

Physics and chemistry of the μ^+ meson and muonium

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This review of the physics and chemistry of muonium and the μ^+ meson includes experiments on the weak interaction, investigation of the properties of solids by means of μ^+ mesons, and problems of the chemistry of the hydrogen-like atom muonium. A new phenomenon is described—the two-frequency precession of muonium, and a means is presented for determination of the contact magnetic field at the μ^+ meson in ferromagnetic materials. The theory of depolarization of μ^+ mesons in matter is presented. Results are given of chemical reactions of muonium.

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INTRODUCTION

This review is a brief report of a major series of studies of the physics and chemistry of μ^+ mesons and muonium, carried out jointly by investigators at three institutes: the I. V. Kurchatov Atomic Energy Institute, the Institute of Theoretical and Experimental Physics, and the Joint Institute for Nuclear Research. This series of studies includes experiments on the weak interaction in $\mu^+ \rightarrow e^+$ decay, on studies of the properties of solids by means of μ^+ mesons, and problems of the chemistry of the hydrogen-like muonium atom. All these studies have as a common ground the problem of studying the interactions of μ^+ mesons with matter, which arises to one degree or another in any of the experiments presented. All of the experiments were carried out in the JINR synchrotron at Dubna.

1. STUDY OF THE WEAK INTERACTION IN $\mu^+ \rightarrow e^+$ DECAY

We shall begin with a study of the angular distributions of the positrons of $\mu^+ \rightarrow e^+$ decay relative to the direction of the μ^+ -meson spin.^{1,12} We shall first discuss briefly the question of what kind of information can be obtained from experimental investigation of $\mu^+ \rightarrow e^+ + \nu_e + \bar{\nu}_\mu$ decay.

Experimentally in $\mu^+ \rightarrow e^+$ decay we can measure the μ^+ -meson lifetime and the energy spectrum, angular distribution, and polarization of the decay positrons. The most general form of the weak-interaction Hamiltonian leads to the following spectrum of positrons produced in decay of stationary, completely polarized μ^+ mesons:

$$dN(x, \theta) = \{M(x, \rho, \eta) + \xi B(x, \delta) \cos \theta\} x^2 dx d\theta$$

Here x is the positron momentum, θ is the angle between the directions of the positron momentum and the μ^+ -meson spin, ρ , η , ξ , and δ are parameters representing bilinear combinations of the constants of the weak-interaction Hamiltonian; $M(x, \rho, \eta)$ and $B(x, \delta)$ are functions determining the energy dependence of the isotropic and anisotropic parts of the spectrum.¹³ Thus, from an experiment on $\mu^+ \rightarrow e^+$ decay it is possible to determine a total of six parameters: ρ , ξ , δ , and η from the spectrum $dN(x, \theta)$ and in addition τ_0 —the μ^+ -meson lifetime, and h —the helicity of the decay positron.

It is well known that the data existing at the present time on weak interactions are in good agreement with the theory of a universal $V-A$ interaction. The theoretical values of the $V-A$ parameters of $\mu^+ \rightarrow e^+$ decay are as follows: $\rho = \delta = 3/4$; $\xi = -h = 1$; $\eta = 0$.

An experimental check of $V-A$ theory in $\mu^+ \rightarrow e^+$ decay requires the most accurate possible determination of the parameters ρ , δ , and ξ , and so forth. Studies in this direction continue to be carried out in a number of laboratories up to the present time. The current experimental values of the parameters ρ , δ , ξ , η , h , and τ_0 are given below: $\rho = 0.756 \pm 0.006$, $\delta = 0.754 \pm 0.009$, $\xi = 0.974 \pm 0.014$, $\eta = -0.8 \pm 0.4$, $h = -1.00 \pm 0.13$, $\tau_0 = (2.19711 \pm 0.00008) \times 10^{-6}$ sec.¹⁾ The studies being presented are devoted to determination of the parameter ξ , which was previously known substantially less accurately than the parameters ρ and δ of the spectrum of positrons from $\mu^+ \rightarrow e^+$ decay. The value obtained $\xi = 0.975 \pm 0.15$ remains the most accurate value up to the present time. The average experimental value of ξ presented above is determined mainly by this result.

For a precise measurement of the parameter ξ it is convenient to use the angular distribution of $\mu^+ \rightarrow e^+$ -decay positrons indicated over energy

$$dN(\theta) \sim 1 + (\xi \cos \theta)/3,$$

which is practically independent of the values of the remaining five parameters which determine $\mu^+ \rightarrow e^+$ decay. We therefore chose as a detector of $\mu^+ \rightarrow e^+$ decay events nuclear emulsion, which has the following advantages:

1) In nuclear emulsion it is possible to measure accurately the emission angles of positrons from $\mu^+ \rightarrow e^+$ decay.

2) In measurements in nuclear emulsion there is essentially no energy threshold for detection of $\mu^+ \rightarrow e^+$ decay positrons and therefore the true integrated-over-energy positron angular distribution is measured.

¹⁾The most accurate value, measured recently in the JINR cyclotron in the experiments of M. P. Balandin, V. M. Grebenyuk, V. G. Zinov, A. D. Konin, and A. N. Ponomarev. See JINR preprint R1-7892, Dubna, 1974.

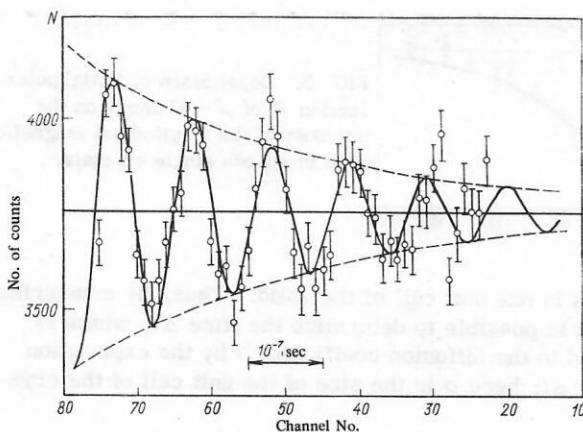


FIG. 1. Precession of triplet muonium in crystalline quartz. Intensity of transverse magnetic fields 7.17.

3) In measurements in nuclear emulsion there is no kinematic polarization of μ^+ mesons, since the π^+ mesons decay at rest.

