Depolarization of Negative Muons in Solids

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The method of spin precession in a magnetic field has been used to investigate the residual polarization of negative muons in solids with zero nuclear spin (metals, semiconductors, insulators) and in the oxygen of a number of oxygen compounds. The results obtained are not described by the theory of cascade depolarization for an isolated mesic atom without taking into account the dynamics of filling of the electron shell of the mesic atom in the course of the de-excitation process. To explain the measured values of residual polarization, it is necessary to take into account the interaction of the mesic atoms with the surrounding medium, the high degree of ionization of the mesic atom during the mesic atom cascade, and the paramagnetism of its electron shell.

IN decay of pions, negative muons are produced completely polarized longitudinally in the pion rest system. Muon beams from accelerators are partially depolarized, and their polarization is about 0.8. In the course of slowing down in matter, the depolarization of muons is unimportant.[1] After slowing down and stopping, strong depolarization of muons occurs in the formation of mesic atoms and in the cascade transitions of muons to the K shell of these mesic atoms. Muon depolarization in these processes has been discussed theoretically in a number of papers[2-5] for nuclei with zero spin. Presence of spin in the nucleus leads to an additional depolarization as the result of the hyperfine interaction.[5,6]

In atomic capture of negative muons, mesic atoms are formed in excited states. After completing a large number of cascade radiative and radiationless transitions, the muons fall into the K shell of the mesic atom. The depolarization of the mesons is due to the spin-orbit interaction and occurs at those levels[2] at which the distance between levels $l \pm \frac{1}{2}$ of the fine structure is much greater than the level width. In light mesic atoms muons are depolarized at the lower levels ($n_0 \leq 5$), where radiative transitions are dominant, and the levels $l_0 \pm \frac{1}{2}$ form an incoherent mixture. The polarization of muons in these levels n_0 from which depolarization in subsequent cascade transitions begins[5] depends on l and amounts to about $\frac{1}{3}$ of the initial value P_0.[3-5] The residual polarization at the end of the cascade[5] is given approximately by the expression

$$P(l_0) = 0.3(l_0 + 3)(2l_0 + 1)^{-1}P_0$$

and falls off with increasing l to a limiting value 0.156. For a statistical population of the sublevels l_0 of the level n_0, the residual polarization of muons in the ground state of the mesic atom is determined by the expression

$$P(n_0) = 0.15(5 + n_0)n_0^{-1}P_0.$$

The nature of the population of the level n_0 depends on the initial population of the highly excited levels of the mesic atom from which the cascade transitions are completed to the level n_0.

Investigation of the details of the earliest stage of the cascade ($n \sim 30-50$ in the region of the valence electron orbits) is very complex, and discussion of the mesic atom without regard for its neighboring atoms is not justified. The outer electron shells have an important effect on the capture of μ^- mesons in chemical compounds.[7] However, there are no theoretical evaluations of the population of the highest excited levels of the mesic atom and of the cascade transitions between them. Cascade transitions have been investigated theoretically beginning with n = 14, where the mesic-atom orbits lie in the region of the K-electron orbit of the atom.[8-10] However, the calculations made by de Borde[8] are rather approximate, the calculations of the level population by Mann and Rose[4] are inaccurate,[11] the more complete estimates made by Martin[9] refer K⁻ mesic atoms, and the calculations of Au-Yang and Cohen[10] were made numerically only for certain heavy mesic atoms.

To explain the observed x-ray yield from mesic atoms, Eisenberg and Kessler[12,13] used various initial populations over the sublevels l in the level n = 14 in calculations of cascade transitions. A distribution of the form $(2l + 1)e^{\alpha l}$, $\alpha \approx 0.1$ gave the best correspondence to the experimental data for light mesic atoms,[13,14] and the statistical distribution ($\sim(2l + 1)$) for medium atoms.[14] The experimental facts are also well described by an initial statistical distribution in l, but only in the case of shells strongly ionized at the time of the mesic cascade;[15] for example, calcium should have on the average only two electrons each in the K and L shells of the atom on passage of the cascade, and for magnesium, only one electron each.

