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ON THE NATURE OF THE FAST DEPOLARIZATION OF MUONS IN CONDESED NITROGEN

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Temperature dependences of the depolarization rate, the muon precession initial amplitude and phase in liquid and crystalline nitrogen with the oxygen content of 10^{-6} have been measured. It has been shown that muon spin relaxation parameters in nitrogen do not change at the reduction of the oxygen impurity content from $0.7 \cdot 10^{-4}$ to 10^{-6}. The fast depolarization of muons in condensed nitrogen is apparently due to the formation of muonium atoms. The muon precession initial phase has been measured as a function of the perpendicular magnetic field to determine the state of short-lived muonium in nitrogen. It has been determined that muonium in nitrogen is in an excited state.

Temperature dependences of the muon depolarization rate Λ, the initial amplitude A and the muon precession initial phase φ in condensed nitrogen have been studied in ref. /1/. The fast depolarization of muons for times unobservable in the experiment ($t \lesssim 10^{-8}$s) has been found both in liquid and crystalline nitrogen.

The fast depolarization of muons in nitrogen may be due to the formation of a hydrogen-like atom of muonium /2/. It is known that in gaseous nitrogen 84% of muons form long-lived atoms of muonium /3/. Long-lived muonium has not been found in condensed nitrogen /1/. The signal of a short-lived ($\tau < 10^{-8}$s) muonium atom cannot be observed in the Fourier spectrum of the precession and its existence can be revealed only by the initial amplitude decrease and the muon precession phase shift /4/. The formation of singlet muonium results in the muon depolarization at times $t_0 = 1/\omega_0$, where ω_0 is the muonium hyperfine splitting frequency. The muon depolarization does not take place in triplet muonium. However, the muonium electron interaction with the medium at which a spin flip occurs, causes additional depolarization of muons /2/.

The fast depolarization of muons in nitrogen may be also due to their interaction with paramagnetic impurities, particularly with oxygen ones. As the electron magnetic moment of oxygen exceeds the nuclear magnetic moment of nitrogen ^{14}N by about 4 orders of magnitude, the presence of O_2 impurities may critically affect the muon depolarization process. The oxygen content in the starting gas was measured in ref. /1/ before condensation and amounted to $0.7 \cdot 10^{-4}$.

As far as the authors know, the mobility of positive charges in diatomic cryocrystals has been measured only in solid hydrogen /5/ and constitutes of the order of 10^{-5}-10^{-6} cm^{2}V^{-1}s^{-1}. At this mobility and the oxygen concentration noted above the muon coming to the O_2 molecule takes place at times of the order of 10^{-5}-10^{-6}s, that appreciably exceeds the "dead" time of the electronics. Regretfully, the mobility of positive charges in crystalline nitrogen was not determined /6/. The instrumentation sensitivity limit was of the order of 10^{-4}cm^{2}V^{-1}s^{-1}. If this value is taken as the upper limit of the mobility of the muon-formed complex in nitrogen at the oxygen concentration of $0.7 \cdot 10^{-4}$ and temperatures of the order of 60K, one can obtain the value of time of a μ^+

coming to a O_2 molecule of the order of $5 \cdot 10^{-8}$s. At this time value the fast depolarization may be due to impurities of paramagnetic oxygen.

The present paper reports the study of the cause of the fast depolarization of muons in condensed nitrogen.

The muon precession parameters in liquid and crystalline nitrogen have been experimentally studied using the standard μSR-method /2/. The technique of sample preparation, temperature variation and measurement is described in ref. /7/. The experimental spectra were treated in accordance with the expression:

$$N(t) = N_0 \, exp(-t/\tau_\mu) \left[1 + Ae^{-\Lambda t} \cos(\omega t + \varphi)\right] + F, \qquad (1)$$

where A is the muon precession amplitude at the time t = 0; Λ is the depolarization rate, ω is the muon precession frequency, φ is the initial phase of the precession, τ_μ – the muon lifetime, F is the background, N_0 – the normalization constant. The resolving time of the electronics was $5.5 \cdot 10^{-9}$s. The number of events in the spectrum was not less than 10^6.