The principal difficulty arising in measurement of the angular distribution of $\mu^+ \rightarrow e^+$ decay positrons in nuclear emulsion (and in any material) is that the medium where the μ^+ meson slows down and stops (depolarizes) it. To suppress the depolarizing influence of the emulsion we used a longitudinal magnetic field (along the direction of the μ^+ -meson spin) of strength up to 140 kOe. This field was obtained in a volume of 0.7 liter in a pulsed mode by discharging into a specially constructed solenoid a capacitor bank of capacitance 0.1 F charged to 2000 volts (2×10^5 joules). The pulsed magnetic field was synchronized with the operation of the accelerator. We achieved a high degree of reliability of operation of the synchronization circuit, which is necessary with the emulsion method of detection of $\pi^+ \rightarrow \mu^+ \rightarrow e^+$ decay events. The ξ value mentioned above was obtained in measurement in emulsion of about 400 thousand cases of $\pi^+ \rightarrow \mu^+ \rightarrow e^+$ decay.

The value obtained $\xi = 0.975 \pm 0.015$ agrees within experimental error with $\xi = 1$ which follows from the $V-A$ theory of weak interactions, and is thus an experimental confirmation of this theory.

A more general form than $V-A$ is the $V - \varepsilon A$ interaction, which corresponds to a two-component neutrino. The ξ value found permits estimation of the relative contributions of V and A interactions to $\mu^+ \rightarrow e^+$ decay, i.e., the coefficient $\varepsilon = |\varepsilon| \exp(i\varphi)$. In two-component neutrino theory the relation between ξ and ε has the form $\xi = 2\varepsilon \cos\varphi / (|\varepsilon|^2 + 1)$. From this relation and the measured value of ξ it is possible to obtain the estimates $0.75 \leq |\varepsilon| \leq 1.34$, which are also the most accurate at the present time.¹⁴

2. STUDY OF SOLIDS BY MEANS OF μ^+ MESONS

The muonic method of studying the properties of matter has recently been undergoing intensive development. The proposed new method of studying matter and, in particular, solids by means of μ^+ mesons is based on the fact that the μ^+ meson is a labeled particle whose

spin direction can be traced on the basis of the asymmetry of the angular distribution of positrons from $\mu^+ \rightarrow e^+$ decay. Interaction of the magnetic moment of the positive muon with the medium leads to a loss of polarization by the muon. By measuring the residual polarization of muons as a function of time and in magnetic fields of various strengths and directions it is possible to study a wide range of physical and chemical properties of the material. Below we describe briefly studies of several properties of solids carried out by means of μ^+ mesons.

The size of a muonium impurity atom in matter.¹⁵⁻²⁸

Muonium is a hydrogen-like atom consisting of a μ^+ meson and an electron. The muonium atom is formed on slowing down of a μ^+ meson in matter, when its velocity becomes comparable with the velocities of the atomic electrons. The muonium formed is then slowed down to thermal velocities (in a time of about 10^{-11} sec).

The observation and study of atomic muonium in matter is an important problem, since this is necessary for study of the conditions of its formation and subsequent interactions. In view of the high reactivity of muonium, like atomic hydrogen, it is natural to expect that muonium can continue to exist as an atomic system only in chemically inert materials. The studies which have been carried out^{24,25} have confirmed this assumption—the precession of a muonium impurity atom in a transverse magnetic field was first observed in condensed media—in crystalline and fused quartz (Fig. 1), carbon dioxide, ice (-196°C), and germanium single crystals²⁶ at -196°C .

The size of the muonium impurity atom in matter, or more precisely the frequency ω_0 of the hyperfine splitting of muonium, has been determined by two methods. We shall first describe the results obtained by the method of two-frequency precession of muonium.

The phenomenon of two-frequency precession or beating was predicted at the Kurchatov Atomic Energy Institute and was observed experimentally by Gurevich.¹⁷ This phenomenon is that the time dependence $P(t)$ of the μ^+ -meson spin direction in a transverse magnetic field is determined by two frequencies:

$$P(t) = (\cos \omega t \cos \Omega t)/2,$$

where $\omega = eB/(2m_e c)$ is the Larmor frequency precession of muonium (m_e is the electron mass) in a field B ; $\Omega = \omega^2/\omega_0$ is the beat frequency. The experimental dependence of $P(t)$ in fused quartz is shown in Fig. 2. This two-frequency dependence $P(t)$ is explained by the fact that the μ^+ -meson spin interacts with the external field B and with the spin of the muonium electron. As can be seen from the expression for $P(t)$, experimental observation of two-frequency precession permits determination of the frequency of hyperfine splitting of the ground state of the muonium atom.

Experiments on determination of ω_0 by the method of two-frequency precession have been carried out in ice, quartz, and germanium. It was found that in ice and quartz the frequency ω_0 corresponds to the vacuum value

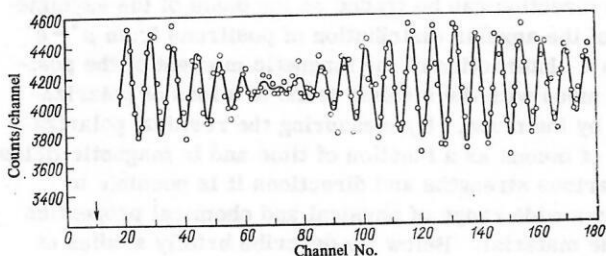


FIG. 2. Two-frequency precession (beating) of the spin of the μ^+ meson of muonium in fused quartz: solid line—theoretical dependence $P(t)$ with parameters ω and Ω selected by the method of maximum likelihood; time-analyzer channel width 1 nsec; $B=98$ Oe. The arrow shows the channel corresponding to $t=0$.

of this quantity $(\omega_0)_{\text{vac}}/2\pi = 4463.25$ MHz, while in germanium $(\omega_0)_{\text{Ge}} = 0.58(\omega_0)_{\text{vac}}$. This result shows that the size of the muonium impurity atom in ice and quartz is the same as in vacuum. In germanium the muonium impurity atom is dilated, so that its Bohr radius $a \sim (\omega_0)^{-1/3}$ is greater than in vacuum: $a_{\text{Ge}} = (1.20 \pm 0.01)a_{\text{vac}}$.

The energy of the hyperfine splitting in the muonium atom has been determined also on the basis of restoration of the polarization of μ^+ mesons in longitudinal magnetic fields.²⁷ For quartz²⁷ it was found that $(\omega_0)_{\text{SiO}_2} = (1.02 \pm 0.05)(\omega_0)_{\text{vac}}$, which agrees with the vacuum value; in the same way it was shown that muonium under these conditions behaves as an isolated atom. A similar correspondence to the vacuum values was obtained for corundum.²⁷ At the same time in a semiconducting material (silicon of high purity) the method of longitudinal magnetic fields²⁸ gave a value $(\omega_0)_{\text{Si}} = (0.41 \pm 0.03)(\omega_0)_{\text{vac}}$, i.e., the radius of the Bohr orbit is $a_{\text{Si}} = 0.719 \pm 0.016 \text{ \AA} = (1.35 \pm 0.03)a_{\text{vac}}$ (Fig. 3).