As the cascade develops, the population deviates from the initial population and the population of sublevels with lower l increases.[12] Above the level n_0, de-excitation of the mesic atom occurs, for the most part, as the result of emission of Auger electrons, and below n_0 radiative transitions are dominant. In some materials emission of Auger electrons in the high levels can lead to partial or complete ionization of the atom.[8,15] If the filling of the atomic shells by electrons occurs more rapidly than the next Auger-electron emission, the rate of Auger transitions is not reduced. Slow filling can lead to formation of a mesic ion in an excited, and to a slowing of its rate of de-excitation as the result of reduction in the rate of radiationless transitions. This is equivalent to the depolarization beginning from a higher level $n > n_0$, and the residual polarization in the K shell of the mesic atom decreases. A decrease in the Auger transition rate by a factor of ten near n_0 increases n_0 by one to two units.

For a mesic atom in the ground state, depolarization

of the muon can be continued as the result of interaction with the magnetic moment of the electron shell[3] of the mesic atom. The characteristic relaxation time of the muon spin as the result of this interaction depends on what atomic orbit the unpaired electron is located in ($10^{-11}-2 \times 10^{-13}$ sec for carbon). This paramagnetic depolarization of the mesic atom depends on the interaction with the surrounding atoms, and also on the possibility of compensation of the magnetic moment of the electron shell and formation of a diamagnetic compound.[16,17] In the neutralization process it is also possible to form electron combinations in the mesic ion leading to temporary appearance of a magnetic moment in the electron shell, i.e., to an additional polarization of the muon during the cascade, if the cascade transition rate decreases (to $10^{11}-5 \times 10^{12}$ sec^{-1} in carbon) in certain stages as the result of reduction of the radiationless transition rate.

For a muon in the K shell of the mesic atom, the Coulomb field of the nucleus affects the spectrum of decay electrons and their angular distribution,[18] decreasing the asymmetry in this distribution. However, this effect exists only for mesic atoms with high Z. For $Z \approx 10$ the decrease in residual polarization is less than 10^{-2} from the initial value. The Coulomb field of the neighboring nuclei can increase the depolarization as the result of the mechanism of Coulomb breakup[19] of the mesic atom and a muonic cascade on repeated formation of mesic atoms. However, this mechanism requires high velocities of the initial mesic atom, i.e., formation of the mesic atom by a muon in flight, and the probability of this process cannot be appreciable.

In some materials a decrease in polarization is possible as the result of the Stark effect (mixing of the levels l)[20] and capture of a fraction of the muons in common mesic-molecule orbitals of chemical compounds [21,22] (the large mesic molecule model). Mixing of the levels plays an important role for mesic-atom processes in hydrogen and helium.[20] In heavier mesic atoms it can appear as the result of ionization of atoms in the process of mesic-atom formation or as the result of displacement of a mesic atom from a crystal lattice site into the electric field of the surrounding atoms. The transition rate depends on collisions with neighboring atoms and is important in the region of large n. Balancing of the distributions in l decreases the depolarization, since it increases with increasing l. For muons captured into common molecular levels, the depolarization should be small since, as the result of removal of the forbiddenness in Δl, the main role is played by radiative transitions immediately to the ground level of the mesic atom,[22] the number of cascade steps is sharply cut off, and for transitions with $\Delta l > 1$ the depolarization for large l is the same[3] as for a dipole transition. However, in materials which do not contain hydrogen, the region of common molecular levels is strongly narrowed[21] the fraction of muons captured into these levels is small, and this effect is neglected in the subsequent discussion.

Theoretically, cascade depolarization of muons has been discussed rather well[2-5] for a certain ideal case in which there is no magnetic moment in the electron shell and the filling of vacancies produced as the result of Auger electron emission occurs rapidly. However,

there is no quantitative discussion of the action of the surrounding medium on this depolarization: capture into high-lying mesic-atom or mesic-molecule levels, the effect of paramagnetism of the electron shell of the mesic atom in de-excitation and in the ground state, ionization of the mesic atom and the surrounding atoms, the effect of Stark transitions and perturbations of the crystal lattice, and chemical reactions with the mesic atom. Detailed theoretical and experimental studies of these effects will permit use of mesic atoms (for study of various quantum processes occurring in real media.