The oxygen content in a specially purified starting gas was measured by means of the "Fluorit" gasanalyzer, having the sensitivity not worse than 0.1 ppm. The measurements showed that the content of oxygen impurities amounted to $\simeq$ 1 ppm. The independent analysis of gaseous nitrogen just before the experiment was carried out using the "quadravac" analyzing system, which sensitivity makes it possible to measure the amount of impurities of the order of 10^{-4}. No oxygen impurities were found. Regretfully, the experiment conditions did not allow to carry out the analysis of nitrogen being condensed as a sample upon completion of the μSR-measurements. The estimate of the time of a positive muon coming to an O_2 molecule at the oxygen concentration of $\sim 10^{-6}$ gives the value of the order of 10^{-6} s even for the upper limit of mobility.

This value makes it possible to rule out the interaction with oxygen impurities as a possible cause of the fast depolarization of muons.

The results of measurements are shown in fig. 1. The temperature dependence of the muon depolarization rate is shown in the upper part of the figure. The value of Λ is temperature-independent and amounts to $\Lambda = (0.14 \pm 0.02)\ \mu s^{-1}$, which within the error is in good agreement with the depolarization rate value, obtained in ref. /1/. The consideration of dipole-dipole interaction of the magnetic moment of a free muon located in a lattice interstice or in a nitrogen lattice vacancy, with the surrounding nitrogen molecules does not ensure the depolarization rate value observed in the experiment. The obtained value of Λ corresponds to the muon depolarization in a stationary ion $N_2\mu^+$ /7/ having the geometry parameters of the analogous ion N_2p^+ /8/. The binding energy of the N_2p^+ ion is known to reach 5.1 eV /8/. The muon in the $N_2\mu^+$ ion has the same binding energy within the accuracy of the isotope effect. Thus, this state is quite favourable energetically and seems to be the most probable.

The temperature dependence of the precession initial amplitude A/A_0 is shown in the central part of fig. 1, where $A_0 = 0.160 \pm 0.002$ is the muon precession initial amplitude in a copper sample, in which no muonium is known to be formed and the precession amplitude corresponds to the total muon amplitude. Within the whole temperature range the A/A_0 value is appreciably less than 1. This means that fast depolarization of muons occurs in nitrogen with the oxygen concentration $\sim 10^{-6}$. The type of the temperature dependence of A/A_0 is the same as that in ref. /1/. The values of the initial amplitude obtained in both experiments are in good agreement within the error of measurements both in liquid nitrogen and in the α- and β-phases of crystalline nitrogen. In its bottom part fig. 1 presents the temperature dependence of the muon precession initial phase $\varphi_{Mu} = \varphi - \varphi_0$, where $\varphi_0 = (0.060 \pm 0.015)$rad is the phase assigned to the presence of a definite angle between the muon polarization vector and the axis of the positron telescope. The value of φ_{Mu} within the experimental error agrees with the results obtained in ref. /1/.

Thus, it has been experimentally shown that the fast depolarization of muons in nitrogen is not due to oxygen impurities. The muon precession parameters in condensed nitrogen do not depend on the oxygen content at the O_2 concentration less than 10^{-4}.

The fast depolarization of muons in condensed nitrogen is apparently due to the formation of muonium atoms. The absence of long-lived muonium can be explained in view of the chemical reaction proposed in /1/

$$Mu + N_2 \rightarrow N_2\mu^+ \qquad (2)$$

Within the time τ an irreversible transition from the paramagnetic state Mu into a diamagnetic complex $N_2\mu^+$ takes place. If the reaction runs fast enough, the initial state of muonium may be not observed.