The experimental data obtained in these studies have important significance for experimental verification of theories describing the state of an impurity atom in matter. Checking of existing theories by this means, which became possible after the experiments mentioned above on determination of the frequency ω_0 of a muonium impurity atom, has been carried out by Wang and Kittel.^{23, 29}

Diffusion of μ^+ mesons in solids.²⁹⁻³²

The method of determining the diffusion of a μ^+ meson is based on observation of the dipole interactions of the magnetic moments of the μ^+ meson and the nuclei of the material. Dipole interactions appear in that the spins of different μ^+ mesons precess with different frequencies. Therefore the experimentally observable precession of a μ^+ meson in a transverse magnetic field damps out. On diffusion of a μ^+ meson over the crystal lattice the nuclear magnetic fields in it become variable in time, as a result of which the damping of the precession amplitude decreases. The damping rate Λ (rate of dipole relaxation of the μ^+ -meson spin) of the diffusing μ^+ meson is determined by the time Δt which the μ^+ meson

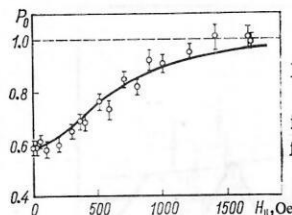


FIG. 3. Dependence of initial polarization P of $\mu^+ \rightarrow e^+$ decay on the intensity of the longitudinal magnetic field in silicon single crystals.

spends in one unit cell of the solid. Thus, by measuring Λ ,³⁰ it is possible to determine the time Δt , which is related to the diffusion coefficient D by the expression $D = \alpha^2/\Delta t$; here α is the size of the unit cell of the crystal.

The temperature dependence $\Lambda(t)$ in copper is shown in Fig. 4. It is evident from this figure that with increase of the temperature the relaxation rate Λ of the μ^+ -meson spin falls off. The constancy of the value of Λ for $T \lesssim 60^\circ \text{K}$ is explained by the fact that during the time of observation (which is limited by the lifetime of the muon) the μ^+ meson in copper at these temperatures practically does not diffuse at all.

This method was used for the first measurements of the temperature dependence of the probability of the diffusion transition $1/\Delta t$ in one of the materials (in copper). The dependence $1/\Delta t = f(T)$ in copper is well described by the expression $1/\Delta t = \nu_\mu \exp(-Q_\mu/T)$, where $\nu_\mu = 10^{7.50 \pm 0.02} \text{ sec}^{-1}$ and $Q_\mu = 560 \pm 15^\circ \text{K}$; the temperature T is expressed in degrees Kelvin. The experimental dependence $1/\Delta t = f(1/T)$ for a polycrystalline sample of copper is shown in Fig. 5.

The proposed method for study of the diffusion of a μ^+ meson permits studying under the same experimental conditions the diffusion of the light isotope of the proton—the μ^+ meson—in a broad class of materials, for example, in metals.

The possibility of observing the diffusion of such a light particle as the μ^+ meson has led to discovery of a new phenomenon—sub-barrier diffusion of the μ^+ meson. The sub-barrier nature of the diffusion of the μ^+ meson follows from the anomalously small value of the pre-exponential factor $\nu_\mu = 10^{7.5} \text{ sec}^{-1}$, which for diffusion of protons in copper and other metals is $\nu_p = 10^{13} \text{ sec}^{-1}$. The coincidence of the experimental value $\nu_\mu \approx 10^{13} \text{ sec}^{-1}$ with the frequency of vibration of particles in the crystal lattice of a metal permits interpretation of the obtained dependence $1/\Delta t = \nu_p \exp(-Q_p/T)$ for a proton (and for heavier particles) as diffusion over the barrier, where $1/\Delta t$ is the probability of jumping of particles from one unit cell to another, and Q_p is the height of the potential barrier. It was found that the height of the potential barrier Q_p for the proton in copper is $(Q_p)_{\text{Cu}} = 4600^\circ \text{K}$. The value $\nu_\mu = 10^{7.5} \text{ sec}^{-1}$ is too small to be interpreted as the frequency of vibration of the μ^+ meson in the copper lattice. Hence it follows that diffusion of a μ^+ meson in copper cannot be over the barrier and the parameter ν_μ is not, as for the proton, the frequency of vibrations.

²⁹For more detail see the review by I. I. Gurevich and B. A. Nikol'skii, preprint IAE-2437, 1974.

³⁰The relation of the quantities, D , Λ , and Δt can be found in the book by Abragam.^[33]

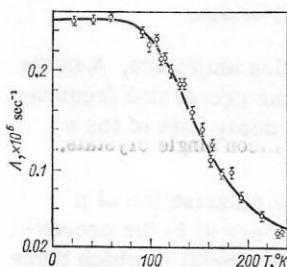


FIG. 4. Temperature dependence of rate of damping of the amplitude of μ^+ -meson precession in a polycrystalline sample of copper: the smooth curve is the theoretical dependences $\Lambda(T)$ obtained on the assumption that the time Δt determining the rate of damping depends on the temperature in accordance with $f(T)$ with parameters $\nu_\mu = 10^{11.5} \text{ sec}^{-1}$ and $Q_\mu = 560^\circ \text{K}$.

A simple calculation shows that the low mass of the μ^+ meson should lead to sub-barrier diffusion of these particles, which also explains the experimentally observed features of μ^+ -meson diffusion in copper. For sub-barrier diffusion the probability $1/\Delta t$ has the following form:

$$1/\Delta t = \nu_0 \exp \left[-2 \sqrt{2mv} b/h \right] F(T).$$

Here ν and b are respectively the height and width of the barrier; m is the mass of the μ^+ meson. The low value of the experimental parameter $\nu_\mu < 10^{13} \text{ sec}^{-1}$ is, thus, the result of the low transparency of the potential barrier. The parameter Q_μ which determines the temperature dependence $F(T)$ in the expression for $1/\Delta t$ can be interpreted as the activation energy which must be expended in widening the threshold to the neighboring cell when the μ^+ meson diffuses.

The experimental observation of sub-barrier diffusion of μ^+ mesons opens up additional possibilities for checking and determining parameters in the theories which describe the diffusion of light atoms in solids.