Depolarization of negative muons in various materials with zero nuclear spin has been studied experimentally by a number of workers.[23-35] Most studies have utilized the method of muon spin precession in a magnetic field, and the residual polarization was measured at the free muon spin precession frequency ω. The residual polarization P is determined from the relation[1] $P = 3a_0P_0^{-1}$, where P_0 is the polarization of the beam, a_0 is the asymmetry coefficient in the angular distribution of decay electrons for zero electron-detection energy threshold. Experimental values of the asymmetry coefficient taken from [24-35] are given in Table I. They are given relative to the similar quantity in graphite, in order to avoid uncertainty connected with the different polarization of beams in different accelerators, which is not always accurately known, and also the inaccuracies resulting from reduction of the asymmetry coefficient to zero threshold.

The experimental results obtained in [24-35] can be summarized briefly as follows. In liquified gases[25-27] —helium, oxygen, argon—the residual polarization is close to zero, and the same in paramagnetic metals[28] —chromium, molybdenum, palladium, and tungsten. In hydrocarbon compounds[26,34] and in water[26,34] it amounts to 0.5 of the residual polarization in graphite, and in metals, semiconductors, and dielectrics[26,28-33,35] it is greater than 0.5 but less than unity, although the spread in the measured values is quite substantial. Only

Table I

Decay electron asymmetry coefficient for negative muons in materials with zero nuclear spin, expressed in fractions of the asymmetry for graphite

Material	a/a_C	Material	a/a_C
Helium (liquid)	-0.02 ± 0.08[25] 0.04 ± 0.11[26] 0.44 ± 0.34[27]	Magnesium	0.82 ± 0.07[26] 1.45 ± 0.27[28] 0.82 ± 0.16[29] 0.77 ± 0.18[31]
Styrene	0.54 ± 0.03[34]	Silicon	0.67 ± 0.17[31] 0.83 ± 0.03[32]
Polyethylene	0.45 ± 0.09[22] 1.05 ± 0.25[28] 0.52 ± 0.04[34]	Sulfur	1.05 ± 0.20[28] 0.96 ± 0.34[29] 0.58 ± 0.13[31] 0.55 ± 0.03[32] 0.99 ± 0.15[35]
Paraffin	0.56 ± 0.09[26] 0.95 ± 0.24[28] 0.50 ± 0.05[34]		
Cyclohexane	0.51 ± 0.04[34]	Argon (liquid)	0.17 ± 0.12[25]
Toluene	0.45 ± 0.05[26] 0.30 ± 0.06[34]	Calcium	0.76 ± 0.04[30] 0.67 ± 0.03[32] 1.00 ± 0.16[33]
Benzene	0.41 ± 0.05[34]		
Phenylcyclohexane	0.43 ± 0.04[34]	Chromium	0.00 ± 0.22[28]
Polystyrene	0.32 ± 0.04[34]	Zinc	1.45 ± 0.32[28]
Polyvinyl	0.33 ± 0.09[26]	Molybdenum	0.12 ± 0.25[28]
Oxygen (liquid)	0.09 ± 0.11[26[	Palladium	0.00 ± 0.25[28]
Water	0.38 ± 0.11[26] 0.43 ± 0.03[34] 1.07 ± 0.18[28]	Cadmium	1.37 ± 0.34[28]
		Tungsten	0.12 ± 0.25[28]
		Lead	1.35 ± 0.35[28]

the data on metals and semiconductors are close to the limiting values expected from calculations according to the cascade model. The qualitatively zero residual polarization in gases is explained by the paramagnetism of mesic atoms,[17] and the residual polarization values in water and hydrocarbon compounds hardly can be understood without taking into account the effect of neighboring atoms in the process of de-excitation of mesic atoms or of their interactions with the surrounding medium.

In the present work we have presented the results of measurement of the residual polarization of muons in solid materials—metals, dielectrics, and insulators—for the purpose of comparison with the theory of cascade depolarization and to clarify the effect of the electron shells. We have also measured the depolarization in oxygen compounds in order to evaluate the effect of the bond with neighboring atoms on the residual polarization in the mesic atom of oxygen.