Analogous processes of the $Mu \rightarrow \mu^+$ reaction were observed in a number of substances /4,9,10/. The analytical expressions for the muon precession initial amplitude (residual polarization) and phase, in the presence of the muonium stage depolarization of μ^+, were obtained by Ivanter and Smilga /11,12/ on the case of an isotropic muonium atom (normal muonium). The results obtained in /1/ and in the present paper on the assumption of the isotropic hyperfine interaction in muonium make it possible to draw certain conclusions on the theory parameters - the hyperfine interaction ω_0, the time τ before μ^+ enters into a chemical reaction and the frequency ν of the muonium electron spin flip on interaction with the medium. The estimate of the time $\tau \sim 10^{-10}$s obtained in /1/ from the measurements of the muon precession initial phase value φ_{Mu} is confirmed by the present experiment. Schenck in his work /2/ calculated on the assumption of $\nu = 0$, the dependence of the initial phase φ_{Mu} on the time τ of μ^+ entering into a chemical reaction in the perpendicular field H=100 Oe. According to calculations the phase φ_{Mu} is positive only at the time of the chemical reaction $10^{-11} < \tau < 10^{-10}$s, which confirms the above-noted estimation of τ. The fact that the muon precession initial phase turns out to be positive, apparently shows that the processes that take place in singlet muonium contribute substantially to the "accumulated" phase. This can take place if the complete depolarization of a muon in singlet muonium does not occur within the muonium lifetime τ. The time of muon depolarization in singlet muonium $t_0 = 1/\omega_0$, i.e. the condition $1/\omega_0 > \tau$ is fulfilled. From this condition one can obtain the upper limit of the muonium atom hyperfine splitting frequency in nitrogen $\omega_0 < 10^{10}$ rad$\cdot$s^{-1}, which confirms the assumption made in ref. /1/ of the excited state of muonium ($\omega_0 < \omega_0^* = 2.8 \cdot 10^{10}$ rad$\cdot$s^{-1} - the muonium hyperfine splitting frequency in the ground state).

If muonium in condensed nitrogen is in the ground 1S state, reaction (2) is highly improbable, as the binding energy of muonium in the ground state (13.54 eV) is appreciably higher than that of the muon in the $N_2\mu^+$ complex ($\sim$ 5 eV) /1/. If muonium is in an excited state, its binding energy turns out to be considerably less than 13 eV and reaction (2) may take place. The lifetime of the excited states of atoms is known to reach 10^{-8}-10^{-9}s/13/. The lifetime of a hydrogen atom in the 2P state amounts to $1.6 \cdot 10^{-9}$s. The 2S state is generally metastable with the lifetime of 0.14 s. The estimate of the time $\tau \sim 10^{-10}$s, obtained in /1/ turns out to be much less than the lifetime of the excited state of muonium. Thus, reaction (2) apparently ends before the muonium atom goes into the ground state. The excited states of muonium were observed in experiments on determination of the Lamb shift (see, e.g. /14/). When the muon hit the foil, the muonium atom in the excited state $2S_{1/2}$ escaped from it. The transition $2S_{1/2} \rightarrow 1S_{1/2}$ is forbidden for muonium as well as for the hydrogen atom. The process $2S_{1/2} \rightarrow 2P_{1/2}$ made it possible to observe the transition $2P_{1/2} \rightarrow 1S_{1/2}$, which was detected in the experiment.

The state of short-lived muonium in a matter, entering into a reaction of type (2), can be determined by measuring the dependence of the muon precession initial phase φ (see expression (1)) on the value of the perpendicular magnetic field. In a large magnetic field (Paschen-Back limit) the Zeeman energy of the electron and muon magnetic moments turns out to exceed the hyperfine interaction in muonium, which results in decoupling of the electron and muon magnetic moments, the "accumulating" muonium phase being equal to zero. The classical analog corresponds to a very fast rotation of the electron magnetic moment in an external magnetic field. As a result of fast rotation the electron magnetic field "at the muon site" becomes equal to zero. That results in the absence of the muonium phase. In low magnetic fields the hyperfine interaction exceeds the Zeeman energy. The coupling between the electron and muon magnetic moments is preserved and $M = I+S$, where I and S are the muon and electron magnetic moments, is a good quantum number. The muonium phase turns out to be an increasing function of the magnetic field $\varphi_{Mu} \sim \omega_{Mu} \tau$, where ω_{Mu} is the Larmor frequency of muonium. The transition from the region in which the hyperfine interaction predominated into the Paschen-Back region takes place under the condition $2\mu_e H_0 = \hbar\omega_0$, where μ_e is the electron magnetic moment, ω_0 is the frequency of the hyperfine interaction (we neglected the term $\mu_\mu H_0 \ll \mu_e H_0$, where μ_μ is the muon magnetic moment). For muonium in the ground 1S state in vacuum $H_0^{vac} = 1585$ Oe is the muon magnetic field "at the muonium electron site". The dependence of the muon precession initial phase φ_{Mu} on the perpendicular magnetic field in case of an irreversible transition of the ground 1S state of isotropic muonium into the muon diamagnetic state in the absence of the electron spin flip processes ($\nu = 0$) is shown in ref. /4/. In this case the dependence $\varphi_{Mu}(H)$ has its maximum at $H = 1/2H_0^{vac}$ /12/.