As was pointed out at the beginning of this section, diffusion of a μ^+ meson is observed as the result of dipole interactions of the magnetic moments of the μ^+ meson and the nuclei of the material in which the μ^+ meson is diffusing. The magnetic-dipole interaction of the μ^+ meson in matter can be used also to obtain another interesting piece of information on the structure of the material. It is possible by this method to study the location of the impurity atom of muonium (the μ^+ meson)

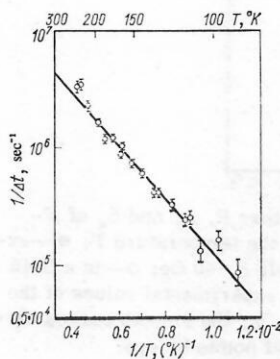


FIG. 5. Dependence of $1/\Delta t = f(1/T)$ for a polycrystalline sample of copper: The straight line is the dependence $1/\Delta t = \nu_\mu \exp(-Q_\mu/T)$ with parameters ν_μ and Q_μ chosen by the method of maximum likelihood.

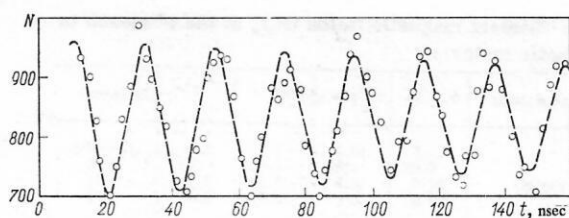


FIG. 6. Precession of μ^+ -meson spin in iron for $B=0$: the smooth curve is the theoretical dependence $N(t) = N_0[1 - a \times \exp(-\Lambda t) \cos \omega t]$ with parameters N_0 , a , Λ , and ω chosen by the method of maximum likelihood; the theoretical and experimental dependences of $N(t)$ are "corrected" for the decay exponential $\exp(-\Lambda_0 t)$ of the μ^+ meson ($\Lambda_0 = 4.55 \times 10^5 \text{ sec}^{-1}$).

in the crystal lattice, to determine the parameters of the μ^+ molecule, and so forth. In an earlier paper³⁰ we have studied the isotopic effect: The difference in the muon-spin relaxation rate in ice of ordinary and heavy water. As a result we concluded that the time Δt which the muonium impurity atom spends in one unit cell of ice is $\Delta t > 0.5 \times 10^{-6} \text{ sec}$.

The magnetic field at the μ^+ meson in ferromagnetic materials³⁴⁻³⁶

The possibility has been demonstrated of observing experimentally the precession of a μ^+ meson in iron, nickel, gadolinium, and cobalt. Precession of a μ^+ meson in these ferromagnetic materials is observed also in the absence of an external magnetic field B . Precession of a μ^+ meson in iron for $B=0$ is shown in Fig. 6.

The frequency ω_μ of precession of a μ^+ meson in a ferromagnetic material is determined by the magnetic field B_μ at the μ^+ meson:

$$\omega_\mu = eB_\mu/(mc),$$

where m is the mass of the μ^+ meson. The local field B_μ is determined by the external field B and the internal magnetic fields in the ferromagnetic material:

$$B_\mu = B + B_{\text{dip}} + (B_c)_\mu.$$

Here B_{dip} is the dipole field of the magnetized atoms, i. e., of the localized atomic electrons; $(B_c)_\mu$ is the contact field of the polarized electrons at the μ^+ meson, mainly of collectivized conduction electrons. For $B=0$ the field B_μ is determined only by the internal fields of the magnetized domains of the ferromagnetic material. The very fact of observation of a definite value B_μ for $B=0$ means that the magnetic field B_{dom} in all domains of an unmagnetized ferromagnetic material is a completely determined quantity. An analysis carried out shows that $B_{\text{dom}} = B_{\text{sat}}$, where B_{sat} is the saturation magnetic induction; $B_{\text{sat}} = 4\pi M_{\text{sat}}$, where M_{sat} is the saturation magnetization. The equality $B_{\text{dom}} = B_{\text{sat}}$ is preserved also on magnetization of the ferromagnetic material, i. e., for $B > 0$.

Another result of study of ferromagnetic materials by means of μ^+ mesons is the determination of the contact field $(B_c)_\mu$ at the μ^+ meson. The field $(B_c)_\mu$ is deter-

TABLE I. Contact magnetic fields $(B_c)_\mu$ at the μ^+ meson in ferromagnetic materials.

Ferromagnetic material	B_{sat} , kG	$(B_c)_\mu$, kG	P_e^{min} , %	$(B_c)_n$, kG	Reference
Iron	21.6	10.7	-6.50	1.4	[37-39]
Nickel	6.08	0.69	-0.42	0.75	[40]
Gadolinium (130° K)	24.0	6.3	-3.85	2.9	[41]
Cobalt	17.9	6.0	-3.65	2.0	[42]

Note. P_e^{min} are the minimum values of polarization of the conduction electrons of a ferromagnetic material; $(B_c)_n$ are the contact fields at the neutron, measured in experiments on magnetic scattering of neutrons.

mined from the expression for B_μ , in which the field B_μ is measured experimentally, and the dipole field B_{dip} can be obtained quite reliably by theoretical means. The values of $(B_c)_\mu$ obtained in this way for various ferromagnetic materials are given in Table I.

The contact fields $(B_c)_\mu$ in a ferromagnetic material are determined by the polarization P_e of the conduction electrons and the density $\rho(0)$ of the electron wave function at the μ^+ meson:

$$(B_c)_\mu = 8\pi\beta_e P_e \rho(0)/3,$$

where β_e is the magnetic moment of the electron. It follows from this relation that the field $(B_c)_\mu$ is determined by $P_e \rho(0)$, i. e., by the electron magnetic moment at the μ^+ meson. The values found for $(B_c)_\mu$ permit estimation of the lower limit of polarization of conduction electrons of the ferromagnetic material, which is obtained if we take as $\rho(0)$ the maximum possible value of this quantity $[\rho(0)]_{\text{max}} = 2.1 \times 10^{24} \text{ cm}^{-3}$ equal to the density of the electron wave function at the μ^+ meson in the free muonium atom. The minimum possible values P_e^{min} obtained in this way for various ferromagnetic materials are given in the table; the negative sign of P_e^{min} means that the direction of polarization of the conduction electrons, like the direction of the field B_c , is opposite to the direction of the magnetization vector M .

The values found for $(B_c)_\mu$ can be compared with the values of the contact fields $(B_c)_n$ of the conduction electrons at a neutron, obtained by the method of magnetic scattering of polarized neutrons in a ferromagnetic material. Values of $(B_c)_n$ are also given in the table. The direction of the fields $(B_c)_n$ at the neutron, like those of the fields $(B_c)_\mu$ at the μ^+ meson, are opposite to the direction of M . From comparison of the values of $(B_c)_\mu$ and $(B_c)_n$ it is possible to evaluate experimentally the degree of deformation of the electron wave function of the ferromagnetic material at the μ^+ meson, i. e., at an impurity center with charge $Z = +1$.

