Study of metals by means of positive muons

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This review discusses a number of questions related to the use of the muon method for the study of metals. The possibilities of the method for the study of normal metals, superconductors, and ferromagnetic materials are analyzed. A systematic comparison of theory and experiment is made. It is shown that the method permits determination of the local magnetic fields at crystallographic sites, the type of magnetic structure in complicated ferromagnetic crystals (rare earth metals), analysis of the magnetic texture (grain orientation) of electrical steels, diagnosis of the interstitial sites occupied by a proton or muon, determination of the diffusion rate and the degree of distortion of the crystal lattice, and study of phase transitions in rare earth metals and spin glasses. In normal metals the muon method permits determination both of the muon diffusion rate and of the nature of diffusion (classical or quantum), the Knight shift in the muon, the charge state of a proton or muon in the metal, and the distortion of the crystal lattice. For superconductors the method permits information to be obtained which is inaccessible by other means, particularly determination of the ratio of the normal and superconducting phases and study of the dynamics of destruction and appearance of the superconducting state. The prospects for development of the muon method for study of the properties of metals are demonstrated.

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CONTENTS

1. INTRODUCTION

When Garwin, Lederman, and Weinrich and independently Friedman and Telegdi in 1957 observed the nonconservation of parity in $\pi-\mu-e$ decay,[1,2] hardly anyone supposed that this fundamental fact, which confirmed the nonconservation of parity in weak interactions and which was extremely important for the theory of elementary particles but apparently remote from ordinary down-to-earth physics, would several years later become the basis of a new method of investigation of the properties of matter and in particular would make it possible to obtain new data on solutions of electrolytes.

We mention this particular example since it is hard to think of more remote and at first glance unrelated areas of physics and chemistry than the weak interactions of elementary particles on the one hand and the properties of electrolytes on the other. And, quite aside from the absolute significance of the muon method, from the prospects of its further development, and from its possibilities, it must be acknowledged that it is in such unexpected associations that one finds the "inner beauty" of science, which in the final analysis is what attracts all of those who are occupied with science.

And since, if we believe the classicists, beauty is the most basic and most valuable argument in favor of a physical theory or experiment, the muon method is assured of a brilliant future.

However, returning to prose, we hope to show that the muon method of study of matter has very definite accomplishments at the present time and has been able to recommend itself in a most worthy manner.

A final digression. The entire history of the muon method illustrates once more that there is nothing more practical than a good theory. Indeed, it is difficult to imagine that after only twenty years the work of Lee and Yang on parity nonconservation in weak interactions[3] can already serve, for example, to determine the technical properties of transformer steels (magnetic texture or grain orientation), but as we shall show the muonic method opens substantially new possibilities in this area.

In fact, immediately after the first experiments on verification of parity nonconservation in $\mu - e$ decay, it could be grasped that we had obtained a new instrument for the study of very diverse properties of matter.

The idea of the method is simple. As a result of the nonconservation of parity the angular distribution of the positrons from decay of a stationary polarized muon is asymmetric with respect to the direction of the spin. The muon decay reaction has the single channel $\mu \to e+\nu+\bar{\nu}$. The mean life of a stationary muon is $\tau_\mu = 2.199\times 10^{-6}$ sec, and the momentum distribution of the decay positrons has the form[4-7]

$$dN = \frac{1}{2}\pi x^2 [(3-2x) + \xi(1-2x)\cos\theta]\, dx\, d\Omega; \quad (1.1)$$

here $x = p/p_{\max}$, where p is the positron momentum, and θ is the angle between the positron momentum and the spin of the decaying muon (the polarization of the muon).

The angular distribution has the form

$$I(\theta)\, d\theta \sim \left(1+\xi\frac{1-2x}{3-2x}\cos\theta\right)\sin\theta\, d\theta \equiv (1+A\cos\theta)\sin\theta\, d\theta. \quad (1.2)$$

As can be seen, the asymmetry parameter A depends on the energy of the positron. In the distribution integrated over energy, $A=\xi/3$. In the so-called V–A variant of weak-interaction theory the value of ξ should be -1.

In the experiments one analyzes the polarization of muons produced in decay of pions (here in principle one can obtain 100% polarization of the muons). For positive muons the spin is directed opposite to the direction of the momentum. In a medium the polarization of a muon changes with time, and the angular distribution of the decay positrons integrated over energy has the form

$$dN(\theta, t) = \frac{1}{4\pi\tau_\mu} e^{-t/\tau_\mu}(1+a\,|\mathbf{P}(t)|\cos\theta)\, d\Omega\, dt, \quad (1.3)$$

where $\mathbf{P}(t)$ is the polarization of the muon at the moment of decay. The coefficient a depends on the detecting apparatus. If decay positrons with different energies are detected with the same efficiency, then $a=1/3$. The factor in front of the expression in parentheses takes into account the correction for decay of the muons.

Thus, from the distribution of the positrons we can determine the direction of the muon spin at the moment of decay. In this way a polarized muon in matter constitutes a unique magnetic probe. All of the experimental information is contained in the polarization $\mathbf{P}(t)$ observed in magnetic fields parallel and perpendicular to the initial polarization of the muon. Usually one studies the average polarization $\langle P\rangle$, the precession frequency ω_μ, and the rate of relaxation of the polarization Λ. By measuring these parameters one can obtain a wealth of information regarding a broad spectrum of physical and chemical properties of matter. In many ways this information is close in nature to that obtained by the EPR and NMR methods, but frequently it is quite specific.

It is interesting to note that the arrangement for detection of the decays is such that one always studies only a single muon stopped in the target, and the final data are obtained after collection of statistics on individual decays. This is one of the few cases in physics when it is possible to study the properties of matter by means of a single individual probe particle. A detailed description of the experimental arrangement can be found, for example, in Ref. 8.

In the initial stages the main purpose of the experimental studies was to check the V–A theory of the weak interaction. A polarized beam of muons was slowed down in a target, and then the asymmetry of the decay was studied. Experiments were performed in magnetic fields parallel and perpendicular to the initial polarization of the beam. The degree of polarization of the initial beam of muons, as a rule, was close to 100%.

Since the initial velocity of the muons is close to the velocity of light ($v\sim c$), it is obvious that the time of slowing down of a muon in matter to Bohr velocities ($v\sim\alpha c$) is of the order $\tau\sim l/c\sim 10^{-10}$–$10^{-9}$ sec (l is the characteristic dimension of the target and α is the fine-structure constant). Naturally, this time and this length must not be confused with the average stopping time and range of ionization (for gases, for example, $\langle\tau\rangle\sim 10^{-6}$ sec). This in particular is responsible for the unsuitability of use of gas targets. The number of stoppings is small, and to collect sufficient statistics it is necessary either to "understop" the beam or to use high-intensity beams.

For velocities of the order αc the cross sections for interaction of a muon with the electrons of matter rise rapidly. Since a positive muon, from the point of view of atomic physics, is simply a light isotope of the proton ($9m_\mu \approx m_p$), as a rule at this stage it captures an electron, forming a hydrogen-like atom of muonium (chemical symbol Mu). Then the muonium is slowed down very rapidly to thermal velocities (it is "thermalized"). The thermalization time is estimated to be of the order 10^{-11}–10^{-13} sec.

The hypothesis of formation of the muonium atom was proposed at the very beginning of these studies in order to explain the fact that already after times of the order 10^{-8}–10^{-9} sec the muon has lost a very substantial fraction of its polarization in a large number of cases.[9,10] Simple estimates show the impossibility of a significant depolarization of a bare muon in such short times. These estimates can easily be obtained by recalling that the stochastic magnetic fields in most materials are of the order 1–10 G, while the frequency of the muon spin in a magnetic field is

$$\omega_\mu = g\frac{eB}{2m_\mu c} \approx 8.5\cdot10^4 B\cdot \text{rad/sec} . \quad (1.4)$$

An accomplishment of this period was the work of the group at the Kurchatov Institute, where very accurate measurements were made in strong longitudinal fields, permitting the initial polarization of the muon to be preserved, and the fundamental result $A = 1/3$ was obtained for the asymmetry parameter A, as predicted by the V–A theory of weak interactions.[5,7]

The next step, a very important one for the theory of depolarization, was made by Nosov and Yakovleva.[11] These authors noted for the first time that the Mu atom, which is simply a light isotope of atomic hydrogen, can enter into a chemical reaction with the atoms or molecules of matter (the target), forming a diamagnetic chemical compound.

In the same work a solution was obtained for limiting cases of the equations describing the depolarization of the muon spin in the muonium atom. The complete theory of the muonium mechanism of depolarization of positive muons in matter was developed in a series of papers by Ivanter and Smilga.[12-16]

At the present time the muon method is being developed quite intensively both in the Soviet Union and in other countries. Special prospects are related to the construction of accelerators which will permit very intense muon beams to be obtained (meson factories). Even at the present time the number of publications has exceeded one thousand, and the applications of the method are quite diverse. In the present review we shall confine ourselves to analysis of the possibilites of the muon method for the study of metals. Up to the present time there is no review article in the world's literature where this problem has been discussed systematically and from a single point of view; for this reason the lack of a clear understanding of the physical bases is apparent in some experimental studies. It should be noted that in regard to the study of metals the muon method has at least one important advantage compared to EPR and NMR, in which the size of the sample studied is limited by the thickness of the skin depth and the experiments can be carried out only in colloidal particles or in thin films. For this latter reason, the importance of various surface effects increases rapidly on the one hand, and, on the other hand it is necessary to perform an averaging over the size of the particles. All of these factors hinder the interpretation of the data. By means of muons one can study massive samples and, furthermore, prior to the introduction of high-current accelerators (meson factories) only massive samples could be studied. At the present time the thickness of the target for the condensed phases can be reduced to several microns.

Up to the present time there is no unique answer to the question of whether there exists a proton + electron bound state in metals or, what amounts to the same thing, whether the Mu atom exists in metals. Theoretical estimates are necessarily based on crude models and frequently lead to contradictory conclusions.[17-26] The existing experimental data on the behavior of hydrogen in metals do not permit a unique interpretation. The only direct experiment[27,28] on the search for the muonium atom in Zn, Al, and Cu unfortunately also cannot be interpreted uniquely. As will be shown in detail below, the results can be explained either by assuming that the Mu atom in a metal is very highly "inflated" (or not formed at all), or by the fact that the Kondo temperature[1)] for Mu in the metals studied is rather high ($T_K \sim$ 200–300 K). The second possibility also leads to uncertainty of the result. The situation is complicated by the fact that reliable calculation of the Kondo temperature from first principles is difficult, and the experimental data for various paramagnetic centers in metals show that T_K can vary[56] over the range 0.1–600 K. Thus, the question is open. It is not excluded, for example, that the Mu atom is formed in some metals and is not formed in others.