The apparatus for determination of muon residual polarization has been described previously.[23] The measurements were made in a separated beam of negative muons with momentum 158 MeV/c from the meson channel of the JINR synchrocyclotron with an impurity of pion stoppings of less than 0.002. The beam used was extended in time, with no time structure, and with a polarization 0.75 ± 0.04. Pulses from muon stoppings (from three counters in coincidence) and from decay electrons (from three counters in coincidence) controlled the operation of a converter which transformed the time interval between these two pulses to a signal of variable amplitude for recording by a 100-channel analyzer with a channel width 18–74 nsec. A guard system blocked the analyzer input in the case of appearance of second pulses from muon stopping or electrons for a period of 12 μsec before the time zero and 8 μsec after it. The time interval for detection of decay electrons was chosen from 2 to 6 μsec. Targets of thickness 4–5 g/cm^2 were placed in a magnetic field of 50–400 Oe. As oxygen-containing materials we selected compounds of oxygen with atoms either having nonzero nuclear spin or a short muon lifetime in these atoms relative to oxygen, in order to avoid a contribution of these atoms to the observed asymmetry of decay electrons from oxygen.

Analysis of the time spectra of appearance of decay electrons was carried out in a computer by the method of least squares. For the one-component targets we used the expression

$$N(t) = N_0[1 + a\cos(\omega t + \delta)]e^{-t/\tau} + N_c[1 + 0.017\cos(\omega t + \delta)]e^{-t/\tau_c} + C,$$

where N(t) is the number of counts in an analyzer channel at the time t, τ is the muon lifetime in the target, a is the electron decay asymmetry coefficient, δ is the initial phase produced by an electronic delay circuit and the geometry of the apparatus, C is the time-independent background, ω is the muon spin precession frequency in the magnetic field N_C and τ_C are the corresponding values for the carbon background from the scintillation counters, whose value N_C/N_0 did not exceed 0.05. The value a = 0.017 was determined previously.[34] The quantities N_0, τ, a, δ, N_C, and C were varied in the analysis.

For the oxygen compounds the asymmetry coefficient was measured in oxygen. In the analysis, instead of N_C, τ_C, and the coefficient 0.017, the quantities N, τ, and a were varied for oxygen, and the contribution of the asymmetry coefficient from the scintillation counters was taken into account, although it was several times smaller than the corresponding errors in the asymmetry in oxygen, or at least comparable with them as in the case of CaO and CuO.

Table II gives the results of measurement of the asymmetry coefficient for 15 solid materials. For the one-component targets the background amounted several percent (C/N_0 = 0.03–0.08), and for the oxygen compounds in the worst case (CuO) it was less than 0.2 of the number of electrons from oxygen C/N_0 = 0.08–0.2). The number of electrons recorded for the various targets varied from 5×10^4 (for PbS) to 3×10^6 (for Si). Corrections to the asymmetry for the different target thicknesses did not exceed a few percent. Table II also lists the asymmetry coefficient values relative to the coefficient in graphite.

It can be seen from Table II that the greatest residual polarization value was measured in graphite (P = 0.194 ± 0.011). The depolarization in all other materials measured is greater than in graphite. The high residual polarization in metals can be explained by the high rate of the exchange interaction of the valence electron of the mesic atom with the conduction electrons. The valence electron does not exist in a state with a definite spin direction, and the mesic atom does not have a magnetic moment. In the opposite case zero residual polarization should be recorded at the precession frequency of the free muon spin.

Measurements in titanium gave a large residual polarization 0.155 ± 0.018. Titanium belongs to the group of transition metals like chromium, molybdenum, and tungsten, for which Ignatenko et al.[28] have measured zero residual polarization, which was explained by paramagnetism of the mesic atoms formed. The large asymmetry of decay electrons in titanium indicates that there is no paramagnetism in a mesic atom in the titanium lattice.