Investigation of magnetic phase transitions in gadolinium⁴³

Phase transitions in gadolinium have been studied in the temperature range $T = (100-200)^\circ\text{K}$. The precession of the μ^+ mesons was observed, as usual, by means of counters detecting the positrons from $\mu^+ \rightarrow e^+$ decay. The counting rate $N(T)$ of the positron counter telescope can be represented as follows:

$$N(t) \sim \exp(-\Lambda_0 t) [1 - a \exp(-\Lambda t) \cos \omega_\mu t],$$

where a is the μ^+ -meson precession amplitude, Λ is the precession damping rate, ω_μ is the precession frequency, and $\Lambda_0 = 4.55 \times 10^{-5} \text{ sec}$ is the decay rate of the μ^+ meson.

The amplitude a determines the polarization of μ^+ mesons in gadolinium $P = a/a_0$, where a_0 is the precession amplitude of the meson in a material in which there is no depolarization of the μ^+ meson. The frequency ω_μ determines the average magnetic field B_μ at the μ^+ meson: $\omega_\mu = eB_\mu/(mc)$. The damping rate Λ characterizes the degree of inhomogeneity of the magnetic field δB_μ at individual μ^+ mesons.

The dependence of the experimental parameters P , Λ , and B_μ on temperature is shown in Fig. 7, where a rapid change in all of the precession parameters P , ω , and Λ is easily visible at a temperature $T_c = 289^\circ\text{K}$ for gadolinium, and also at $T_0 \approx 235^\circ\text{K}$. At $T = T_0$ the magnetic induction B_μ drops by a factor of two, the relaxation rate Λ rises rapidly, and the μ^+ -meson polarization P decreases substantially.

The entire temperature interval $T = 235-285^\circ\text{K}$ is also unusual for a normal ferromagnetic material. The field B_μ at these temperatures remains approximately constant, which does not agree with the form of the Brillouin dependence $B_\mu(T)$. The polarization P is unusually small, which indicates the existence of spin relaxation processes fast in comparison with the time of observation for an appreciable fraction of the μ^+ mesons. For

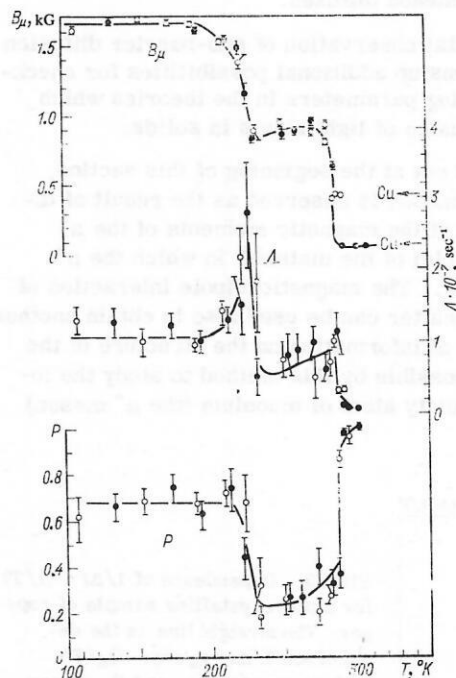


FIG. 7. Dependence of the parameters P , Λ , and B_μ of μ^+ -meson precession in gadolinium on the temperature T : \bullet —experimental values in an external field $H = 60 \text{ Oe}$; \circ —in a field $H = 450 \text{ Oe}$; the arrows indicate the experimental values of the field B_μ in copper at $H = 60$ and 450 Oe ; the smooth curves have been drawn through the experimental points by eye.

$T < T_0$ the parameters P , Λ , and B_μ are practically independent of temperature. The increase of Λ at $T = T_0$ indicates a sharp rise in the degree of inhomogeneity of the fields B_μ at the temperature T_0 of the phase transition, the nature of which remains unclear in studies by known methods up to the present time. The value $P = 2/3$ for $T < T_0$ is natural for an unmagnetized (or weakly magnetized) ferromagnetic material and corresponds to an isotropic distribution of the directions of magnetization of the individual domains.

The muonium mechanism of μ^+ -meson depolarization in matter^{11, 44-51}

Above we have considered the dipole relaxation of the μ^+ -meson spin in matter. This slow relaxation (characteristic relaxation time about 10^{-6} sec) is observed experimentally in the absence of μ^+ -meson diffusion. However, another fast process for μ^+ -meson spin relaxation exists which usually occurs in a time $\leq 10^{-9}$ sec. The entire set of experimental data on fast relaxation of the μ^+ -meson spin is satisfactorily explained if we assume that this relaxation occurs in the formation of muonium.

As we have already pointed out, muonium is formed on slowing down of the μ^+ meson to the velocities of the atomic electrons and exists as a paramagnetic atom until it enters into a chemical bond or another diamagnetic compound with the molecules of the medium. The lifetime of the paramagnetic muonium atom we shall designate as τ . The muonium atom can have spin $I = 0$ and $I = 1$. Formation of muonium in the state with $I = 0$ leads to depolarization of the μ^+ -meson spin as the result of hyperfine interactions in a time of about $1/\omega_0 = 3.6 \times 10^{-11}$ sec. On formation of muonium in a state with $I = 1$ the μ^+ meson is not depolarized in view of the conservation of angular momentum. However, the state with $I = 1$ does not remain unchanged. As the result of interaction with the medium the electron of the muonium changes the direction of its spin, which leads to transitions $(I = 1) \rightleftharpoons (I = 0)$, which lead to further depolarization of the μ^+ meson. The frequency of depolarization of the muonium electron we shall designate as ν . Measurement of the residual polarization of the μ^+ meson in matter after the muonium stage permits determination of the parameters τ and ν characterizing the interaction of muonium in this material. We note that the parameter τ is the rate of chemical reaction of muonium—the light isotope of the hydrogen atom.

The fast relaxation of the μ^+ -meson spin has been studied experimentally in various materials in a transverse magnetic field (with respect to the μ^+ -meson spin). A strong transverse magnetic field ($\gtrsim 1$ kG) should lead to a rapid precession of the μ^+ -meson spin in the muonium stage, and consequently, to an additional depolarization of the μ^+ meson as a result of the fact that the spins of the individual μ^+ mesons are rotated by different angles in the time before the muonium enters into a chemical reaction. No additional depolarization of the μ^+ meson in transverse magnetic fields has been observed experimentally. This effect can be explained by the high frequency $\nu \gtrsim \omega_0$.