The dependence of the muon precession initial phase on the perpendicular magnetic field was measured to determine the state of muonium in condensed nitrogen. The measurements were carried out at temperatures T = 28K (α-phase of N_2), T = 37K (the $\alpha-\beta$-transition region, the maximum of the dependence $A/A_0(T)$), T = 55K (β-phase of N_2) and T = 73.5K (liquid nitrogen). The magnetic field varied within 0÷1000 Oe. The field relative stability and inhomogeneity over the sample were no worse than 10^{-3}. The procedure of the determination of the muonium phase φ_{Mu} was carried out similar to the one described in /7/. The measurement of the "instrumental phase" φ_0 related to the presence of a definite angle between the muon polarization vector and the axis of the positron telescope was carried out in a copper sample, in which no muonium is known to be formed. The change of the field direction to the opposite one resulted in the change of the φ_0 sign, which is due to a different direction of the muon spin rotation in a perpendicular magnetic field. The dependences $\varphi_{Mu}(H)$ at the opposite field directions did not differ in principle. This was determined by measurements at T = 28K. All the results are given to one direction of the field. Fig. 2 presents the dependences of the muon precession initial phase on the perpendicular magnetic field at T = 28K and T = 37K. The analogous dependence at T = 55K is shown in fig. 3.

The determination of H_0 for muonium should be carried out in case of $\nu = 0$ /12/. As seen from fig. 1, the polarization of muons turns out to be equal $\sim 1/2$ only in the vicinity of the $\alpha-\beta$-transition. Apparently, the change of the muonium electron polarization is absent only in this region. The dependence $\varphi_{Mu}(H)$ at T = 37K has a weak maximum at $1/2H_0 \sim 100-150$ Oe. Regretfully, the low accuracy of the phase determination did not allow to carry out more detailed measurements. The obtained value of H_0 should be compared with the vacuum value for muonium in the 1S state ($H_0^{vac} = 1585$ Oe). As $H_0 \sim 1/a_0^3$, where a_0 is the Bohr radius, the muonium atom in nitrogen is apparently "swollen" and has the Bohr radius of ~ 1Å. Thus, it has been experimentally determined, that muonium in nitrogen is in an excited state.

The decrease in the muon precession amplitude A/A_0 less than 1/2 (at least at $T < 35K$ and $T > 40K$) shows that the muon polarization is not preserved in triplet muonium. Such a situation may take place in anomalous muonium (the muonium with an anisotropic hyperfine interaction) /2/, or in an isotropic normal muonium with account of the electron spin flip processes with the frequency ν . The weak intermolecular interaction in a nitrogen crystal as well as a high degree of the lattice symmetry will not probably result in the loss of the spherical symmetry of muonium. As for the flip processes, nowadays it is difficult to draw any conclusions on their nature. The assumption noted in /7/ of the possible interaction of the muonium electron with the electron spin of oxygen impurity molecules in apparently ruled out by the present work. Close values of A/A_0 are observed in the α -phase of crystalline nitrogen and in liquid nitrogen, in which the particle diffusion coefficients differ by several orders of magnitude. That is why the interaction of muonium with the products of the ionization and excitation processes in the track formed by the muon at thermalization seems highly improbable.