The entire problem has not only a purely academic nature, but also great practical value, since the behavior of hydrogen dissolved in a metal is of interest in a wide range of problems of physics and the physics and chemistry of metals.

Observation of the Mu atom in metals is substantially more complicated than in nonconducting materials, since the spin-spin coupling of the electron and muon is highly attenuated as the result of strong exchange scattering of the muonium electron by the electrons of the medium—the thermal reservoir.[29] Therefore, regardless of whether the muonium atom is formed or not, the muon behaves in its general features as if it were a free particle: its spin precesses with a frequency ω_μ, and the depolarization is determined by the direct interaction of the magnetic moment of the muon with the magnetic moments of the nuclei. However, if muonium is formed, the last two statements are only approximately true, and in the following we shall indicate a number of procedures permitting unique determination of whether or not the Mu atom exists in a metal.

Thus, in what follows we shall have in mind two alternative variants: 1) in some interstitial site of the crystal lattice there is a free muon; 2) the Mu atom is formed.

If muonium is not formed, then we can write down simple formal relations which are common for all metals. The polarization of an ensemble of muons $\mathbf{P}(t)$ can be represented as the convolution of the initial polarization vector $\mathbf{P}(0)$ and a tensor of second rank:

$$P_\alpha(t) = \mu_{\alpha\beta}(t)\, P_\beta(0). \quad (1.5)$$

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1) The temperature T_K below which the magnetization of the cloud of conduction electrons compensates the magnetization of the impurity if the exchange interaction is antiferromagnetic.

The tensor $\mu_{\alpha\beta}(t)$ characterizes the magnetic properties of the target-sample in an external field.

In the absence of diffusion, if the microscopic field at the point of location of the muon can be considered constant during the muon lifetime τ_μ, this tensor is the result of averaging of the matrix

$$M_{\alpha\beta}(\mathbf{b}, t) = n_\alpha n_\beta + (\delta_{\alpha\beta} - n_\alpha n_\beta)\cos(\gamma_\mu bt) + e_{\alpha\beta\delta} n_\delta \sin(\gamma_\mu bt), \quad (1.6)$$

which determines the change of the polarization vector in a magnetic field which is constant in time:

$$\mu_{\alpha\beta}(t) = \int M_{\alpha\beta}(\mathbf{b}, t) W(\mathbf{b})\, d\mathbf{b} = \langle M_{\alpha\beta}(t)\rangle; \quad (1.7)$$

here $n_\alpha = b_\alpha/b$, γ_μ is the gyromagnetic ratio for the muon, and $W(\mathbf{b})$ is the probability density of the muon's finding itself in a microscopic field $\mathbf{b}$.

The microscopic field $\mathbf{b}$ is made up of the fields of all dipoles of the sample, the external magnetic field, and the contact field of the conduction electrons of the metal. It is convenient to present $\mathbf{b}$ in the form

$$\mathbf{b} = \mathbf{B}' + \mathbf{b}_r + \mathbf{B}_{cont}, \quad (1.8)$$

where $\mathbf{B}'$ is the average (macroscopic) field produced by all dipoles lying outside some small but macroscopic cavity r constructed around the point of location of the muon, and by the external sources; $\mathbf{b}_r$ is the microscopic field of all dipoles of the cavity r. The size of the cavity r is chosen much smaller than the characteristic length in which the macroscopic field $\mathbf{B}'$ changes. For paramagnetic materials this length can be due to the nonuniformity of the external field, for superconductors of the second type it is determined by the diameter of the superconducting filaments, and for ferromagnetic materials it is determined by the size of the domain or crystallite. The sum $\mathbf{B}' + \mathbf{b}_r$ obviously does not depend on the shape of the cavity r.[2)] For calculation of the microscopic field $\mathbf{b}_r$ it is best to use the well known method of Ewald. In practice it is convenient to choose the cavity r as a sphere, and in this case $\mathbf{B}' = \mathbf{B} - (8\pi\mathbf{M}/3)$, where $\mathbf{M}$ and $\mathbf{B}$ are the macroscopic magnetization and magnetic induction in the region of the cavity. The contact field $\mathbf{B}$ oscillates with a frequency 10^{15}–10^{16} sec^{-1}, and for processes with characteristic times $\tau = 10^{-9}$–10^{-5} sec its average value, which is proportional to the average spin density of the point x, is important.

Let us see now what kind of information can be obtained by analyzing $P_\alpha(t)$. Let us divide $P_\alpha(t)$ into the constant and oscillating parts of the polarization $P_\alpha^c(t)$ and $P_\alpha^v(t)$:

$$P_\alpha(t) = P_\alpha^c + P_\alpha^v(t) \equiv [\mu_{\alpha\beta}^c + \mu_{\alpha\beta}^v(t)]\, P_\beta(0), \quad (1.9)$$

where

$$\mu_{\alpha\beta}^c = \langle n_\alpha n_\beta\rangle,$$
$$\mu_{\alpha\beta}^v(t) = \langle(\delta_{\alpha\beta} - n_\alpha n_\beta)\cos(\gamma_\mu bt)\rangle + e_{\alpha\beta\delta}\langle n_\delta \sin(\gamma_\mu bt)\rangle. \quad (1.10)$$

Experimentally P_α^c and P_α^v are easily observed individually: P_α^c, for example, can be found by taking the average polarization value over a time interval much greater than the period of its oscillations. Since the tensor $\mu_{\alpha\beta}(t)$ has nine independent components in the general case, to determine it one must measure projections of the vector $\mathbf{P}(t)$ onto three noncoplanar directions for three independent values of $\mathbf{P}(0)$. Usually one can restrict oneself to a smaller number of elements, since as a rule in magnetic respects the target possesses symmetry axes.

As follows directly from Eq. (1.10), the tensor $\mu_{\alpha\beta}^c$, which we shall call the orientation tensor, is determined by the constant part P_α^c. The real part of the Fourier transform of $\mu_{\alpha\beta}^v(t)$ in the argument $\gamma_\mu t$ is the result of averaging over angle the tensor $\delta_{\alpha\beta} - n_\alpha n_\beta$.

$$\mathrm{Re}\,\mu_{\alpha\beta}^v(b) = \int (\delta_{\alpha\beta} - n_\alpha n_\beta)\, W(\mathbf{b})\, b^2\, d\mathbf{n}. \quad (1.11)$$

The probability density of the field distribution in modulus $W_1(b) = \int W(\mathbf{b}) b^2 d\mathbf{n}$, as follows from Eq. (1.11), is determined by the convolution of the symmetric tensor:

$$2W_1(b) = \mathrm{Re}\,\mu_{\alpha\alpha}^v(b). \quad (1.12)$$

From the imaginary part of the Fourier transform of $\mu_{\alpha\beta}^v(t)$ one can find the average direction of the field with modulus b:

$$2\int n_\gamma W(\mathbf{b})\, b^2\, d\mathbf{n} = e_{\gamma\alpha\beta}\, \mathrm{Im}\,\mu_{\alpha\beta}(b). \quad (1.13)$$

We now turn to discussion of concrete metals.

2. NORMAL METALS

Recently the muon method of studying normal metals has been developed at a particularly rapid rate. Nevertheless we are at present in the initial stage of systematic studies and the most interesting results are still ahead.

a) The "muon" depolarization variant

Let us consider the regularities of the behavior of the polarization $\mathbf{P}(t)$ of the muon, assuming that the Mu atom is not formed. The most interesting information can be obtained by analyzing the rate of depolarization. The relaxation is determined only by the direct dipole-dipole interactions of the magnetic moments of the muon and the nuclei of the metal. In fact, the rate of depolarization Λ in the conduction electrons can be estimated from the well-known Korringa relation (see for example Ref. 30): $\Lambda \approx 4\pi\hbar^{-1}(\gamma_\mu/\gamma_e)^2 K^2 k_B T$, where K is the Knight-shift parameter in the muon (the shift of the muon resonance frequency in an external magnetic field due to the average contact field, which is produced by conduction electrons at the point of location of the muon), k_B is the Boltzmann constant, and T is the absolute temperature. Since the relaxation rate is proportional to the density of electrons taking part in the scattering, it is evident that Λ is proportional to the temperature of the medium. For most metals which have been investigated $K \lesssim 10^{-4}$ and even for $T \sim 10^3$ K we obtain $\Lambda \sim 10^2$ sec^{-1}. Therefore the Korringa relaxation can be neglected. The Knight shift in the muon has been studied experimentally by several authors. A summary of the data on the Knight

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2) We note that this statement and Eq. (1.8) are both rigorously proved for crystals by means of the Ewald method, which we shall discuss in more detail below.

shift in the muon as of 1976 can be found in the review by Schenck.[31]

The muon-spin relaxation rate depends substantially on whether the muon is diffusing or whether it is localized at interstitial sites. Therefore study of the muon depolarization opens up prospects for measurement of its diffusion rate.