Theory of depolarization of positive muons in matter^{11, 21, 48-51, 50-56}

A polarized positive meson in matter captures an electron in the thermalization process, forming a hydrogen-like muonium atom μ^+e^- . Here atoms with total spin projection on the quantization axis equal to unity and zero are formed with equal probability. In the latter case the initial state is not stationary for the spin Hamiltonian of the muon+electron system and therefore in it the muon polarization oscillates rapidly with the frequency of the hyperfine interaction ($\omega_0 \approx 3 \times 10^{10}$ sec⁻¹). As a result the contribution of this state to the observed average polarization decreases and in the absence of a magnetic field is equal to zero. The state with total angular momentum projection equal to unity is stationary and the muon polarization is preserved in it. In accordance with this picture in the absence of a magnetic field the observed polarization of a muon (averaged over a time of about 10^{-10} sec) should be $1/2$. However, smaller values have repeatedly been observed experimentally. This fact can be explained if we take into account the fact that as the result of interaction of the electron with the medium its spin relaxes with a frequency ν , which leads to subsequent depolarization of the muon spin.

Nosov and Yakovleva^{11, 48, 49} have made the suggestion, which is extremely important for all subsequent theory, that in a medium (in particular, in solids) the chemically active muonium atom (the light isotope of hydrogen) can enter into a chemical reaction, forming a diamagnetic chemical compound. The characteristic reaction time τ was a new phenomenological parameter of the theory. In these same studies the phenomenological equations for the spin density matrix of muonium in magnetic fields were formulated. These equations were solved for the two limiting cases $\nu \ll \omega_0$ and $\nu \gg \omega_0$ for a magnetic field parallel to the initial polarization, which immediately permitted explanation of the dependence of the residual polarization, observed in emulsions, on the longitudinal magnetic field and made it possible to indicate the magnetic field value necessary for practically complete restoration of polarization.⁶ These indications of the theory have been used in Refs. 1-12.

Ivanter and Smilga^{50, 51} obtained a new solution of the system of equations for the muonium spin matrix for arbitrary relations of the parameters ν , ω_0 , and τ in magnetic fields parallel and perpendicular to the initial polarization.

In fields parallel to the initial polarization of the muons, for the so-called residual or medium polarization, they found the formula

$$P/[2(1-P)] = (1 + 2\nu\tau)/(\tau^2\omega_0^2) + (Y_2 + X^2)/(1 + 2\nu\tau),$$

where $X = \omega'(1 + \zeta)/\omega_0$; ω' is the frequency of precession of the electron spin in an external magnetic field; $\zeta = m_e/m_\mu = 1/207$ is the ratio of the magnetic moment of the μ meson to the magnetic moment of the electron. The physical meaning of the quantity X is obvious—it is the dimensionless external magnetic field. It should be emphasized that the frequency of hyperfine interaction ω_0 in matter is different from the vacuum frequency

and is a phenomenological parameter of the theory. For the case in which the magnetic field is perpendicular to the initial polarization, simple relations for the residual polarization were also obtained. A detailed study was made of the solutions obtained, which permitted, in particular, prediction and development of a complete theory for the phenomenon of two-frequency precession,^{16, 21, 50} which was later observed experimentally,¹⁵⁻²³ and also stopping of the precession of a μ^+ meson in a very strong magnetic field. The results obtained also permitted theoretical interpretation of the previously unclear data on μ^+ -meson depolarization in transverse magnetic fields.⁴⁶ The results also permitted development of a complete phenomenological theory analyzing the chemical reactions of muonium, and in particular made it possible to take into account the possibility of superthermal reactions (the "hot chemistry" channel) and to propose a new method for studying chemical reactions by observation of the initial phase of muonium precession at the mesonic frequency.

The theory developed for analysis of muonium chemical reactions has permitted interpretation of the experimental data and, in particular, accomplishment of muon depolarization in KCl crystals.⁵² In another article Ivanter and Smilga⁵³ proposed a method of studying the magnetic structure of Type II superconductors by studying the precession of μ^+ mesons in transverse magnetic fields and demonstrated the sensitivity of this method to changes in the magnetic structure of Type II superconductors.

In several articles^{49, 54-56} a complete phenomenological theory of the behavior of muon depolarization has been developed in the case of charge exchange (alternating ionization of the muonium atom and electron capture by the muon), which is of interest for analysis of the behavior of muon polarization in semiconductors.

3. CHEMICAL INTERACTIONS OF THE MUONIUM ATOM

Entering into a chemical reaction by a muonium atom, as has been pointed out, fundamentally changes its depolarization factors in a medium.^{11, 57-59} Depending on the muonium chemical reaction rate, the residual polarization of the μ^+ meson in the composition of the molecule changes, which permits us to obtain a relation between the absolute rate constant of the process and the asymmetry coefficient of $\mu^+ \rightarrow e^+$ decay at the meson and muonium frequencies.

The nuclear physics time standards in study of chemical reactions of muonium are the following constants: the frequency of transitions between the (1, 0) and (0, 0) hyperfine structure states and the decay constant of the μ^+ meson, with respect to which the reaction rates are determined. We note that it is possible to observe directly the kinetics of the process and to study the stationary states of the reaction products obtained.

Let us formulate the features present in the muon method of study of chemical processes. First, one of the characteristic conditions is the existence in the entire volume of material studied of only single muonium

atoms, which is due to the specific technical procedure of the experiment. Thus, study of the kinetics of processes occurs with a negligibly small depth of transformation of the initial material, i.e., under initial conditions not complicated by the presence of secondary particles formed in the course of the reaction. Second, use of nuclear-physics constants as independent time standards permits study of a wide spectrum of fast processes and determination of absolute reaction rate constants, which in combination with the short times of observation permits study of processes at a level close to the elementary interaction events. Third, observation of the behavior of the spin and magnetic moment of the μ^+ meson provides the possibility of studying the factors which determine the spin interaction, in the kinetics of the process or under stationary conditions. Fourth, the typical independence of the detection technique of external parameters of the experiment substantially increases the possibility of investigations in different aggregate states of matter.

With use of external magnetic fields to separate the mesonic and muonium components of precession as a methodological condition of the experiment, it is desirable to group the reaction products, including muonium, in accordance with magnetic moment, separating molecules with a diamagnetic electron shell and compounds which have an unpaired electron. In this case taking into account of the kinetics of the processes permits determination of the mechanism of muonium chemical reactions, separating the rate constants for formation of molecular and radical reaction products.