The superhyperfine or nuclear hyperfine interaction /2,4/ of the muonium electron with the nuclear magnetic moments of the nearest neighbouring nitrogen molecules is apparently the cause of the muon precession initial amplitude decrease. Fast rotation of N_2 molecules reduces the interaction similar to the well-known phenomenon of the "dynamic narrowing" /2/.

Freezing of the molecule rotation due to the anisotropic quadrupole-quadrupole interaction results in the increase of the nuclear hyperfine interaction. Apparently, it is the orientational dynamics of N_2 molecules that determines the temperature dependence of the muon precession initial amplitude.

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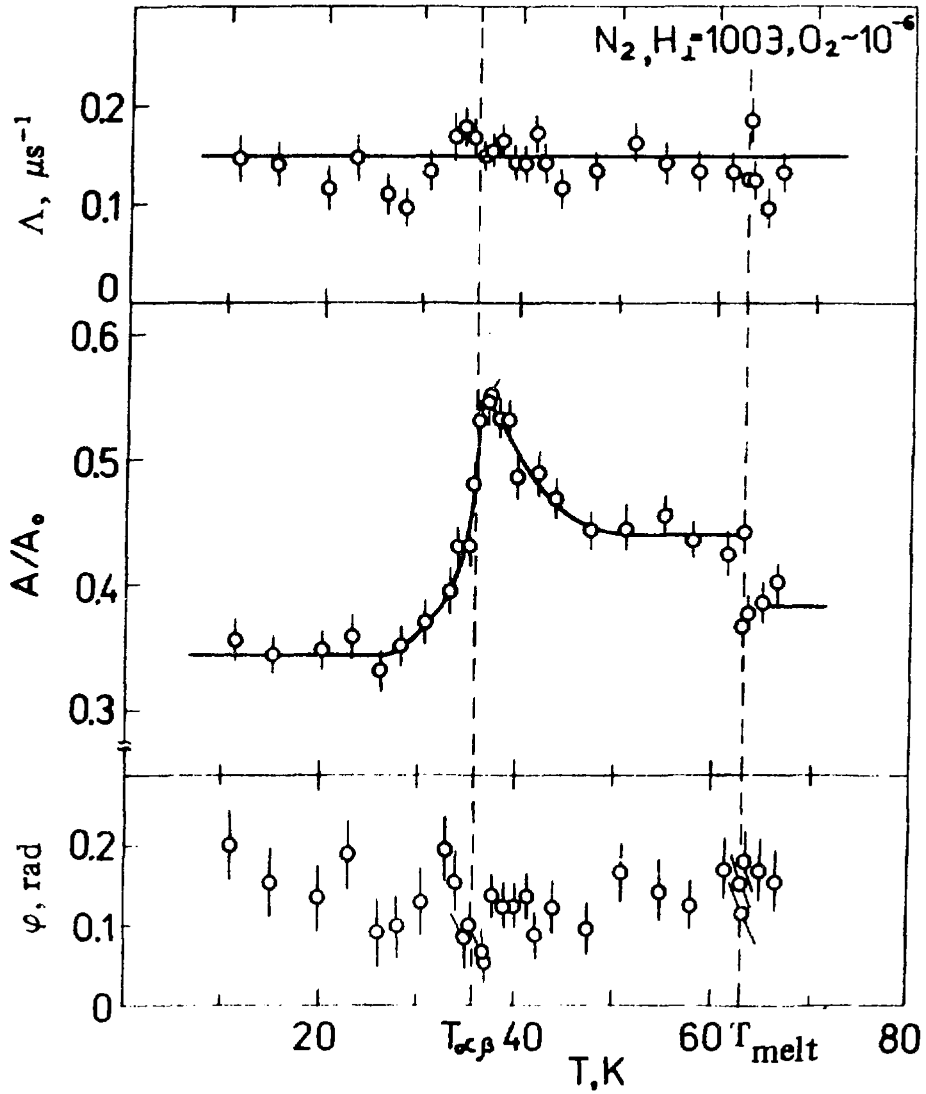

Fig. 1. Temperature dependences of the muon depolarization rate Λ, the muon precession initial amplitude A/A_0 and the precession initial phase φ in condensed nitrogen. The magnetic field $H_\perp$ = 100 Oe.