Suppose that there are N different types of equilibrium positions for muons in the sample. These positions may be due to capture of muons in crystallographically different interstitial sites, in impurity atoms, in lattice defects, and so forth. The polariization of muons localized in positions of type (i) is determined by the evolution operator (matrix) $\mathbf{G}_i^0(t)$, so that in the absence of diffusion we have for the observed polarization

$$\mathbf{P}(t)=\sum_{i=1}^{N} W_i \mathbf{G}_i^0(t)\,\mathbf{P}(0), \tag{2.1}$$

where W_i is the probability of capture of muons in a position of type (i) directly after thermalization. Obviously the operator $\mathbf{G}_i^0(t)$ is the result of averaging of the matrix $M_{\alpha\beta}(t)$, Eq. (1.6), over interstitial sites of type (i). Diffusion processes can be described phenomenologically by means of probabilities λ_{ik} of transfer of a muon from a position of type (k) to a position of type (i) per unit time. Let $P_i(t)$ be the contribution to the polarization from muons which at a moment of time t are at locations of type (i); obviously $\mathbf{P}(t)=\sum_{i=1}^{N}\mathbf{P}_i(t)$ and $\mathbf{P}_i(0)=W_i\mathbf{P}(0)$. The partial polarizations $\mathbf{P}_i(t)$ satisfy the system of integral equations[32]:

$$\mathbf{P}_i(t)=e^{-\lambda_i t}\mathbf{G}_i^0(t)\,W_i\mathbf{P}(0)+\sum_{k=1}^{N}\lambda_{ik}\int_0^t e^{-\lambda_i(t-\tau)}\mathbf{G}_i^0(t-\tau)\,\mathbf{P}_k(\tau)\,d\tau, \tag{2.2}$$

where $\lambda_i=\sum_{k=1}^{N}\lambda_{ki}$ is the total probability of a jump of the muon from a type (i) site per unit time. A Laplace transformation reduces the system of equations (2.2) to a system of linear algebraic equations with constant coefficients, from which it is easy to find the Laplace transform of the observed polarization. Thus, the system (2.2) solves the problem of the behavior of the polarization of diffusing muons if the operators $\mathbf{G}_i^0(t)$ are known.

b) Depolarization of nondiffusing muons

Let us first consider nondiffusing muons. The Hamiltonian of the system has the form

$$H=H_\mu+H_I+H_{\mu I}+H_{II}; \tag{2.3}$$

here H_μ is the interaction of the muon with the external magnetic field, H_I is the interaction of the spins of the nuclei with the external fields, $H_{\mu I}$ is the spin-spin interaction of the muon with the nuclei, and H_{II} is the interaction between the spins of the various nuclei. Simple estimates show that the action of the operator H_{II} in times $t\lesssim 10^{-4}$ sec can be neglected.

We shall write the Hamiltonian H in the form

$$H=H_0+V, \tag{2.4}$$

where H_0 is the basic Hamiltonian and V is a perturbation; the operator V includes the term $H_{\mu I}$. The specific form of breakdown of the Hamiltonian (2.4) depends on the magnitude of the external field. As is well known,[33] the main contribution to depolarization is from the part of the perturbing operator V which is diagonal in the H_0 representation (the so-called secular part of the interaction). The meaning of separation of the secular interactions can easily be understood in classical language: for a rapid precession of the muon and the nuclei the variable components of the local field at the muon are averaged and the main action is exerted by the static components. We note that in this approximation only the polarization component perpendicular to the external field relaxes.

In what follows we shall assume that the external magnetic field $\mathbf{B}$ is directed along the z axis and the initial polarization of the muon is along the x axis. Then $H_\mu=-\hbar\omega_\mu S_z$, where $\omega_\mu=\gamma_\mu B$; $H_I=-\hbar\omega_I\sum_j I_{jz}+H_Q$; here $\omega_I=\gamma_{IB}$, and HQ is the quadrupole interaction of the nuclei with the electric field gradient produced by the muon. The importance of this interaction for nuclei with spin $I>1/2$ has been pointed out by Hartmann.[34] It leads to the result that for the nuclei closest to the muon the direction of the quantization axis does not coincide with the z axis and depends on the magnitude of the applied field $\mathbf{B}$.

If the external field is large $(H_\mu, H_I \gg H_{\mu I})$, then $H_0=H_\mu+H_I$, $V=H_{\mu I}$, and the secular part of the interaction has the form

$$V^s=\hbar^2\gamma_\mu\gamma_I\sum_j r_j^{-3}\,[i_{jz}-3a_{jz}(\mathbf{a}_j\mathbf{i}_j)]\,S_z(\mathbf{I}_j\mathbf{i}_j), \tag{2.5}$$

where $\mathbf{a}_j=\mathbf{r}_j/r_j$ is the unit vector directed from the muon to the j-th nucleus and $\mathbf{i}_j$ is a unit vector directed along the quantization axis of the j-th nucleus. The complex muon polarization $P_+(t)=P_x(t)+iP_y(t)$ is determined by the obvious formula

$$P_+(t)=\mathrm{Sp}\,[\sigma_+(t)\,\rho_\mu(0)\,\rho_I(0)], \tag{2.6}$$

where $\sigma_+(t)=\exp[i\hbar^{-1}(H_0+V^s)t]\sigma_+\exp[-i\hbar^{-1}(H_0+V^s)t]$, $\rho_\mu(0)=1/2+S_xP(0)$, and $\rho_I(0)$ is the Gibbs density matrix of the nuclear spin system. The nuclei are practically always unpolarized. Calculations[32] give

$$P_+(t)=G^0(t)\,P(0)=e^{-i\omega_\mu t}\prod_j\frac{\sin\{[I+1/2]\,\Omega_j t\}}{(2I+1)\sin(\Omega_j t/2)}P_+(0), \tag{2.7}$$

where

$$\Omega_j=\hbar\gamma_\mu\gamma_I r_j^{-3}\,[i_{jz}-3a_{jz}(\mathbf{a}_j\mathbf{i}_j)]. \tag{2.8}$$

In the absence of quadrupole interactions we have $\Omega_j=\hbar\gamma_\mu\gamma_I r_j^{-3}[1-3(a_{jz})^2]$ and Eq. (2.7) goes over into the formula obtained by Selivanov.[35]

As can be seen, the polarization vanishes an infinite number of times, the first zero being reached at $t\approx\pi/[I+(1/2)]\Omega_j^{\max}$. The magnitude of the next maximum is ~1% of $P(0)$.

For an approximate description one often uses the expression $P_+(t)=\exp(-i\omega_\mu t-\sigma^2t^2)P_+(0)$, where σ^2 is defined as the coefficient of t^2 in the expansion of the polarization amplitude in powers of time. From Eq. (2.7) we find

$$\sigma^2=\frac{1}{6}I(I+1)\sum_j\Omega_j^2. \tag{2.9}$$

The frequencies Ω_j depend strongly on the directions of the vectors $\mathbf{i}_j$, which are determined by the relation between the external magnetic field and the electric field gradient. Hartmann[34] gives a relation $\sigma^2(B)$ for different orientations of **B** with respect to the axes of a monocrystalline sample and for various positions of the muon at interstitial sites in the lattice. Comparison with the results of Camani *et al.*[36] shows that in copper the muons are captured in octahedral sites. To achieve quantitative agreement with the theoretical dependence, the authors had to assume that the muon expands the lattice by ~5%.

If the quadrupole interaction is unimportant (the external magnetic field is very large), then σ^2 is determined by the well known van Vleck formula

$$\sigma^2=\frac{1}{6}I(I+1)(\hbar\gamma_\mu\gamma_I)^2\sum_j\frac{[3(a_{jz})^2-1]^2}{r_j^6}. \qquad (2.10)$$

We give a dependence of σ^2 on the orientation of a monocrystalline sample with cubic symmetry of the lattice with respect to the external field:

$$\sigma^2=\frac{1}{6}I(I+1)(\hbar\gamma_\mu\gamma_I)^2[A_0+A_1(n_1^4+n_2^4+n_3^4)], \qquad (2.11)$$

where n_1, n_2, and n_3 are the direction cosines of the magnetic field with respect to the crystallographic axes, $A_0=(1/2)[7\sum_j r_j^{-6}-27\sum_j r_j^{-6}\cos^4\alpha_j]$, $A_1=(9/2)[-\sum_j r_j^{-6}+5\sum_j r_j^{-6}\cos^4\alpha_j]$, and α_j is the angle between the [001] axis and the direction from the muon to the j-th nucleus. For polycrystalline samples

$$\sigma^2=\frac{2}{15}I(I+1)(\hbar\gamma_\mu\gamma_I)^2\sum_j r_j^{-6}. \qquad (2.12)$$

Let us consider now the case of an intermediate magnetic field where $\omega_\mu\gg\omega_{\mu I}$, $\omega_I\sim\omega_{\mu I}$; here $\omega_{\mu I}$ is the field created by the nuclei at the muon (in frequency units). Then $H_0=H_\mu$, $V=H_{\mu I}+H_I$, and instead of Eq. (2.5) for V^s we have

$$V^s=H_I+\hbar^2\gamma_\mu\gamma_I\sum_j r_j^{-3}[I_{jz}-3a_{jz}(\mathbf{a}_j\mathbf{I}_j)]S_z. \qquad (2.13)$$

A calculation again leads to Eq. (2.7), but Ω_j now has the form

$$\Omega_j=\hbar\gamma_\mu\gamma_I r_j^{-3}\sqrt{1+3(a_{jz})^2}. \qquad (2.14)$$

However, if there is a quadrupole interaction, then for nuclei remote from the muon it is necessary in the product (2.7) to take Ω_j from Eq. (2.14), and for near nuclei they must be taken from Eq. (2.8), in which $\mathbf{i}_j=\mathbf{a}_j$: $\Omega_j^{\text{near}}=-2\hbar\gamma_\mu\gamma_I r_j^{-3}a_{jz}$. For σ^2 we obtain

$$\sigma^2=\frac{1}{6}I(I+1)(\hbar\gamma_\mu\gamma_I)^2\Big[\sum_{\substack{\text{all}\\ \text{nuclei}}}\frac{1+3(a_{jz})^2}{r_j^6}-\sum_{\substack{\text{near}\\ \text{nuclei}}}\frac{1-(a_{jz})^2}{r_j^6}\Big]. \qquad (2.15)$$

The first term in Eq. (2.15) does not take quadrupole interactions into account; it was obtained in Ref. 37. However, the second term determines the contribution of quadrupole interactions. We note that for single crystals with cubic symmetry of the interstitial site and for polycrystalline samples σ^2 has the form[32]

$$\sigma^2=\frac{1}{3}I(I+1)\Big[\sum_{\substack{\text{all}\\ \text{nuclei}}}r_j^{-6}-\frac{1}{3}\sum_{\substack{\text{near}\\ \text{nuclei}}}r_j^{-6}\Big] \qquad (2.16)$$

and does not depend on the orientation of the external field. For zero external field σ^2 is a factor of two larger than for an "intermediate" field.

