Muon-Spin-Relaxation Investigation of EuMn$_2$O$_5$

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The magnetic properties of the EuMn$_2$O$_5$ multiferroic (samples consisting of single crystals and ceramic samples) have been investigated by the muon-spin-relaxation (μSR) method in the temperature range of 10–300 K. Below the magnetic ordering temperature $T_N = 40$ K, the loss of the polarization of muons and the effect of the external magnetic field have been observed. Both phenomena can be explained by an additional channel of depolarization of muons owing to the appearance of muons in a medium with a low electron density due to the charge separation process (the redistribution of the electron density in the phase transition process). The “memory” phenomenon has been revealed in a sample in the external magnetic field; the memory relaxation time depends on the size of the structure units of the samples (single crystals or ceramic grains).

Multiferroics are promising materials for the production of elements of devices transforming magnetic signals into electric ones. This property is due to the large magnetoelectric effect observed in a number of compounds, in particular, in RMn$_2$O$_5$ manganites (R is a rare-earth element) in which the antiferromagnetic and ferroelectric orders coexist at low temperatures [1, 2]. Their crystal structure corresponds to an orthorhombic lattice and was studied in, e.g., [3].

Most RMn$_2$O$_5$ manganites have three magnetic subsystems (R$^{3+}$, Mn$^{3+}$, and Mn$^{4+}$). One of the exclusions is the EuMn$_2$O$_5$ compound, where the Eu$^{3+}$ ion is nonmagnetic in the ground state, and its magnetic properties are determined only by the Mn$^{3+}$ and Mn$^{4+}$ ions. A sample is paramagnetic at high temperatures. At the Néel temperature $T_N = 40$ K, the Mn$^{3+}$ and Mn$^{4+}$ ions are ordered and a long-range magnetic order appears. The effect of the magnetic scattering of neutrons is observed [4] and, simultaneously with a weak anomaly in the magnetic susceptibility $\chi$ [1], a sharp peak of the dielectric constant $\varepsilon$ [1, 5, 6], and a jump in the intensity of the passage of millimeter electromagnetic radiation [6]. The analysis of the data indicates that this structure phase transition is initiated by the rotation of the magnetic moments of the Mn$^{3+}$ and Mn$^{4+}$ ions and by the shift of the Mn$^{4+}$ ions along the $c$-axis.

A wide anomaly in the temperature dependence of the dielectric constant $\varepsilon(T)$ was also revealed in the temperature range $T = 13–23$ K [1, 4–6], which is correlated with the bends in the temperature dependence of the magnetic susceptibility $\chi(T)$ and with the features of the magnetic scattering of neutrons. These effects can imply the existence of magnetic and structural inhomogeneities.

Unusual phenomena were also revealed at high temperatures (at 100–130 K), but they likely have another nature and are attributed to the existence of limited correlated magnetic regions [5].

The experimental data considered above demonstrate a strong correlation between the electric and magnetic phenomena (magnetoelectric effect) and indicate the relation between the structural and magnetic properties.

In this work, the comparative muon-spin-relaxation (μSR) investigation of EuMn$_2$O$_5$ samples of two types (consisting of single crystals and ceramic grains) is performed in the temperature range of 15–140 K and indicates the qualitative difference between their properties. The time spectra of decay positrons are measured and used to obtain the temperature depen-
dences of the relaxation parameters of the muon polarization, the frequencies of the muon spin precession in the internal magnetic fields of the samples, and the partial contributions of various precession modes to the total asymmetry.

The samples were studied on a setup mounted at the exit of the muon channel of the synchrocyclotron at the Petersburg Nuclear Physics Institute, Russian Academy of Sciences. The setup, the procedure of the measurement of the μSR spectra, the processing of the spectra, and the procedure of obtaining the relaxation parameters of the muon polarization; the frequencies of the muon spin precession in the internal magnetic fields of the samples; and the partial contributions to the total asymmetry of the muon decay were described in detail in [8, 9]. Polarized μ+ -muons having the momentum distribution with a mean momentum of \( p_0 = 90 \text{ MeV}/c \) and a FWHM of \( \Delta p/p = 0.02 \) stopped in a target, which was a container with 2–3-mm single crystals randomly oriented with respect to the muon spin or a ceramic sample manufactured by solid-phase synthesis with a diameter of 35 mm and a thickness of about 5 mm. The sample was placed in a cryostat, where the necessary temperature in the range under investigation was maintained with an accuracy of \( \pm 0.2 \text{ K} \).