In order to shed light on the role of exchange of electrons of the mesic atom with conduction electrons, we used pure silicon (n type, 200 ohm-cm) and silicon with an impurity of 3×10^{18} atoms/cm^3 of antimony (n type, 0.01 ohm-cm) at room temperature and liquid nitrogen temperature. In spite of the fact that the number of charge carriers was varied over a wide range, the residual polarization remained high ($a/a_C = 0.91 \pm 0.03$) and constant within the experimental error. The fact that depolarization of positive muons in the same sam-

Table II. Decay electron asymmetry coefficient for negative muons in solid materials

Material	−a, %	a/a_C	Material	−a, %	a/a_C
C	4.80±0.10		Cr₂O₃	0.60±0.31	0.12±0.06
C (−196°)	4.36±0.15	0.91±0.05	B₂O₃	0.44±0.12	0.09±0.02
Mg	3.31±0.17	0.70±0.05	CaO	0.24±0.14	0.05±0.03
Si (with impurities)	4.70±0.23	0.98±0.06	SiO₂ (fused)	0.57±0.27	0.12±0.06
Si(with impurities,−196°)	4.30±0.27	0.90±0.06	SiO₂ (crystal)	0.42±0.29	0.09±0.06
Si (pure)	4.10±0.41	0.85±0.08	CuO	3.24±0.50	0.65±0.10
Si (pure, −196°)	4.30±0.46	0.90±0.10	CuO (−196°)	0.70±0.35	0.15±0.07
S	3.32±0.80		KOH	0.83±0.25	0.17±0.05
Ca	4.25±0.68	0.89±0.11	NaOH	1.22±0.37	0.25±0.08
Ti	3.90±0.38	0.80±0.08	PbS	6.1±1.9	1.27±0.40
Al₂O₃ (single crystal)	0.26±0.28	0.05±0.06	PbO	1.33±0.28	0.28±0.05

ples depends sharply both on the amount of impurity and on the temperature shows that the exchange interaction mechanism is not dominant in the case of mesic atoms. A temperature dependence of the residual polarization also was not found in graphite.

In the other semiconductors investigated, CuO and PbS, the residual polarization for oxygen and sulfur is also high. In all these cases the mesic atom formed has a paramagnetic electron shell, and to observe the observed value of residual polarization it is necessary to introduce mechanisms leading to compensation of this paramagnetism. For example, in silicon a mesic atom is formed which has the electron shell of aluminum with one unpaired electron. This mesic atom occupies an acceptor level in the silicon lattice, which leads to rapid compensation of the paramagnetism of its electron shell. The large measured polarization value can be explained by assuming that the mesic atom of aluminum occupies the level for a time substantially less than $\sim 3 \times 10^{-12}$ sec, since a decrease in the residual polarization by a factor of two occurs as the result of paramagnetic depolarization[3] in a time $\sim 3 \times 10^{-12}$ sec. It is possible that a similar situation occurs for CuO and PbS. However, here it is necessary, in principle, to take into account also the possibility of occurrence of fast chemical reactions of mesic atoms in the medium with formation of diamagnetic compounds. Thus, the temperature dependence of the residual polarization in CuO can be considered to be the result of temperature dependence of the rate of a chemical reaction with formation of the diamagnetic compound $Cu_\mu NO$, although occurrence of this reaction requires very specific conditions.

The measured average value of asymmetry coefficient for silicon differs less from the value in graphite than for other materials. The smaller polarization in silicon may be due to reduction[12] of the population of levels with small l in comparison with graphite in the region $n_0 \sim 5$, where depolarization in the cascade is already substantial. From Tables I and II we can calculate the weighted mean values of residual polarization relative to the polarization in graphite for a number of elements. This value is 0.76 ± 0.04 for magnesium, 0.87 ± 0.03 for silicon, 0.63 ± 0.03 for sulfur, 0.72 ± 0.03 for calcium, and 0.80 ± 0.08 for titanium. The results obtained indicate some connection of the atomic electron shell with muonic depolarization in the mesic atom,[7] associated with the nature of the cascade transitions and with the population of the lower levels. This is illustrated in Fig. 1, where we have shown the value of a/a_C as a function of Z of the mesic atom. The values for mesic atoms with nonzero nuclear spin—lithium, beryllium, and nitrogen[36]—have been corrected for the depolarization produced by hyperfine interaction of the muon and the nucleus, calculated theoretically.[5] In Fig. 1 we can see both a general reduction in residual polarization with increasing Z, and inside the series of the periodic table from the beginning of the series to its end. The behavior for the oxygen compounds is similar.