Inclusion of quadrupole interactions leads to the result that the calculated value of σ^2 decreases by about 30%. For example, for copper we obtain $\sigma\approx 0.34$ μsec^{-1} if the muon is localized in a tetrahedral site and $\sigma\approx 0.27$ μsec^{-1} if it is localized in an octahedral site. The latter value is in good agreement with the experimental data of Grebinnik *et al.*,[38] who obtained the value $\sigma=0.252\pm0.007$ μsec^{-1}. Exact agreement can be obtained if we assume that the interstitial site is expanded by ~3%.

c) Diffusion and "strange" diffusion of muons

Let us turn now to discussion of diffusing muons, following the work of Mikaélyan and Smilga[32]; here we first restrict ourselves to the case in which muons are captured in equilibrium positions of only a single type. This case is realized, apparently, in copper. Then from Eq. (2.2) we have for the Laplace transform of the polarization:

$$\tilde{\mathbf{P}}(p)=\tilde{\mathbf{G}}^0(p+\lambda)[1-\lambda\tilde{\mathbf{G}}^0(p+\lambda)]^{-1}\mathbf{P}(0), \qquad (2.17)$$

where $\lambda=\lambda_1=\lambda_{11}$ is the frequency of jumps of the muon and

$$\tilde{\mathbf{P}}(p)=\int_0^\infty\exp(-pt)\mathbf{P}(t)\,dt,\qquad \tilde{\mathbf{G}}^0(p+\lambda)=\int_0^\infty\exp[-(p+\lambda)t]\mathbf{G}^0(t)\,dt.$$

If λ is small in comparison with the eigenfrequencies ω_μ and ω_I of the Hamiltonian H_0, then the high-frequency part of the field is averaged over the time spent by the muon, and instead of the exact operator $\mathbf{G}^0$ one can use the function G^0 defined by Eq. (2.7). From Eq. (2.17) we obtain

$$P_z(t)=P_z(0), \qquad (2.18)$$

$$P_+(t)=\frac{1}{2\pi i}\int_{-i\infty+\varepsilon}^{i\infty+\varepsilon}\frac{\tilde{G}^0(p+\lambda)}{1-\lambda\tilde{G}^0(p+\lambda)}e^{pt}\,dp. \qquad (2.19)$$

At the present time one usually uses the well known semiempirical formula[33]

$$P_+(t)=e^{-i\omega t}\exp\Big[-2\frac{\sigma^2}{\lambda^2}(e^{-\lambda t}+\lambda t-1)\Big]P(0) \qquad (2.20)$$

for description of experimental data.

For $\lambda\to 0$ it follows from this that $G^0(t)\approx\exp(-i\omega_\mu t-\sigma^2t^2)$. If this expression is used in Eq. (2.19), then for the two limiting cases $\lambda\ll\sigma$ and $\lambda\gg\sigma$ Eqs. (2.19) and (2.20) coincide, while in the intermediate region $\lambda\sim\sigma$, as a calculation shows, the differences are insignificant. Thus, Eq. (2.20) gives a qualitatively correct result.

For rapid diffusion ($\lambda\gg\omega_I$) in calculation of $\mathbf{P}(t)$ it is necessary to use Eq. (2.17) with the exact operator $\tilde{\mathbf{G}}^0(p+\lambda)$. The calculation is greatly simplified, however, as the result of the presence of the small parameter $\omega_{\mu I}/\lambda$ (we recall that $\omega_{\mu I}$ is the field created by the nuclei at the muon). We give only the final formulas:

$$P_z(t)=e^{-\Lambda_1 t}P_z(0),$$
$$P_+(t)=e^{-(i\omega+\Lambda_2)t}P_+(0). \qquad (2.21)$$

Here

$$\Lambda_1=\frac{2}{3}\frac{\lambda\langle(\omega_{\mu I})^2\rangle}{\omega_\mu^2+\lambda^2},\quad \Lambda_2=\frac{1}{3}\frac{\langle(\omega_{\mu I})^2\rangle}{\lambda}\frac{\omega_\mu^2+2\lambda^2}{\omega_\mu^2+\lambda^2},$$
$$\omega=\omega_\mu\Big(1+\frac{1}{3}\frac{\langle(\omega_{\mu I})^2\rangle}{\omega_\mu^2+\lambda^2}\Big)\quad \langle(\omega_{\mu I})^2\rangle=2I(I+1)(\hbar\gamma_\mu\gamma_I)^2\sum_j r_j^{-6}. \qquad (2.22)$$

It follows from this that for rapid diffusion the precession frequency of the muons differs from ω_μ, the correction reaching several tenths of a percent and decreasing rapidly with increase of the external magnetic field and temperature. For example, in copper in a field $B = 62$ G for $\lambda = \omega_\mu$ it is ~0.38%. Grebinnik *et al.*[27,28] carried out a search for the Mu atom in copper on the basis of the temperature dependence of the precession frequency; here a small decrease of ω with increase of T was observed in a field of 62 G and was not observed at a field of 700 G. In principle this effect can be explained by means of the first of Eqs. (2.22).

As follows from Eqs. (2.21) and (2.22), the longitudinal component of the polarization (parallel to the field) is damped at a rate Λ_1 which reaches a maximum for $\lambda = \omega_\mu$ and approaches zero both for $\lambda \ll \omega_\mu$ and for $\lambda \gg \omega_\mu$. In copper in a field of 62 G the maximum value of Λ_1 is ~0.04 μsec^{-1}.

The relaxation rate Λ_2 of the transverse component differs from the value given by Eq. (2.20). In fact, for $\lambda \gg \sigma$ we find from (2.20) $\Lambda_2 = 2\sigma^2/\lambda$, where σ^2 is given by Eq. (2.9). In the case of a large external field this result is valid for $\lambda \ll \omega_I$. For $\omega_I \ll \lambda \ll \omega_\mu$ and for $\lambda \gg \omega_\mu$ we have respectively $\Lambda_2 = \langle(\omega_{\mu I})^2\rangle/3\lambda$ and $\Lambda_2 = 2\langle(\omega_{\mu I})^2\rangle/3\lambda$. Let us consider the fact that the quantity $\langle(\omega_{\mu I})^2\rangle$ does not depend on the orientation of the external magnetic field with respect to the axes of the single crystal.

The equations (2.22) can be obtained also from the well known Wangsness-Bloch equation. In Refs. 39 and 40 the muon polarization is calculated by means of a somewhat modified Wangsness-Bloch equation (see for example Ref. 41). This equation is used for large correlation times, i.e., over the entire range of diffusion velocities. In Ref. 39 the correlator of the local field at the muon $\langle\{h_\alpha(t), h_\beta(0)\}\rangle/2$, knowledge of which is necessary in this approach, is calculated quantum-mechanically on the basis of the random-walk method. For a comparison of the results with experiment, a computer analysis is necessary. On the other hand, in Ref. 40 it is assumed that the components of the local field at the muon are statistically independent and the correlator has the form $\langle h_\alpha(t) h_\beta(0)\rangle = \langle h^2(0)\rangle \delta_{\alpha\beta} f(t)$. We note that this assumption is valid only for not too strong external fields ($B \lesssim 100$ G), where the nuclei precess slowly, or for very rapid diffusion. In the opposite case it is necessary to take this precession into account in the correlator, and the formulas obtained in that work become incorrect.

The formulas presented above show that with increase of the frequency of jumps λ the relaxation rate of the polarization component perpendicular to the external field falls off monotonically, approaching zero. However, recently an unusual nonmonotonic dependence of $\Lambda(T)$ on temperature has been observed in a considerable number of metals.[42-46] In niobium[43,44] the relaxation rate first falls off and reaches a minimum for $T \approx 25$ K, then rises and reaches a plateau, and subsequently drops again. A similar behavior of $\Lambda(T)$ has been observed also in bismuth.[42] In tantalum and beryllium[45] the picture is somewhat different: in tantalum the relaxation rate is low at low temperatures, then rises, after which it falls off again. In beryllium two plateaus are observed for $\Lambda(T)$. In Ref. 46 the depolarization of muons in aluminum was investigated and it was established that Λ is very small even at low temperatures. At the same time in the alloy Al + 1% Cu the dependence $\Lambda(T)$ is qualitatively the same as in pure copper.

This behavior of $\Lambda(T)$ can be explained if we assume that in the crystal there are at least two types of equilibrium positions of muons with different binding energies. We shall not discuss here the possibilities associated with coherent quantum diffusion, since it is obvious that it can appear only in specially prepared pure single crystals.

For definiteness we shall assume that the positions of type (2) are more stable, i.e., $\lambda_{21} \gg \lambda_{12}$. As was mentioned above, in the first approximation we can assume that the amplitude of the polarization component perpendicular to the external field for muons localized in sites of types (1) and (2) falls off according to laws $\exp(-\sigma_1^2 t^2)$ and $\exp(-\sigma_2^2 t^2)$, respectively. We shall consider the region of temperatures for which $\lambda_{11} \gg \sigma_1$ and $\lambda_{22} \approx \lambda_{12} \approx 0$. From the system of equations (2.2) we can obtain[156]

$$P(t) = \Big\{W_1 \exp[-(2\sigma_1^2/\lambda_{11} + \lambda_{21})t] + W_2 \exp(-\sigma_2^2 t^2) + W_1 \lambda_{21} \int_0^t \exp\Big[-\sigma_2^2 (t-\tau)^2 - \Big(\frac{2\sigma_1^2}{\lambda_{11}} + \lambda_{21}\Big)\tau\Big] d\tau\Big\} P(0). \quad (2.23)$$

If the sites (2) are due to capture into impurity atoms, then $W_2 \approx 0$ and $\lambda_{21}/\lambda_{11} \approx c$, where c is the relative concentration of the impurities. Then for $\lambda_{21} \ll \sigma_2$ in Eq. (2.23) the main contribution is from the first term and we find for the relaxation rate

$$\Lambda \approx \frac{2\sigma_1^2}{\lambda_{11}} + \lambda_{21} \approx \frac{2\sigma_1^2}{\lambda_{11}} + c\lambda_{11}. \quad (2.24)$$

From this it follows that for $\lambda_{11} \approx \sigma_1 \sqrt{2/c}$ the relaxation rate is minimal, and $\Lambda_{min} \approx 2\sigma_1\sqrt{2c}$. Thus, on decrease of the impurity concentration the location of the minimum of Λ should be shifted toward higher temperatures and $\Lambda_{min} \ll \sigma_1$. These conclusions are inconsistent with the results obtained in niobium and bismuth.[42-44] In fact, in these experiments samples with $c \sim 10^{-4}$ were studied and it was found that $\Lambda_{min}/\sigma_1 \approx 0.35$–$0.6$, while the calculated value is ~0.03. Therefore the sites (1) and (2) in this case are evidently interstitial sites of different types. For a final clarification of the question more detailed studies are necessary, for example, measurement of the dependence of Λ on the concentration of the impurities.

d) Behavior of muon polarization on formation of a muonium atom

In this section we shall discuss the principal regularities of muon polarization behavior if a Mu atom is formed in a normal metal. The manifestation of the qualitative regularities of the muon depolarization process in this case has a fundamental nature, since as we have already remarked above the question of the charge state of a positive muon in metals has not been unambiguously solved at the present time.