The absolute rate constants of muonium reactions have been determined for a number of organic compounds, and the dependence of the reactivity of covalent organic molecules on their structure has been studied.⁶⁰⁻⁶⁹ As for atomic hydrogen, muonium reactions with unsaturated compounds are dominated by addition reactions with a multiple bond with formation of the corresponding radical product. Absolute rate constants have been determined for substitution and addition reactions with successive methylation of the benzene ring and of the side chain, and with successive hydriding of unsaturated compounds. The dependence of the reactivity of organic compounds on the type of halogen in the ring has been studied, as well as on introduction of several halogen atoms into the side chain. It has been shown that redistribution of substitution and addition reaction rates occurs within one or two C-C bonds.

Study of the temperature dependence^{63, 64} of muonium reaction rates has permitted determination of the activation energy of interactions with organic (cyclohexane, bromoform, bromobenzene) and inorganic (water) compounds. For the water-ice phase transition a sharp jump is observed in the asymmetry coefficient, corresponding to the change in the reaction rate by more than two orders of magnitude (Fig. 8); the activation energies of processes above and below 0° are close to zero. As we have pointed out, in the crystalline structure of ice, precession of the triplet muonium atom has been identified.²⁵ The data obtained confirm the idea that a coordination-closed system of hydrogen bonds (Fig. 9) is

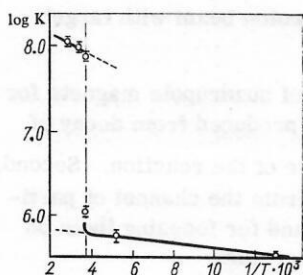


FIG. 8. Temperature dependence of the rate constant for interaction of muonium with water (ice).

formed in the process of crystallization of ice and that there are no free sites for stabilization of muonium in this system.

The influence of longitudinal magnetic fields on the polarization of μ^+ mesons in unsaturated chemical compounds has been studied for several monomeric⁷⁰ and polymeric^{65,66} materials (Fig. 10). The change in the strength of the critical magnetic field in comparison with the vacuum value for the muonium atom, due to the system of spin interactions of the magnetic moments of the generally delocalized electron, meson, and other nuclei of the molecule, permits investigation of the distribution of electron density in radicals, and experimental determination of the average effective distances between interacting particles possessing magnetic moments. The limiting asymptote of polarization as a function of longitudinal magnetic field strength, which corresponds to the combined rate for reactions with formation of molecules and of radical products, agrees satisfactorily with the values obtained by the method of competing acceptors. It has been shown that the restoration of μ^+ -meson polarization with increase of the field strength is due to the longitudinal component of the magnetic field vector.

Interactions of a μ^+ meson with a crystalline lattice have been studied; these present interest for solution of several problems of solid-state physics.^{52,67-69} The parameters of the phenomenological theory of muonium depolarization of a μ^+ meson in a medium have been determined for the case of the ionic crystal of potassium chloride.⁵² Another means of separate determination of the parameters of the theory, which consists of measurement experimentally of the shift of the phase and amplitude of the residual polarization vector of μ^+ mesons as a function of the external transverse magnetic field strength, has been used to study single crystals of germanium.⁷¹ We note that this technique permits determination of the contribution of the so-called prompt processes ($\tau \ll \omega_0^{-1}$) and is the most precise in measure-

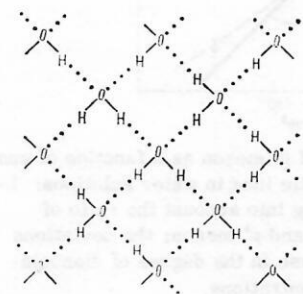


FIG. 9. System of hydrogen bonds in a crystal of ice with tetrahedral coordination around each oxygen atom.

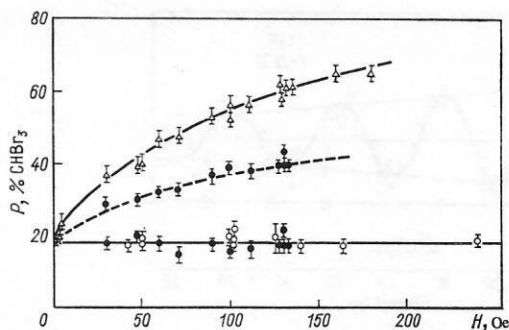


FIG. 10. Polarization in plastic scintillator based on polystyrene as a function of magnetic field strength: \circ —transverse field; Δ —longitudinal field; \bullet —field vector at an angle 45° (135°) to the μ^+ -meson spin (upper and lower points with longitudinal and transverse components of the magnetic field vector, respectively).

ments of the duration of existence of free muonium atoms in a medium.

The phenomenon observed in some cases of a slow (microsecond scale) depolarization at the meson frequency is important in study of spin-lattice relaxation, crystal bond structure, and the role of local magnetic fields. The temperature dependence of the relaxation time of the μ^+ -meson spin is characteristic—with increase of temperature a decrease in the depolarization rate in crystals occurs (Fig. 11). In relatively weak longitudinal magnetic fields (individually for each compound) the slow depolarization effect is removed (Fig. 12). It has been shown that processes leading to slow depolarization of μ^+ mesons in most cases do not exert an important effect on the muonium stage of depolarization. Study of the effect of crystal structure defects has shown that the latter are appreciable but do not change the general nature of the observed phenomena.

Considerable interest is presented by study of the behavior of muonium (the μ^+ meson) in semiconducting materials.^{25,72} Introduction of alloying impurities, and also change of temperature, have revealed a complete spectrum of variation of μ^+ -meson polarization from di-

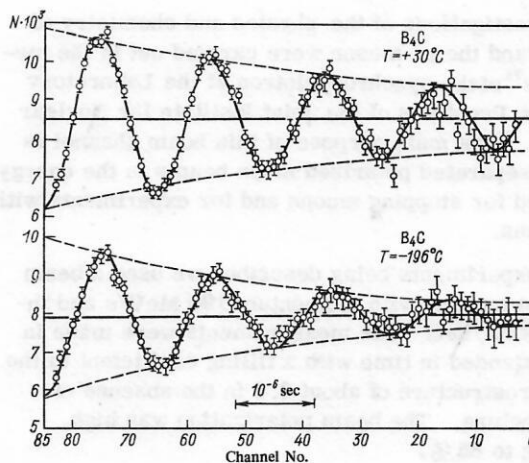


FIG. 11. Spin relaxation of μ^+ meson in boron carbide at various temperatures in a transverse magnetic field ($H = 50, 8$ Oe).

