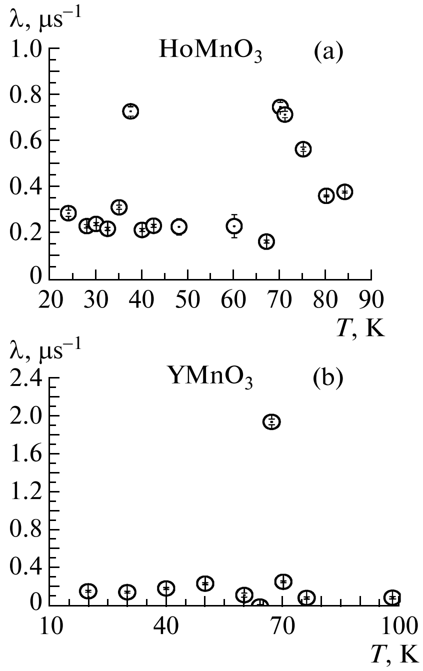

Fig. 1. Temperature dependence of the relaxation rate of stopped muon polarization in (a) $HoMnO_3$ and (b) $YMnO_3$ samples in a zero magnetic field.

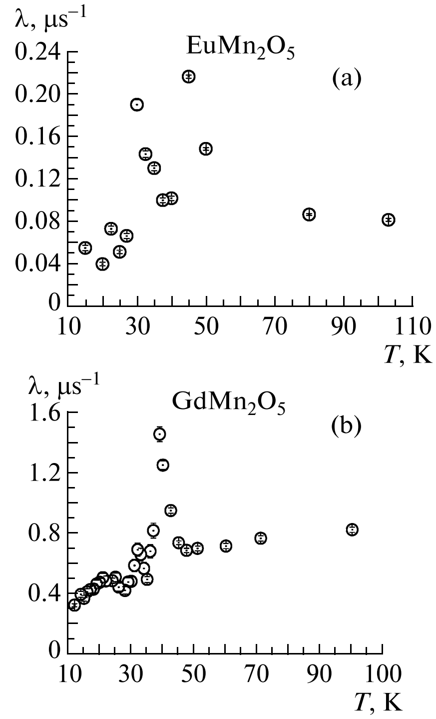

Fig. 2. Temperature dependence of the relaxation rate of stopped muon polarization in (a) $EuMn_2O_5$ and (b) $GdMn_2O_5$ sample in a zero magnetic field.

Factorization of the relaxation function is used in analyzing the experimental data:

$$G_s(t) = G_d(t) \times G_{st}(t), \quad (2)$$

where $G_d(t)$ describes the relaxation due to the dynamic effects, and $G_{st}(t)$ is the function of relaxation in static fields. In the investigation of dynamic phenomena leading to relaxation of the spin of muons, the relaxation function is determined by the dependence

$$G_d(t) = \exp(-\lambda \times t), \quad (3)$$

where λ is the rate of dynamic relaxation.

Figure 1a shows the dependence of the relaxation rate λ of stopped muon in the $HoMnO_3$ sample on the temperature. We can see two peaks (at 76 and 40 K), which correspond to two phase transitions. The first one (T_N = 76 K) is the transition from the paramagnetic state to the state of antiferromagnetic ordering. The second one ($T \approx 40$ K) is associated with a 90° turn of Mn spins (a spin–rotational transition). This is agreed with the results obtained by other methods [9, 10].

Similar investigations were performed with the $YMnO_3$ sample. In the behavior of the dependence of relaxation rate λ for the polarization of muons stopped in the $YMnO_3$ sample on the sample temperature, we can see the peak at temperature $T_N \approx 66$ K that corresponds to the phase transition paramagnetic–antiferromagnetic (Fig. 1b) [11].

Figure 2 shows the temperature dependence of relaxation rate λ for the polarization of muons, stopped in the samples of monocrystals $EuMn_2O_5$ (Fig. 2a) and $GdMn_2O_5$ (Fig. 2b). The behavior of relaxation parameter λ allows us to determine the temperature value of the phase transition, there is a sharp peak in the λ values near this point, due to critical fluctuations. There are two distinct peaks for the $EuMn_2O_5$ sample ($T_N \sim 40$ K and $T_C \sim 30$ K) that correspond to the two transitions found out by other methods. Near ~22.5 K, there is a sign of a third transition that will be studied further. For the $GdMn_2O_5$ sample, we can clearly see the antiferromagnetic transition near $T_N \sim 40$ K and two more transitions at $T_C \sim$ 30 and 22.5 K, as with the $EuMn_2O_5$. The samples' rates of relaxation for the polarization of stopped muons appeared to be different, especially for the $GdMn_2O_5$ sample; Fig. 2b shows that it is much higher.

CONCLUSIONS

The narrow width of the peaks of dynamic relaxation λ near the transition temperatures gives evidence to the homogeneity of the studied samples, which allowed us to perform more detailed μ*SR*-investigations of these samples in temperature range 10–70 K (depending on the sample) in a zero external magnetic field.

For all of the studied samples, the dependence of the precession frequency on the temperature is well approximated by the Curie–Weiss curve $F \sim F_{max}(1 - T/T_N)^{\beta}$ with the exponent $\beta = 0.39 \pm 0.02$, which corresponds to a model of a 3D-magnetic of the Heisenberg type.

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