The Mu atom in polarizable media (metals and semiconductors) has been discussed theoretically by several authors.[47-49] In Refs. 47 and 48 some regularities of the muon polarization behavior were studied. In Ref. 48 it was correctly noted that in a polarizable medium the magnetic moment of the muonium electron is renormalized, which was not taken into account in Ref. 47. In other respects the initial equations, which were obtained in Ref. 47 from phenomenological considerations and in Ref. 48 on the basis of an analysis of exchange scattering in a polarizable gas,[50] are practically identical. However, the basic equations are, generally speaking, erroneous, since they do not take into account the inverse influence of the magnetic moment of the nucleus (muon) on the electron spin. As a result, for example, the muonium equilibrium state obtained in these studies does not satisfy the Gibbs distribution [see Eq. (15) of Ref. 47]. In Ref. 49 a systematic theory of the relaxation process was developed. We shall present briefly the principal results.

We write the Hamiltonian of the system in the form

$$H = H_0 + H_T + V, \tag{2.25}$$

where H_0 is the Hamiltonian of the dynamical subsystem (muonium), H_T is the Hamiltonian of the thermal reservoir, and V is the potential for interaction of the dynamical subsystem with the thermal reservoir, the part of the interaction potential diagonal in the thermal reservoir being equal to zero or assigned to H_0.

The explicit form of the interaction potential is determined by the model of the thermal reservoir. In metals the relaxation of the muonium electron spin is due mainly to exchange scattering by the electrons of the medium. As usual,[51-54] the model Hamiltonian used was

$$V(r) = \frac{J}{n}\sum_t \sigma_e\sigma_t\delta(\mathbf{r}_e - \mathbf{r}_t), \tag{2.26}$$

where σ_e and σ_t are the Pauli operators of the electrons of the muonium and the medium, respectively; J is the exchange integral and n is the density of electrons.

As a basis we took the well known relaxation equations of NMR and EPR theory (see for example Refs. 41 and 55). In the calculation it was assumed that the wave functions of the conduction electrons of the metal are plane waves and we neglected their distortion near the muonium. Accordingly we did not consider effects related to the Kondo effect, i.e., it was assumed that the temperature $T > T_K$—the Kondo temperature. It was assumed also that the wave function of the muonium is the s-function of a hydrogen-like atom. As a result we obtained for the muonium spin density matrix the equation

$$\frac{\partial\rho}{\partial t} + i\hbar^{-1}[H_{\text{eff}}, \rho] = \frac{1}{2}[\sigma^+, \rho\tilde{G}] + \frac{1}{2}[\sigma^-, \rho G] + [\sigma^z, \rho G_z] + \text{H.c.}, \tag{2.27}$$

where H_{eff} is the Hamiltonian H_0 with renormalized magnetic moments of the muonium electron μ_e and of the muon μ_μ and a frequency of hyperfine interaction in the metal ω_0; G is a matrix with elements $g(\omega_{ln})\sigma^+_{ln}$, $\tilde{G}$ is a matrix with elements $\sigma^-_{ln}g(\omega_{nl})$, G_z is a matrix with elements $g(\omega_{ln})\sigma^z_{ln}$,

$$g(\omega_{ln}) = \nu\frac{\hbar\omega_{ln}/2T}{\operatorname{sh}(\hbar\omega_{ln}/2T)}\exp\frac{\hbar\omega_{ln}}{2T}, \quad \nu = \frac{3\pi}{8}\left(\frac{J}{2\varepsilon_f}\right)^2\frac{T}{\hbar}, \tag{2.28}$$

$\hbar\omega_{ln} = E_l - E_n$, where E_n are the eigenvalues of H_{eff} and ε_f is the Fermi energy.

The coefficients $g(\omega_{ln})$ determine the frequencies of transitions between different levels and can be interpreted as effective collision frequencies. We note that for $\hbar\omega_{ln} \gg T$ the coefficients increase linearly with increase of ω_{ln}. This corresponds to the fact that electrons with energy $\varepsilon \geq \varepsilon_f - \hbar\omega_{ln}$ can also take part in the scattering.

In the general case the system of equations (2.27) breaks up into a system of five equations which determine the longitudinal component P_z of the polarization (the z axis is chosen in the direction of the external magnetic field B), two Hermitian-conjugate systems of four equations which determine $P_\pm = P_x \pm iP_y$, and two Hermitian-conjugate equations.

In large external magnetic fields $\exp(\hbar\omega/2T) \gg 1 (\hbar\omega = 2\mu_e B)$ the components of the density matrix have two characteristic relaxation rates—fast and slow. For the diagonal components of the density matrix this corresponds to the fact that the population of the two highest triplet levels falls off with a short decay time $[g(\omega_{ln})]^{-1}$, after which the system approaches thermodynamic equilibrium relatively slowly. In this case we have for the longitudinal component of the polarization

$$P_z(t) = P_z(\infty)(1 - e^{-\tau_1^{-1}t}) + P_z(0)e^{-\tau_1^{-1}t}. \tag{2.29}$$

For the condition $\hbar\omega_0, \zeta\hbar\omega \ll T (\zeta = |\mu_\mu/\mu_e|)$ the inverse value of the longitudinal relaxation time has the form

$$\tau_1^{-1} = 2\nu\left(\frac{\omega_0}{\omega}\right)^2\frac{\hbar(\omega_0 + 2\zeta\omega)}{2T}\operatorname{cth}\frac{\hbar(\omega_0 + 2\zeta\omega)}{4T}. \tag{2.30}$$

The transverse components of the polarization vary with time in the following manner:

$$P_-(t) = P_+^*(t) = P_-(0)\exp[-(\tau_2^{-1} + i\Omega_\perp)t], \tag{2.31}$$

where $\tau_2 = 2\tau_1$ is the transverse relaxation time and the precession frequency

$$\Omega_\perp = \zeta\omega + \frac{\omega_0}{2} \tag{2.32}$$

does not depend on temperature.

For the condition $\hbar\omega_0 \ll T$ the formula (2.30) for the longitudinal relaxation time coincides with the results of Ref. 51. As can be seen from Eq. (2.32), one can search for muonium in metals by measuring the shift of $\Omega_\perp$ in comparison with the muon frequency $\zeta\omega$. At the present time it is possible to resolve muon precession frequencies less than 10^{-9} sec^{-1}; therefore the proposed experiment is possible for $B \sim 5\times10^3$–10^4 G and $T \sim 0.1$ K.

We note that if the relaxation time $\tau_1 < \tau_\mu$, then in principle one can search for muonium by analysis of the equilibrium state, since the polarization $P_z(\infty)$ differs appreciably from the equilibrium polarization of a free muon $P_\mu^{\text{fr}}(\infty) = \text{th}(\hbar\zeta\omega/2T)$. In fact, in fields $\hbar\omega \gtrsim T$ we have

$$P_z(\infty) = \text{th}\left(\frac{\hbar\omega}{2T}\right)\text{th}\left(\frac{\hbar\omega_0}{4T}\right) + P_\mu^{\text{fr}}(\infty). \tag{2.33}$$

For example, at $T = 0.5$ K, $B = 10^4$ G, and $\omega_0 = 0.1\omega_{0v}$ ($\omega_{0v} = 2.8\times10^{10}$ sec^{-1} is the hyperfine splitting frequency in vacuum) we have $P_z(\infty)\approx 0.025$, while $P_\mu^{\mathrm{fr}}(\infty)\approx 0.015$. An experiment in a field perpendicular to the polarization at the initial moment of time is probably most suitable. We note that in such an arrangement, one can carry out a compensation experiment by reversing the direction of the field.

For high temperatures $T \gg \hbar\omega, \hbar\omega_0$ the matrices G, $\tilde{G}$, and G_z can be expanded in a complete set of 4×4 spin matrices. Equation (2.27) can now be written in the following form, retaining only terms linear in $\hbar\omega/T$ and $\hbar\omega_0/T$;

$$\dot{\rho} + i\hbar^{-1}[H_{\mathrm{eff}}, \rho] = 2\nu\Big\{\sigma^n_e\rho\sigma^n_e - 3\rho + ie_{kln}\frac{\hbar\omega_n}{3T}\sigma^k_e\rho\sigma^l_c - \frac{\hbar}{2T}[(\omega_n\sigma^n_e)\rho + \rho(\omega_n\sigma^n_e)] + i\frac{\hbar\omega_0}{8T}e_{kln}(\sigma^n_e\rho\sigma^k_e\sigma^l_\mu - \sigma^k_e\sigma^l_\mu\rho\sigma^n_e) - \frac{\hbar\omega_0}{4T}[(\sigma^n_e\sigma^n_\mu)\rho + \rho(\sigma^n_e\sigma^n_\mu)]\Big\}. \quad (2.34)$$

The equation (2.34) differs from the equations of Refs. 47 and 48 for description of relaxation processes at infinite temperatures. The relaxation term is determined by the polarization of the muonium electron, and the stationary solution is the Gibbs distribution.

Solution of the system (2.34) is obtained by standard methods.[11-13] For the longitudinal component of the muon polarization for $\nu \gg \omega_0$ with accuracy to terms of order $(\omega_0/\nu)^2$ inclusive, we obtain

$$\tau_1^{-1} = \frac{4\nu\omega_0^2}{(8\nu)^2 + \omega_0^2 + \omega^2}\left(1 + \frac{\hbar\omega_0}{4T}\right). \quad (2.35)$$

Equation (2.35) is valid also for $\nu \ll \omega_0$ with the difference that, in the equation for the polarization $P_z(t)$, rapidly oscillating terms are present which are averaged in observation. Thus, as an interpolation formula, Eq. (2.35) is satisfactory with no restrictions. Since $\nu \propto T$ [see Eq. (2.28)], it is easy to see that τ_1^{-1} has a maximum as a function of temperature. The falling branch was obtained previously in Ref. 11.