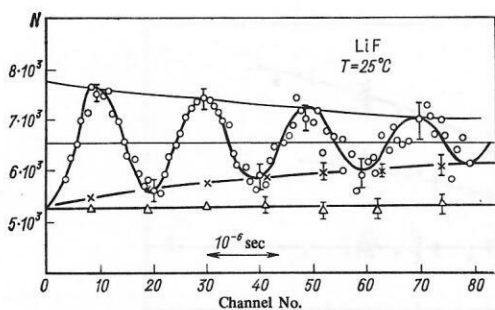


FIG. 12. Change of μ^+ -meson spin relaxation rate in LiF in magnetic fields: \circ —transverse field $H=50$ Oe; \times —longitudinal field $H=0.8$ Oe; Δ — $H=180$ Oe.

electrics to metals. A number of new effects have been observed: dependence of depolarization processes on the type of conduction and the concentration of charge carriers, the nature of the dependence of polarization and spin relaxation rate on temperature, observation of the precession of triplet muonium, and the distortion, mentioned above, of the wave functions of muonium implanted in the crystal lattice of germanium and silicon.

Oxidation and reduction reactions of muonium with anions and cations have been studied, the effect of ionic strength and the degree of dissociation of the electrolytes in concentrated solutions has been investigated, and the fundamentally new fact of dependence of the residual polarization of μ^+ mesons on the concentration of the electrolyte has been experimentally established and theoretically justified in Refs. 60 and 73; the possibility of investigation of the interactions of magnetic moments of μ^+ mesons and a neighboring nucleus in diatomic molecules have been considered in Ref. 58; interaction of magnetic moments of μ^+ mesons and paramagnetic ions in water solutions has been studied and has demonstrated a deep quantitative analogy with the NMR method. The possibility of study of the degree of dissociation of strong electrolytes (Fig. 13) has been investigated in Refs. 59 and 74.

4. PRODUCTION OF HIGH-INTENSITY BEAMS OF POLARIZED MESONS

The investigations of the physics and chemistry of muonium and the μ^+ meson were carried out in the meson beams⁷⁵ of the synchrocyclotron at the Laboratory of Nuclear Problems of the Joint Institute for Nuclear Research. The main purpose of this beam channel is to obtain separated polarized muon beams in the energy range used for stopping muons and for experiments with fast mesons.

In the experiments being described we used a beam of positive mesons with momentum 190 MeV/c and intensity 2×10^5 /sec. The measurements were made in a beam extended in time with a filling coefficient in the time macrostructure of about 0.8 in the absence of microstructure. The beam polarization was high, amounting to 85 %.

The meson channel includes three systems:

- 1) a system for injection into the channel of pions

produced in interaction of a proton beam with target nuclei in the accelerator.

- 2) a strong-focusing array of quadrupole magnets for continuous focusing of muons produced from decay of pions;

- 3) a system for extraction from the channel of particles of a definite momentum and for focusing them on detectors in experimental apparatus.

The channel consists of 31 quadrupole magnets (of type ML-29 and ML-30) and three magnets with gradient focusing (of the type ML-31). The aperture of the channel is 20 cm. The channel length is about 20 m.

The systems of adjustment and transport belonging to the physics installations act on the beams of the meson channel to obtain in each apparatus optimum conditions for carrying out measurements. When necessary the adjustment in the horizontal plane is carried out by remote control. To be able to install a large number of pieces of equipment and effectively use the existing area, the analyzing magnets with two lenses can be rotated by about 90° , providing extraction of beams in two directions.

In the meson channel it is possible to produce more than twenty beams. This substantial muon intensity cannot be achieved in most of the beams from other channels. The meson channel which we are describing is the only one of its type in the Socialist countries. Some other useful features of the meson channel beams make possible their wide use for experiments⁷⁶ in various fields of science and technology. We note some of these features:

- 1) low impurities in the beams;
- 2) possibility of control of the geometrical parameters of the beams on exit from the channel;

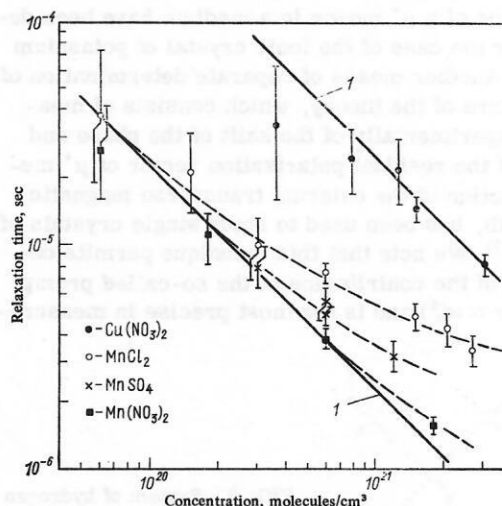


FIG. 13. Spin relaxation time of μ^+ meson as a function of concentration of salts of paramagnetic ions in water solutions: 1—extrapolation of NMR data, taking into account the ratio of magnetic moments of the proton and μ^+ meson; the deviations from the line 1 are due to decrease in the degree of dissociation of the salts at high ion concentrations.

- 3) the possibility of obtaining beams of particles with rather high resolution over a wide range of momenta in a single experimental apparatus;
- 4) the possibility of obtaining particles of both signs;
- 5) longitudinal polarization of the beam, variable in magnitude and direction;
- 6) a significant number of stopped mesons and high density of stoppings;
- 7) smoothing of the time structure for muon beams.

CONCLUSION

It is interesting to note that the discovery of parity nonconservation in weak interactions—phenomena, it would appear, from an extremely remote field, elementary particle physics—after 10–15 years has led to appearance of a new method of studying matter.

The results of the studies carried out show that we can now speak of creation of a new, very promising method of studying the physical and chemical properties of matter. In its essence the mesonic method is an original analog of NMR and EPR. In comparison with these methods the μ^+ -meson method of investigation has certain substantial advantages (and also certain deficiencies), and therefore all of these methods mutually reinforce each other. In particular, for study of the properties of metals the mesonic method is significantly more convenient, since it permits investigation of massive samples, while for both NMR and EPR we are forced to limit ourselves to study of small colloidal particles. Study of the properties of metals (in particular, ferromagnetic materials) has already provided new and interesting results, and very extensive prospects are being opened up in that field.

As is well known, the μ^+ -meson method has permitted investigation of the characteristics of the wave function of a muonium impurity atom in such practically important semiconductors as germanium and silicon, which could not be done for the proton.

The study of matter by means of the μ^+ -meson method, and particularly the study of μ^+ -meson diffusion in copper, has led to discovery of a new phenomenon: sub-barrier diffusion of the μ^+ meson.

We should also emphasize the possibilities of the meson method for investigation of fast chemical reactions of atomic hydrogen; here it is of particular interest to learn the role of the tunneling effect, which should be extremely important for such a light isotope of hydrogen as the muonium atom. On the other hand, we should note that the method is important for study of fast reactions (about 10^{-10} – 10^{-12} sec), which is difficult or impossible to do by other means. This method may also be very useful for study of the places of localization of hydrogen in the lattice of metals.

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