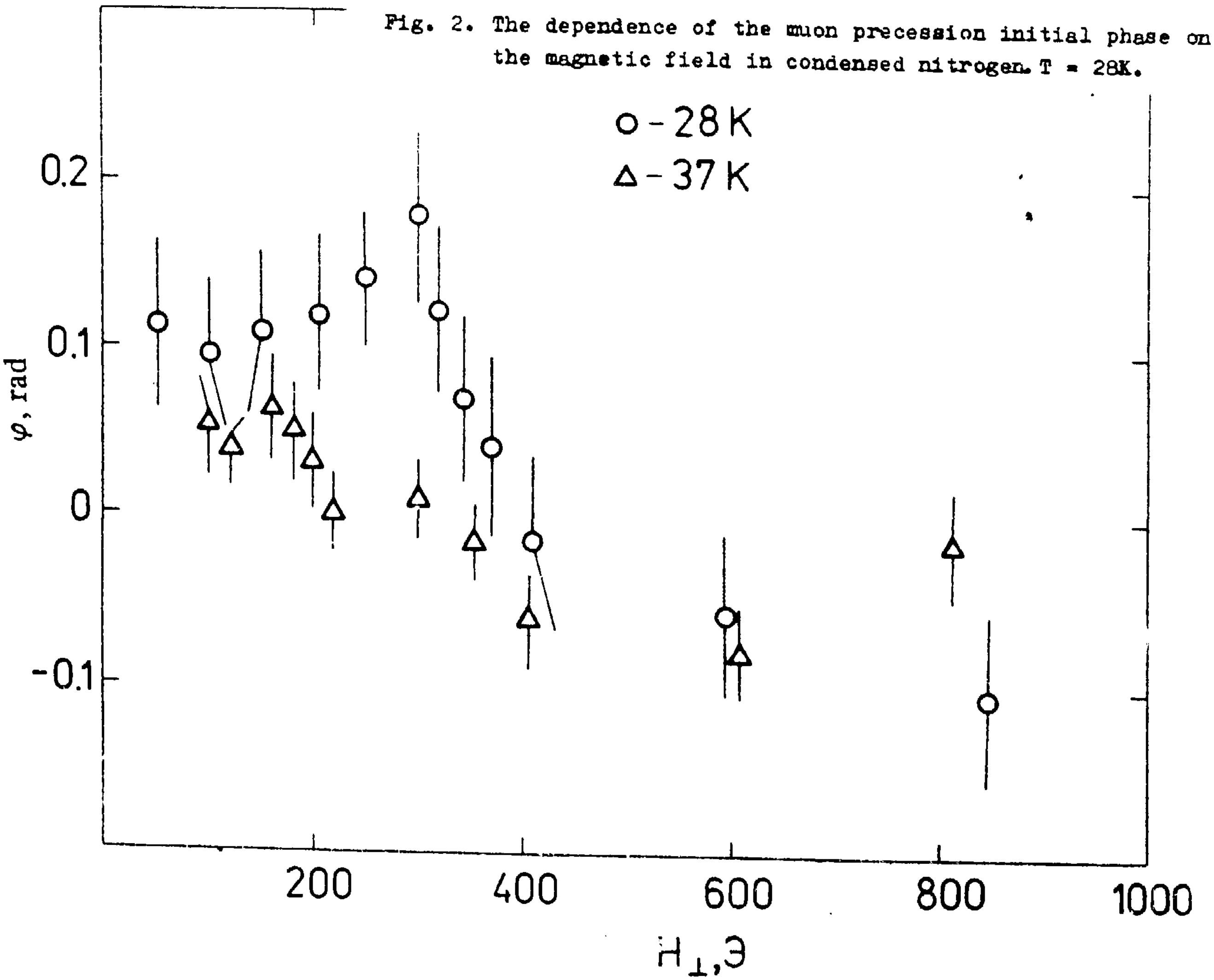

Fig. 2. The dependence of the muon precession initial phase on the magnetic field in condensed nitrogen. T = 28K.