The weighted mean absolute value of residual polarization in graphite, including the data of [28, 31, 35], is 0.208 ± 0.011. According to the cascade model[5] this quantity achieves values of 0.30 for a statistical population over the l sublevels of the level $n_0 = 5$ and 0.35 for a popula-
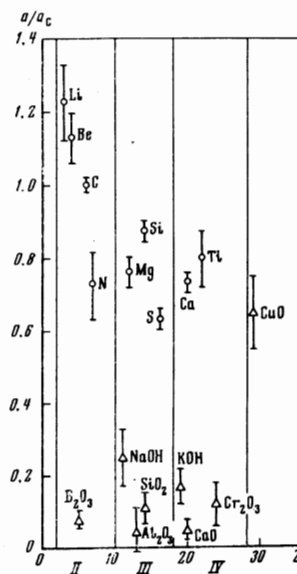

FIG. 1. Residual polarization (relative to carbon) as a function of Z of the mesic atoms and of Z of the atoms in oxygen compounds. The vertical lines correspond to the limits of the series of the periodic table.

tion following from the higher-lying cascade transitions.[12] In Fig. 2 we have shown the residual polarization as a function of n_0 for these distributions in l. From the fact that the measured value is less than 0.3, it follows that there is either incomplete compensation of the magnetic moment of the mesic atom electron shell by the interaction with the surrounding atoms, or that muon depolarization occurs also in levels $n_0 > 5$. When the experimental errors are taken into account, agreement with theory begins only for $n_0 > 9$. In the same figure we have shown the data for metals (the value of n_0 corresponds to the mesic-atom level in which the Auger transition rate is 100 times greater than the radiative transition rate[8] for circular orbits). All the values lie appreciably below the curves obtained from the model of cascade depolarization without inclusion of the effect of the electron shell and of the slowing down of the rate of radiationless transitions. This comparison shows that to explain the observed depolarization values it is necessary to take into account the effects of the surrounding medium, which we have discussed above.

It is evident from Table II that in most oxides the residual polarization does not exceed 0.1 of the value in carbon (except CuO), in spite of the completely dif-
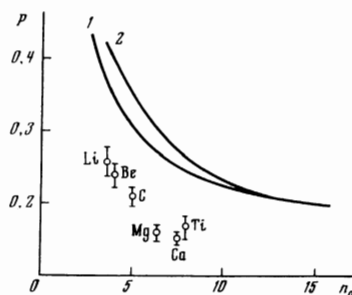

FIG. 2. Residual polarization as a function of the quantum number n_0 of the mesic-atom level below which depolarization is important. [5] For curve 1 a statistical population over the l sublevels of the n_0 level was used, and for curve 2 the population following from the higher-lying cascade transitions in carbon. [12] Residual polarization values are shown for metals (the weighted means of the experimental results of the present work and refs. 26, 28–33, 36, and 35) and graphite.

ferent structure of the materials investigated: single crystals (Al_2O_3, SiO_2), powders (B_2O_3, Cr_2O_3, CaO, KOH, NaOH), and fused materials (SiO_2). Apparently the structural bonds of oxygen with the other atoms are not the main factor affecting the substantial depolarization of muons in oxygen. In these compounds, a mesic nitrogen atom is formed and the measured polarization value is determined by the probability of compensation of the paramagnetism of its electron shell.

In summarizing, we can draw certain conclusions:

1. The measured absolute values of residual polarization of negative muons in graphite and metals are significantly lower than predicted by the theory of cascade depolarization for an isolated mesic atom (without inclusion of the dynamics of filling of the mesic-atom electron shell). These results can be interpreted by assuming the existence of a high degree of ionization of the mesic atom during the mesic-atom cascade process.

2. The large measured value of residual polarization in silicon and the independence of this value on the concentration of impurities and the temperature can be explained by taking into account the fact that the aluminum mesic atom formed rapidly occupies a level in the silicon lattice with compensation of the paramagnetism of its electron shell. To explain the large measured value of residual polarization in other semiconductors studied, it is necessary also to take into account the possibility of occurrence of fast chemical reactions with formation of diamagnetic compounds.

3. From the measurements in oxygen compounds, a residual polarization was obtained which is on the average an order of magnitude smaller than in graphite. To explain this value it is necessary to consider chemical interactions of the nitrogen mesic atom with formation of diamagnetic compounds.

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