The transverse component of the polarization is determined for $\nu \gg \omega_0$ by a single root, as in the case of strong fields (2.31), where with accuracy to terms of order $(\omega_0/\nu)^2$ we have

$$\tau_2^{-1} = \frac{\omega_0^2}{32\nu} + \frac{2\nu\omega_0^2}{(8\nu)^2 + [1 + \zeta - (\hbar\omega_0/2T)]^2\omega^2}, \quad (2.36)$$

$$\Omega_\perp = \left\{\zeta + \frac{\hbar\omega_0}{4T} + \frac{(1+\zeta)\omega_0^2}{(16\nu)^2 + 4[1 + \zeta - (\hbar\omega_0/2T)]^2\omega^2}\right\}\omega. \quad (2.37)$$

In Eq. (2.37) for $\omega_0 \sim 10^8$ sec^{-1} and $\nu = 10^9 T$ sec^{-1}, the last term can be neglected even for $T \gtrsim 1$ K, and then the precession frequency can be written as

$$\Omega_\perp = \left(\zeta + \frac{\hbar\omega_0}{4T}\right)\omega, \quad (2.38)$$

which coincides with the precession frequency given in Refs. 27 and 18. For $\nu \ll \omega_0$, ω the solution practially coincides with the result obtained in Ref. 31.

The temperature dependence of the precession frequency (2.38) is easily explained. In fact, in a magnetic field perpendicular to the initial polarization of the muon, the electron of the Mu atom is rapidly polarized along the field direction. Since it does not belong to the Fermi gas of conduction electrons of the metal, its polarization is determined simply by the Boltzmann formula and turns out to be much greater than the polarization of the conduction electrons. Accordingly, a contact-field component appears at the point of location of the muon parallel to the external magnetic field, and therefore the muon effectively precesses in a field $B_{\mathrm{ext}} + \Delta B_{\mathrm{cont}}$. As can be seen from Eq. (2.35), the maximum of τ_1^{-1} is reached for $(8\nu)^2 = \omega_0^2 + \omega^2$. Here τ_1^{-1} is

$$\tau_{1\,\max}^{-1} = \frac{\omega_0}{4\sqrt{1 + (\omega^2/\omega_0^2)}}. \quad (2.39)$$

Thus, a maximum can be observed even for $\omega_0 \sim 10^7$ sec^{-1}, which for a field $B \sim 10^2$–10^3 G corresponds to $\tau_{1\,\max}^{-1} \sim 10^4$–$10^5$ sec^{-1}. Estimates give $\nu = (10^8$–$10^{10})T$ sec^{-1}, and accordingly the maximum should be observed at a temperature $T \sim 0.1$–10 K (Fig. 1). In the region where Eq. (2.36) is valid, the quantity τ_2^{-1} does not have a maximum and falls monotonically with increase of temperature, and at the point of the maximum for τ_1^{-1} it takes on a value (for $\omega^2 \gg \omega_0^2$)

$$\tau_2^{-1} = \tau_{1\,\max}^{-1} + \frac{\omega_0}{4}\frac{\sqrt{1 + (\omega^2/\omega_0^2)}}{1 + (2\omega^2/\omega_0^2)}. \quad (2.40)$$

For specified values of ω and ω_0, we always have $\tau_{1\,\max}^{-1} < \tau_2^{-1} < 2\tau_{1\,\max}^{-1}$.

It follows from Eq. (2.30) that the relaxation rate $\tau_{1\,\max}^{-1} = \tau_1^{-1}/2$ in strong fields ($\exp(\hbar\omega/2T) \gg 1$, $\omega \gg \omega_0$) rises linearly with increase of the temperature, and therefore τ_2^{-1} has a maximum; however, it is observed at temperatures lower than the point of the maximum for τ_1^{-1}. For fields $B \sim 10^3$–10^5 G the maximum for τ_2^{-1} is reached at a temperature $T \sim 0.1$–10 K. For τ_1^{-1} in these fields the maximum should be observed at a temperature $T \sim 10$–100 K. Experimentally it is possible to observe muonium in such fields at the level $\omega_0 \sim 10^8$–10^{10} sec^{-1}. In the case where $(8\nu)^2 \gg \omega_0^2 + \omega^2$ Eqs. (2.35) and (2.36) go over to the corresponding results of Ref. 51.

It should be specially emphasized that both the results of Ref. 49 and the semiquantitative considerations formulated previously in Refs. 27 and 28 are based on a simplified model of the metal. Therefore all of the conclusions of Ref. 49, like those of Refs. 27 and 28, which apply to the equilibrium polarization of the

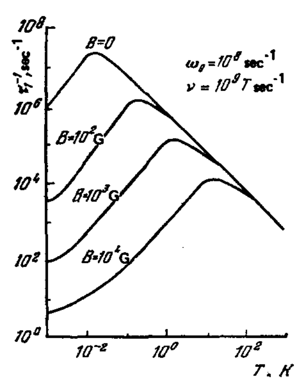

FIG. 1. Dependence of relaxation rate τ_1^{-1} of the longitudinal component of polarization of a positive muon in muonium on the temperature T for various values of the external magnetic field B.

electron and muon in the muonium atom, are valid only in the case where $T > T_K$, the Kondo temperature, when the paramagnetic impurity is a Mu atom. In fact it is well known[56-58] that for $T < T_K$ the magnetic susceptibility of the impurity is small and does not depend on the temperature. Therefore for $T < T_K$ the equilibrium polarization of the electron and muon in a magnetic field is practically equal to zero and does not depend on T. In Refs. 27 and 28 with accuracy 0.1–0.01% no temperature dependence of ω_μ was observed in Al, Zn, and Cu in the temperature range 4–90 K. This fact can be interpreted not only as the absence of a μ^+e^- bound state but also as the fact that T_K for the Mu atom in these metals is greater than the temperature at which the experiment was carried out. Reliable theoretical estimates of the Kondo temperature for this impurity (in particular, for Mu) essentially do not exist at the present time. As shown by the experiment of Ref. 56, for alloys of various types $0.1\ \mathrm{K} < T_K < 600$ K. Therefore the temperature correction to ω_μ could be observed at high temperatures ($T \sim 300$–600 K), where clearly $T > T_K$. However, even for $\omega_0 \sim \omega_{0B}$, the correction will already amount to less than 0.1% for $T \sim 100$ K.

We emphasize also that a clean experiment must be carried out with metals (or isotopes) in which the spins of the nuclei are equal to zero. Indeed, direct dipole-dipole interactions can give a correction to the precession frequency which depends on the diffusion rate [see Eq. (2.22)]. It is also obvious that they can make a decisive contribution to the relaxation rate. Nevertheless the results of Refs. 27 and 28 provide the basis for the assumption that if muonium exists in Al, Zn, and Cu, it is highly "inflated" in view of the screening of the Coulomb interaction by the conduction electrons of the metal. In this way these data place in doubt the theoretical calculations carried out in Ref. 26, where it was found that the hydrogen atom in a metal has practically the same radius of the first Bohr orbit as in vacuum.

Summing up, we can say that for search for the Mu atom in metals it is appreciably more appropriate to study the temperature dependence of the relaxation rate, since the qualitative conclusions of Ref. 49, in particular, the existence of a maximum for τ_1^{-1} (Fig. 1), do not depend on the temperature region in which we are working ($T > T_K$ or $T < T_K$).

Finally, we must note the following fact. It is evident, if $\nu \ll \omega_0$, that in transverse fields it is possible to observe precession with the muonium frequency. The muonium atom is located in interstitial sites of the crystal lattice. If the crystal field at the site has axial symmetry, then the spin Hamiltonian of the muonium is not isotropic,[59] and in monocrystalline samples the precession frequency will depend on the orientation of the crystal in the magnetic field. It is natural to expect this pattern in semimetals. Hartmann *et al.*[60] investigated the precession of the muon spin in samples of As, Bi, and Sb. In the As and Bi samples they observed a very small difference (less than 0.01%), and in Sb a large difference, from the muonium precession frequency observed in Cu. In monocrystalline Sb the precession frequency depended on the orientation of the sample in the magnetic field: The depolarization rate was about $0.2 \times 10^6\ \mathrm{sec}^{-1}$. The Sb lattice has axial symmetry, and the data of Ref. 60 can be interpreted as existence of Mu in Sb. It was also found that the temperature dependence differs somewhat from T^{-1}, but this difference can also be explained by the nonsphericity of the hyperfine-interaction Hamiltonian.[61] It follows from Ref. 60 that the Kondo temperature for Mu in Sb is less than 1 K.

3. SUPERCONDUCTORS

a) Study of type I and type II superconductors

Let us consider the possibilities that the muon method presents for study of an inhomogeneous superconducting state.[62-64] The conceptual aspect of the use of muons reduces as before to the probing of internal magnetic fields.[62,63] As is well known, in superconductors very frequently a situation exists in which the magnetic field partially penetrates the thickness of the material. For example, this may be the so-called intermediate state for Type I superconductors, when as a result of geometrical factors for certain portions of the material the external magnetic field exceeds the critical field and as a result there arises a peculiar mixture of the normal and superconducting phases, often with a very queer geometry. Islands of the normal phase with a frozen magnetic field may remain in a superconductor even after cooling to a temperature much less than the critical temperature T_c if the sample is substantially nonuniform in its physical properties.

As a result of the same inhomogeneity factor in Type I superconductors, normal-phase regions can arise on partial destruction of the superconducting state by a magnetic field or by currents. Generalizing, we can say that any superconductor-normal phase transition of metals, in view of the inhomogeneity of the material, can begin in individual seed regions.