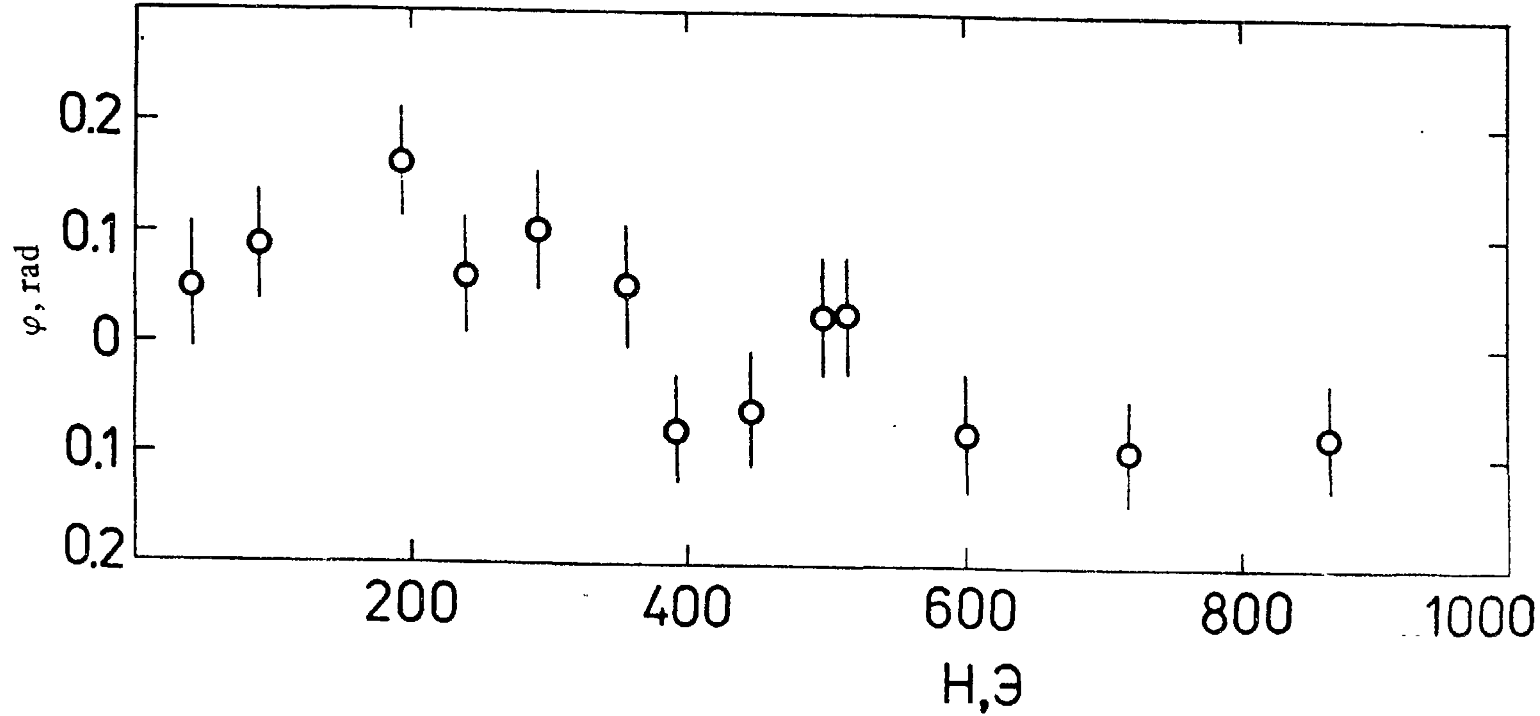

Fig. 3. The dependence of the muon precession initial phase on the magnetic field in condensed nitrogen . T = 55 K.

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