Muon depolarization during the magnetic phase transition in V_2O_3

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The temperature dependence of the residual polarization of negative muons in the mesic oxygen atom produced in V_2O_3 is investigated by the method of spin precession in a magnetic field. The dependence is observed to undergo a jump at the Néel temperature. The relaxation of the spin of $\mu^{\pm}$ mesons in V_2O_3 is also obtained.

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In recent years investigations of the residual polarization of muons stopped in solids has attracted much interest.[1–6] In the present study we investigated the change of the residual polarization of negative muons captured by oxygen atoms of the oxide V_2O_3, in the temperature interval 77–300 K. We measured also the residual polarization of μ^+ mesons at temperatures 77 and 300 K.

The oxide V_2O_3 is known to undergo a phase transition near the temperature $T_N = 168$ K (the Neel temperature).[7,8] Above this temperature, V_2O_3 is a conductor and has paramagnetism, whereas below 168 K it is an antiferromagnet and its conductivity decreases by a factor 10^6. During the phase transition the magnetic susceptibility undergoes a jump and the crystal structure of this oxide changes.

The experiments were performed with the separated muon beam from the proton synchrotron of the Laboratory for Nuclear Problems of our Institute. The apparatus for the measurements and the data reduction we described earlier.[1,4] The polarized muons were stopped in a target of polycrystalline V_2O_3 of thickness 3.7 g/cm^2, placed in a transverse magnetic field of approximate intensity 120 Oe. The target was placed in a cryostat in which the temperature was maintained accurate to $\pm$ 1 K. The electrons from the decay of muons in the mesic atoms of oxygen and vanadium were separated in accordance with the muon lifetimes. The distribution $N(t)$ of the electrons from the muon decay in the oxygen (after taking into account the time-constant background and the exponentials due to the lifetime of the mesic atoms) is of the form

$$N(t) \sim [1 + a \exp(-t\lambda_r) \cos(\omega t + \delta)],$$

where a is the asymmetry coefficient of the electrons from the muon decay in the mesic oxygen atom at $t = 0$ (time of stopping of the muon in the target), λ_r is the muon-spin relaxation rate in the mesic atom, ω is the frequency of the Larmor precession of the quasi-free spin of the muon in the transverse magnetic field, and δ is the initial phase of the precession. The temporal spectra of the decay electrons were processed with a computer by least squares.

The measured values of $\bar{a}$ for the μ^- mesons are shown in the figure relative to a_C in graphite. In the case of positive muons it was found that $\bar{a}/a_{br} = 0.77 \pm 0.01$ at 77 K and $\bar{a}/a_{br} = 0.19 \pm 0.02$ at 300 K (a_{br} is the asymmetry coefficient for μ^+ in bromoform). The cited values of $\bar{a}a_C^{-1}$ ($\bar{a}a_{br}^{-1}$) were obtained at the value $\exp(-t\lambda_r) = 1$. This means that in the case $\lambda_r \neq 0$ averaging of aa_C^{-1} (aa_{br}^{-1}) over the measurement time is carried out. The value of aa_C^{-1} is equal to the ratio of the residual polarizations PP_C^{-1} in the oxygen of the oxide and of the graphite. The values of a were corrected for muon stopping in the scintillation counter ahead of the target and in the walls of the cryostat, as indicated in Ref. 1.

It follows from the presented data that above the phase-transition temperature one observes a residual polarization at the precession frequency of the free muons of both signs, and that its values are close to the maximum. The residual polarization of the negative muons near the phase-transition point varies with temperature jumpwise. At liquid-nitrogen temperature one observes at the free-spin precession frequency of the muon a nonzero residual polarization of the positive and negative muons. Relaxation of the muon spins was observed at room temperature: For the case of μ^+ mesons $\lambda_r = 0.11 \pm 0.01$ μsec^{-1} and the relaxation rate for μ^- is $\lambda_p = 0.34 \pm 0.05$ μsec^{-1}. Accordingly $a/a_{br} = 0.96 \pm 0.02$ and $a/a_C = 1.10 \pm 0.05$; λ_r at other temperatures was not determined with sufficient accuracy.

After the Coulomb capture of the muon by the oxygen atom and the subsequent cascade transitions of the muon, a mesic atom $z - 1 = 7$ (mesic nitrogen) is produced with nonzero magnetic moment of the electron shell. The hyperfine interaction of this moment with the magnetic moment of the muon should lead to a partial depolarization of the muon spin within a time $\sim 10^{-10}$ sec, and to absence of residual polarization at the precession frequency of the quasi-free spin of the muons. The fact that a nonzero residual polariza-

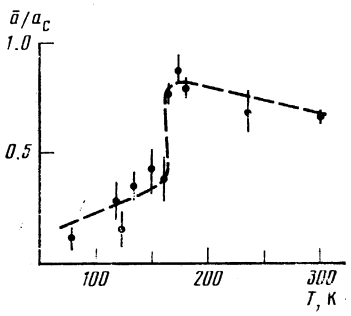

FIG. 1. Residual polarization $\bar{a}a_C^{-1}$ of the negative muons in the mesic atom of oxygen of V_2O_3 (relative to graphite) as a function of temperature.

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TABLE I. Residual polarization of negative muons in the mesic atom of oxygen for antiferromagnetic oxides at room temperature and at the temperature of liquid nitrogen, values[8] of the Néel temperature, and the muon-spin relaxation rates.