The time spectra of decay positrons were measured in long (~10 μs \( \approx 4.5\tau_p \), where \( \tau_p = 2.19711 \text{ μs} \) is the muon lifetime) and short (~1–1.1 μs) ranges with channel steps of 4.9 and 0.8 ns, respectively. These spectra were approximated by the expression

\[
N_0(t) = N_0 \exp(-t/\tau_p) [1 + a_s G_s(t) + a_b G_b(t)] + B, \tag{1}
\]

where \( N_0 \) is the normalization constant proportional to the number of detected positrons; \( a_s \) and \( a_b \) are the contributions to the observed decay asymmetry from muons stopped in the sample and the construction elements of the setup, respectively; \( G_s(t) \) and \( G_b(t) \) are the respective polarization relaxation functions; and \( B \) is the contribution from the random coincidence background. The parameter \( a_b \) is determined from the processing of the time spectrum measured in the external magnetic field at the sample temperature below the Néel temperature (~25 K). The parameter \( a_s \) can depend on the sample temperature if the variation of the temperature is accompanied by the appearance of additional polarization loss channels (e.g., the formation of muonium with the subsequent fast depolarization).

The polarization relaxation function can usually be represented in the form

\[
G_s(t) = G_0(t) G_w(t). \tag{2}
\]

Such a representation is valid beyond the region of critical fluctuations and phase transitions. The dynamic function \( G_0(t) = \exp(-\lambda_0 t) \) describes the average spatial fluctuations of the internal magnetic fields of the sample at large distances (about the muon diffusion length). The form and parameters of the static function \( G_w(t) \) are determined by the structure of the sample at a given temperature and, as a result, by the distribution of magnetic fields in regions preferable for the localization of muons after their deceleration in the sample and the end of their thermalization process.

The static relaxation function of the muon polarization for magnets below the temperature \( T_N \) of the paramagnetic—antiferromagnetic transition has the form

\[
G_w(t) = 1/3 + 2/3 \cos(\Omega t) \exp(-\Delta t), \tag{3}
\]

where \( \Omega = 2\pi F \) is the frequency of the muon spin precession in the local magnetic field acting on the muon and \( \Delta \) is the rate of precession damping due to the spread of the local magnetic fields. It is easy to see that in the limiting cases, \( G_w(t) = 1 \) (in the paramagnetic state) and \( G_w(t) \approx 1/3 \) (if the sample is in a magnetically ordered state and other depolarization channels are absent) at times \( t \gg 1/\Delta \) when the static relaxation of the polarization can be neglected.

From the processing of the experimental data in the time range \( t = 0.6–10 \text{ μs} \), where the effect of static magnetic fields is negligibly small, the dynamic relaxation rate \( \lambda_d \) and the residual asymmetry \( a_s \) were obtained throughout the temperature range under investigation. The initial asymmetry \( a_0 \) was determined from the processing of the measured time spectra of the muon spin precession in the external magnetic field at temperatures \( T > T_N = 40 \text{ K} \) when the sample is paramagnetic. The results are presented in Figs. 1 and 2. The ratio \( a_s/a_0 \) is also shown in Fig. 2.

The temperature dependences of the parameter \( \lambda_d \) are the same for both samples and exhibit a sharp peak at the temperature \( T_N \approx 40 \text{ K} \), which is attributed to the paramagnetic—antiferromagnetic phase transition.

Anomalies in the behavior of \( \lambda_d \) are observed at temperatures of 30 K (FE1 transition) and 22 K (FE2 transition).

The behavior of the residual asymmetry (see Fig. 2) remains almost unchanged above the Néel temperature \( T_N \approx 40 \text{ K} \). However, the residual asymmetry \( a_s/3 \) below the Néel temperature \( T_N \) is much smaller than \( a_0/3 \), indicating the appearance of the additional channel of muon depolarization, e.g., on the formation of long-lived muonium Mu (the \( \mu^+e^- \) bound state) in the samples. Muonium is rapidly depolarized and, thus, the observed asymmetry is partially lost. However, for the formation of muonium, it is necessary to change the electron (charge) density. The charge density decreases at the sites where muons prefer to stop. The redistribution of the charge density is