At the present time it is rather complicated to determine satisfactorily the volumes of the normal and superconducting phases when both are present in the sample. The volumes of the normal and superconducting phases are determined extremely easily with the aid of positive muons. Let the external magnetic field **B** be perpendicular to the initial polarization of the muons. Take the direction of polarization for the x axis and the direction of the field for the z axis. Then, if there exists a mixture of normal and superconducting phases in the sample with volumes $1-d$ and d, respectively, the observed precession pattern is described by the formula

$$P_x(t) = (1-d)\int \cos(\gamma b t)\, W(b)\, db + d, \qquad (3.1)$$

$$P_y(t) = (1-d)\int \sin(\gamma b t)\, W(b)\, db.$$

Here $W(b)$ is the probability of a given value of field in the normal phase. The initial polarization we assume equal to unity.

Actually, the superconducting phase enters into the precession pattern as a single piece of the sample in which there is no magnetic field. Averaging of the functions $\cos(\gamma bt)$ and $\sin(\gamma bt)$ with a weight $W(b)$ leads as usual to a damping of the precession pattern. However, it follows from Eq. (3.1) that by measuring the initial amplitude of the precession and the constant component of the polarization we can find the values of $1-d$ and d, i.e., the relative concentrations of the normal and superconducting phases. The error in the determination of the parameter d is due in the last analysis to the set of statistics and without great expenditure of time an experiment can be performed at the present time with accuracy 1% or better.

The probability $W(b)$ is simply the time Fourier amplitude of $P(t)$, and by means of Eq. (3.1) we can determine the probability of distribution of the magnetic field in the sample studied. We note also that the muon method can be useful in observation of any phase transitions, for example, in Kondo systems (for example in alloys of the type $(La_{1-x}Ce_x)Al_2$).[64-66] If the sample studied is thermally well stabilized, the muon method is an extremely reliable instrument for measurement of T_c. As has been noted in the literature,[64,66,67] the existing methods of determination of T_c on the basis of measurement of the magnetization and electrical resistivity in Type I superconductors do not provide the possibility of measuring T_c for samples which are inhomogeneous over their volume, i.e., of determining the existence of residual islands (finely dispersed admixtures) of the normal or superconducting phase. An unambiguous conclusion regarding the volume of the superconducting phase can be obtained only by measuring the specific heat of the sample (the electrons of the normal phase provide a contribution to the heat capacity $c_e \sim T$). Without discussing here the comparative advantages of the two methods, we establish that by means of muons one can determine the same quantity by a new, independent means.

In the study of phase transitions the muon method is equally applicable to Type II superconductors. In addition, since it is just Type II superconductors which, on the one hand, present the greatest practical interest and, on the other hand, are as a rule inhomogeneous in their structure, which leads to irreversibility of phase transitions, the method considered is most promising for study of just these materials.

A magnetic field greater than H_{c_1} in a Type II superconductor forms a completely definite structure (Fig. 2), and in accordance with Eq. (3.1) it is possible to

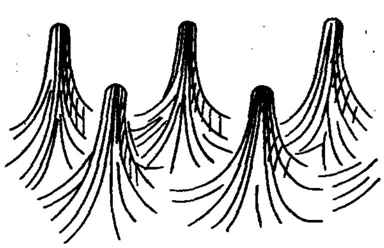

FIG. 2. Structure of magnetic flux for superconductors.

find $W(b)$ by means of muons. As follows from the analytic solution obtained in the classic work of Abrikosov[68] for fields close to H_{c_2}, the system of filaments forms a two-dimensional lattice. For temperatures substantially less than T_c and fields differing appreciably from H_{c_2}, only numerical solutions exist.[68]

The experimental study of the vortex structure in Type II superconductors has been carried out by various means, of which the most promising have turned out to be microphotography of surface powder figures[69] and neutron diffraction. The results have shown that in most cases a triangular lattice is realized, but in a number of samples a square lattice has also been observed, as well as a layering of the sample into macroscopic regions of the Meissner phase (Type I superconductor) and a mixed state.[66,67] The period of the lattice in fields sufficiently large compared to H_{c_2} is of the order 10^2-10^3 Å.

The idea of the possibility of studying a Type II superconductor by means of muons was proposed by Ivanter and Smilga in Ref. 70. In that work they calculated the expected time dependence of the polarization $P(t)$ for triangular and square lattices on the basis of the Abrikosov solution and showed that on the basis of the form of $P(t)$ one can reliably distinguish the types of plane lattice that arise. It is important to note that they were analyzing the variant in which the muon diffusion rate is small and in the course of $10^{-5}-10^{-6}$ sec the muon can be displaced by distances small in comparison with the period of the vortex structure.

It must be said that the question of the diffusion rate of a muon in a Type II superconductor is rather unclear. As a rule, Type II superconductors are alloys with an extremely irregular structure and numerous defects in the lattice. Therefore quantum diffusion along the regular lattice should be suppressed by scattering at the impurities. On the other hand, one can expect that the muons will be trapped in the defects and therefore it is difficult to obtain any theoretical estimates.

We note, however, the attractive possibility of a purely experimental measurement of the diffusion factor. It is evident that if the diffusion rate is sufficiently high, then for each muon on the average we will observe a precession in the effective average field:

$$\langle b \rangle = \int bW(b)\, db. \tag{3.2}$$

In this connection we note that

$$\langle b \rangle S = \Phi_0, \tag{3.3}$$

where S is the area per filament in the vortex lattice (the area of the Bravais unit cell) and $\Phi_0 = 2\times 10^{-7}$ G-cm^2 is the quantum of magnetic flux. For a triangular lattice $S=\sqrt{3}a^2$, where a is the lattice period; for a square lattice $S=a^2$.

It is evident that Eq. (3.3) is valid if the muon during its lifetime travels a distance much greater than the spatial period of the vortex structure, i.e., $\langle r\rangle \sim 10^{-5}-10^{-4}$ cm. Then the effective field in which each muon precesses will not depend on the initial conditions, and on the basis of the precession pattern we shall determine the field $\langle b\rangle$ with a characteristic stochastic spread near this value.

As can be seen from Eq. (3.3), if the period of the lattice for given conditions and a given sample is measured by an independent means (for example, by means of decoration microscopy), then we can find $\langle b\rangle$ and compare it with the observed pattern. However, the most direct possibility of manifestation of the diffusion of positive muons consists in a parallel study of the sample by means of negative muons.

Negative muons are captured by nuclei into a K orbit and, although they lose a significant fraction of their polarization in the cascade process, the remaining polarization is sufficient to reveal the precession pattern. Since the diffusion factor is excluded for negative muons, we obtain here the true distribution $W(b)$. By comparing data on experiments with μ^+ and μ^- mesons we can separate the role of diffusion.

Without going into details, we note only that in experiments with μ^- mesons it is convenient for simplicity of the analysis to select isotopes with zero spins of the nuclei.

At the present time data have been published[71,72] on studies of the alloy $Pb_{0.90}In_{0.10}$ and of Nb by the muon method. The principal results of these authors are shown in Fig. 3.

As can be seen, in the transition of the sample to the superconducting state the Fourier amplitude corresponding to the value of the external field falls off with reduction of the temperature both for PbIn and Nb. On cooling of a PbIn sample in an external field to a temperature 3 K a second peak appears which the authors interpret as the probability of arrival of a muon at the saddle point between normal filaments.

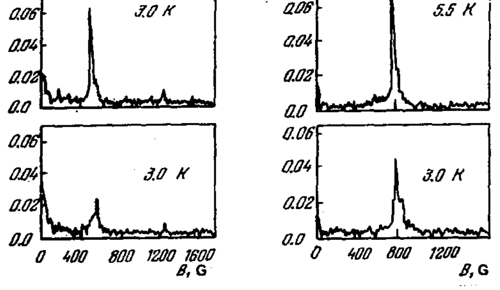

FIG. 3. Fourier transformation of amplitudes of muon polarization precession in Nb and in $Pb_{0.90}In_{0.10}$.

For niobium they observe only a decrease of the Fourier amplitude [or $W(b)$] corresponding to the external field value, and the authors somewhat unexpectedly interpret this circumstance as an indication of rapid diffusion of muons in niobium. As is clear from the foregoing, a shift of the maximum Fourier amplitude in diffusion should be observed to the region of fields lower than the external field, and also a characteristic Gaussian broadening of the line. We note that in Nb the external field, as can be seen from Fig. 3, is 524 G, which is significantly less than the critical field $H_c \sim 2\times10^3$ G. Therefore in the mixed state we cannot expect in practice a constant field value over the cross section of the sample, and $\langle b\rangle$ should differ appreciably from the external field B.

At first glance the results of the authors show that in Nb the vortex filaments have a sharp boundary which separates the normal and superconducting phases. In the absence of diffusion in this case we would observe the pattern shown in Fig. 3. However, in view of the extraordinarily compressed form of the article and the practical absence of detailed discussion by the authors it is difficult to draw any definite conclusions regarding the results of the experiment.

Summing up, we can conclude that the muon method can turn out to be an extremely convenient tool for investigation of the vortex structures in Type II superconductors. It appears that it can compete effectively with neutron diffraction[73] and it is obviously of great interest to study the same samples simultaneously by the two methods under appropriate conditions. The study of vortex structures in the flow regime may also turn out to be very promising. It is obvious that the motion of filaments on flow of a transport current for the observed precession pattern will lead to the same defects as the diffusion of muons in a fixed vortex lattice. On the basis of estimates and the experimental data (which, however, are not too reliable[66,67]) in the flow regime the velocities of the filaments are of the order 10^{-1}–1 cm/sec, and during the muon lifetime of 10^{-6} sec they can move by a distance of the order of the period of the filament lattice.

b) Possibilities of observation of the Mu atom in type I superconductors

Let us consider now the possibility of observing the muonium atom in Type I superconductors. At temperatures close to the critical temperature $T\approx T_c$ most electrons of a superconductor belong to the normal phase, and as in a normal metal the exchange scattering is quite intense. However, on further reduction of the temperature more and more conduction electrons drop into the Cooper condensate and no longer take part in exchange scattering. The number of electrons in the superconducting phase is determined by the characteristic exponential dependence on T, and accordingly the frequency of exchanges of the electron spin, ν, falls off exponentially with temperature.[74-76] Therefore, by lowering the temperature in superconductors, it is easy to traverse the entire region of variation of ν from $\nu\gg\omega_0$ to $\nu\ll\omega_0$. In the analysis of muon spin relaxation in Type I superconductors it is necessary to take into account also the relaxation associated with direct interaction of the muon magnetic moment with the medium. This may be, for example, a dipole-dipole interaction with the magnetic moments of the nuclei or the well known Korringa relaxation.