Oxides	T_N, K	T, K	$a(T)a_C^{-1}$	$\bar{a}(T)a_C^{-1}$	λ_p, μsec^{-1}
V_2O_3	167	300	1.10±0.08	0.66±0.03	0.34±0.06
	167	77	–	0.11±0.05	–
Cr_2O_3	308	300	–	0.12±0.06	–
Mn_2O_3	80	300	0.42±0.05	0.28±0.04	0.28±0.12
	80	77	–	0.04±0.02	–
MnO	122	300	1.10±0.19	0.42±0.06	0.66±0.18
	122	77	–	0.05±0.01	–
Fe_2O_3	953	300	–	0.02±0.05	–
FeO	198	300	–	0.00±0.04	–
CoO	291	300	0.73±0.14	0.31±0.04	0.53±0.14
	291	77	–	0.05±0.07	–
NiO	523	300	–	0.06±0.08	–
CuO	230	300	–	0.75±0.05	–
	230	77	–	0.15±0.07	–

tion was observed at this frequency is evidence of the presence of mechanisms that stop the hyperfine interaction. Such mechanisms can be the following: interaction of the mesic atom with the conduction electrons, a rapid chemical reaction of the mesic atom with formation of diamagnetic products, occupation by it of an impurity acceptor level,[1,5,9] and others.

In the metallic phase of V_2O_3 the frequency ν of the exchange with the spins is determined by the number of electrons with unpaired spins[10] (by the density of the spin states near the Fermi surface), and if ν is estimated by using the expression of Ref. 11 and the data of Refs. 7 and 12, then $\nu \sim 7 \times 10^{12}$ sec^{-1}. The hyperfine interaction frequency ω_0 in the mesic oxygen atom is close to 10^9 sec^{-1} (Refs. 9, 13). Thus, $\nu \gg \omega$ in the metallic phase. This means in fact that there is no depolarization on account of the hyperfine interaction of the muon with the electrons in the mesic atom. A large value of the residual polarization is indeed observed in experiment. The relaxation of the spins of the μ^+ and μ^- mesons, observed at room temperature, is apparently due to the dipole interactions of the magnetic moment of the mesons and the paramagnetic atoms of the metal. The difference obtained between the values of λ_r can be attributed to the fact that the μ^+ meson diffuses in the crystal, whereas the diffusion of the mesic atom can be neglected. Owing to the diffusion, the average value of the magnetic field at the μ^+ meson is less than at ath μ^- meson, and consequently λ_r for μ^+ mesons should be smaller than for μ^- mesons.

The fact that V_2O_3 is antiferromagnetic at $T < T_N$ explains the jump in the $\bar{a}(T)a_C^{-1}$ dependence. Indeed, as noted in Ref. 11, as $T \to T_N$ the frequency of the oscillations of the atomic spins is effectively decreased because of the ordering of the spins in the antiferromagnetic phase, and λ_r increases strongly, i.e., the residual polarization becomes close to zero. A nearly-zero residual polarization was measured in the following metals: the ferromagnets Fe (Ref. 4) and Ni (Ref. 14) and in the antiferromagnet Cr.[15] A zero residual polarization was observed also for the antiferromagnetic phase in rare-earth metals[11] in experiments with positive muons. What remains unanswered, however, is the question why the residual polarization in the antiferromagnetic phase in V_2O_3, at the frequency of the quasi-free spin of the muon, differs from zero. This apparently means that V_2O_3 contains regions with disordered magnetic structure, owing to the use of a polycrystalline target. This is indicated, in particular, by the presence of a noticeable paramagnetic susceptibility in the antiferromagnetic phase,[7] which is due to impurities and to violation of the structure.[8] The difference between the precession frequency of the free spin of the muon in the paramagnetic phase of V_2O_3 from the precession frequency of the spin of the free muon (the Knight shift) was not observed within the limits of the experimental accuracy ($\pm 0.2\%$).

We have also determined A (V/O), which is the ratio of the probabilities of the atomic capture of muons by the vanadium and by the oxygen in V_2O_3. Within the limits of the masurement accuracy, we did not find a temperature dependence of A (V/O). The mean value of A (V/O) was found to be 2.43 ± 0.05, which agrees with the quantity 2.19 ± 0.18 obtained earlier at 300 K (Ref. 16).

As noted above, at a disordered magnetic structure (for antiferromagnets below the Néel temperature) in weak external magnetic fields, one expects small values of the residual polarization. The table lists the values of the residual polarization in oxygen mesic atoms at temperatures 300 and 77 K for antiferromagnetic oxides, values obtained by us earlier[1,5,7] and in the present paper, as well as the values T_N of the Néel temperature.[8] It can be seen from the table that near-zero values of the residual polarization were obtained at a temperature below T_N for all the oxides in the table. For temperatures higher than T_N, the measured values of aa_C^{-1} differ noticeably from zero (0.4–1) and the mechanisms that compensate for the paramagnetism of the electron shell of the mesic atom of oxygen, referred to above, are in operation. An exception is FeO, where even above T_N the residual polarization is close to zero. For FeO, however, the data on the magnetic properties are not reliable in connection with the instability of the compounds with Fe^{+2} (Ref. 8) and can differ in the employed target T_N from those given in the table.

The present paper shows that negative muons can be used to identify the magnetic paramagnet–antiferromagnet (paramagnet–ferromagnet) phase transition. One can hope that more detailed investigations of the residual polarization and of the muon spin relaxation will uncover new possibilities for the study of phase transitons.

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