The Wangsness-Bloch equations for the spin density matrix of muonium have the form[77]

$$\frac{\partial\rho}{\partial t} = -\frac{i}{\hbar}[H, \rho] + 2\nu(\sigma_e\rho\sigma_e - 3\rho) + 2\nu_\mu(\sigma_\mu\rho\sigma_\mu - 3\rho); \tag{3.4}$$

here ν_μ is the relaxation frequency for the muon spin. It is easy to see that Eq. (3.4) is obtained immediately if we only assume that the medium is isotropic and the relaxation process depends linearly on the spin density matrix of the subsystem.

As was noted earlier, it is customary to neglect the relaxation due to direct interaction of the muon magnetic moment with the random fields of the medium, since ν_μ is significantly less than ν. Actually it is easy to show[38] that the ratio ν/ν_μ is at least of the order of the ratio of the squares of the corresponding magnetic moments:

$$\frac{\nu}{\nu_\mu} \sim \left(\frac{m_e}{m_\mu}\right)^2 \approx 4\cdot 10^4.$$

However, as soon as $\nu \gg \omega_0$ and the spin coupling of the muon with the rapidly relaxing electron spin is broken, then, as was shown in the preceding section, the muonium mechanism of relaxation leads to times $\tau \sim 10^{-5}$–10^{-6} sec. As is well known, relaxation times resulting from the direct dipole-dipole interaction are of the same order. Therefore it is useful to find general relations. This problem has been discussed and solved in Ref. 77, where the solution of the system of equations (3.4) was analyzed in the case $B=0$ and simple formulas were obtained for $P(t)$ and the averaging polarization $\langle P\rangle = 3(N - N_+)/N_0$, where N_0 is the total number of decays recorded in an experiment and N_- and N_+ are the total numbers of decays backward and forward. Referring to the study indicated for details, let us go over to analysis of the possibilities for an experiment in superconductors.

It is well known that the nuclear spin relaxation rate in superconductors at temperatures substantially below T_c falls off, roughly speaking, as $\exp(-\Delta/T)$, where Δ is the gap width.[74-76] As has been pointed out, this is explained by the fact that the number of free electrons in superconductors drops exponentially with temperature. Although a quantitative calculation of the electron spin relaxation rate in the electron subsystem in superconductors is difficult because the perturbation theory used in the BCS theory in calculation of the nuclear spin relaxation rate is inapplicable, the qualitative relationship naturally should be retained. Thus, lowering the temperature to values much less than T_c, we can achieve a decrease of the electron spin relaxation rate by several orders of magnitude. (We note that many superconductors satisfy the semi-empirical relation that for $T \ll T_c$ the energy gap is $\Delta \approx 2T_c$.) The Mu atom is a paramagnetic center introduced into the metal, and accordingly the wave function of the superconductor is distorted near it. However, as shown by estimates and experimental data on superconducting alloys with magnetic impurities, this distortion should not lead to a complete destruction of the superconducting state near a paramagnetic impurity. Therefore for exchange of the electron spin in a Mu atom it is necessary as before to expend a certain energy.

Thus, if the muon forms muonium in a superconductor, then by lowering the temperature we can easily achieve fulfillment of the condition $\nu \ll \omega_0$, i.e., we can obtain for the muonium the conditions which are ordinarily observed in good insulators. The complete theory of the behavior of $P(t)$ in this case has been developed in Refs. 12–16. As was shown in that work, the average polarization $\langle P\rangle$, which is equal to approximately unity in an ordinary metal, will decrease rapidly to values close to one half when with reduction of the temperature we reach the region where $\nu \ll \omega_0$. Here it may turn out that in the intermediate region $\langle P\rangle$ passes through a minimum.[12,13] However, if the muon does not form a bound state with an electron in the semiconductor, then the average polarization retains values close to unity, reacting only weakly to change of temperature. In fact, the nuclear spin relaxation rates in an electron subsystem at low temperatures are small and during the possible muon observation time ($\sim 10^{-6}$ sec) do not lead to an appreciable change of the polarization.

It must be emphasized that the picture described will be observed only in the case when the magnetic moments of the nuclei of the superconductor are equal to zero and there is no dipole-dipole relaxation of the electron and muon spins. It is therefore necessary in an experiment to choose appropriate isotopes of the superconductors. Many superconductors are known which have isotopes with zero nuclear spin. For example, we point out Zn(T_c = 0.65 K), Pb(T_c = 7.3 K), Sn(T_c = 4.2 K), Nb(T_c = 3 K), and so forth. (A complete table of superconducting elements is given, for example, in the book by Rose-Innes and Rhoderick.[76]) Therefore the choice of appropriate objects for experiments does not present difficulties.

We note in conclusion that for complete analysis great interest is presented by setting up experiments in parallel in isotopes of superconductors having nonzero nuclear spins. The rate of dipole relaxation of the muon spin, as shown by theoretical estimates and experiments, should be of the order 10^4–10^6 sec^{-1}. Accordingly we can expect values 10^6–10^8 sec^{-1} for the dipole relaxation rate of the electron spin. If muonium does not exist, then by measuring $P(t)$ and $\langle P\rangle$ at temperatures much less than T_c we can measure the rate of dipole-dipole relaxation of the nuclear spin in bulk samples of the superconductor. So far as is known to the authors, there is no other similar possibility. If muonium exists, then by making measurements in isotopes with different magnetic moments of the nuclei and in this way changing τ, we can determine through measurements of $P(t)$ and $\langle P\rangle$ first, the parameters ν_μ and τ for the muon and, second, the dependence of τ on the value of the nuclear magnetic moment.

In parallel with the use of positive muons it is of great interest to use also negative muons. The possibilities of study of the precession of μ^- and the prospects of this technique for analysis of the properties of superconductors have been discussed in Refs. 63 and 78. It appears to us that the muon method opens up extensive prospects for the study of superconductors, and we can only regret that the experimental studies in this field have really not yet begun.

4. FERROMAGNETIC METALS

a) Study of internal magnetic fields by means of muons

Study of the properties of ferromagnetic materials by means of positive muons was begun comparatively recently. The first experimental papers[79-85] appeared in 1973–1975, but interest in this field has been growing continuously and at the present time about fifty papers exist. The first object of study has been the classical ferromagnetic materials Fe, Co, and Ni. Measurements of the fields at the muon in polycrystalline samples at room temperature carried out in the early studies showed that the field at the muon is constant right up to the point of complete magnetization of the sample. At the present time rather extensive experimental data have been accumulated for single crystals and polycrystalline samples of Ni, Co, and Fe at various temperatures and external magnetic fields.[79-95]

In a Co single crystal the variation of the direction of the axis of easy magnetization has been traced for $500 < T < 600$ K, and also the phase transition from hexagonal close packed to face centered cubic at $T = 690$ K.[89,90] Similar measurements have been made in a polycrystalline sample of Gd.[90,92,96-101]

Many experimental and theoretical studies have been devoted to the determination of the contact fields in Ni, Co, and Fe. We note that the theoretical calculations[102-108] of the contact fields do not yet provide agreement with experiment. A comparison of the theoretical calculations with the experimental results, and a detailed bibliography, have been given in several reviews.[31,90,109]

Recently more and more attention is being devoted to the study of ferromagnetic properties of the rare earth metals,[90,92,96-100,110-115] to alloys of the type PdFe,[99] and to magnetic dielectrics.[116] We note also that in Ref. 117 the transition to the ordered magnetic state in spin glasses has been observed.

After a brief summary of the main experimental studies, we shall turn to a theoretical analysis of the behavior of a muon in ferromagnetic metals. Following Refs. 63 and 118–121, we shall make a calculation of the magnetic fields acting on a muon stopped in some interstitial site of the ferromagnetic material. In calculation of the microfield we shall assume that the lattice atoms are localized at sites and shall neglect the distortion of the lattice around the muon. A reliable theoretical calculation with inclusion of the deformation is hardly possible at the present time, but as will be shown below the distortion of the unit cell can be determined experimentally from the discrepancy between the experimental field values acting on the muon and the theoretical values. In accordance with Eq. (1.8) the local field acting on the muon is

$$\mathbf{b}(\mathbf{x}) = \mathbf{B} - \frac{8\pi\mathbf{M}}{3} + \mathbf{b}_r + \mathbf{B}_{\text{cont}}. \tag{4.1}$$

We recall that here $\mathbf{b}_r$ is the field of the dipoles in a Lorentz sphere and **B** and **M** are respectively the macroscopic field and the magnetization of the domain. We emphasize that **B** and **M** differ from the average macroscopic fields and magnetizations $\langle\mathbf{B}\rangle$ and $\langle\mathbf{M}\rangle$ which are usually used in the equations of magnetostatics. Actually the vectors $\langle\mathbf{B}\rangle$ and $\langle\mathbf{M}\rangle$ are the result of averaging over many domains and, for example, in a sample of ellipsoidal shape in an external uniform magnetic field they are constant. The vectors **B** and **M** obviously change their direction in the transition from domain to domain and, furthermore, the macroscopic field **B** inside a domain can, generally speaking, change both in magnitude and in direction within a single domain. The domain magnetization **M** is the saturation magnetization at a given temperature and coincides with the averaged magnetization $\langle\mathbf{M}\rangle$ only for a completely magnetized (single-domain) sample. A theoretical calculation of the field **B** can be carried out only by means of the thermodynamic theory of magnetization, and at the present time has been carried out only for definite models of ferromagnetic materials. For "good" ferromagnetic materials in any not completely magnetized sample it turns out that $\mathbf{B} = 4\pi\mathbf{M}$ over the entire domain and correspondingly $\mathbf{H} \equiv \mathbf{B} - 4\pi\mathbf{M} \equiv 0$. However, this fact is neither trivial nor general. Actually it has been established in Ref. 89 that for magnetization of a Co single crystal in a direction perpendicular to the easy magnetization axis the macroscopic field in a domain rises monotonically from the very first stage of the magnetization.

Although in the first studies on the theory of the application of the muon method to the study of ferromagnetic materials[63,120,121] a clearcut definition was already given and calculations were made of the field acting on the muon, all experimental studies up to the present time[79-81,83-101,110-117,141] have used erroneous formulas for the field acting on the muon. Specifically, authors have failed to understand the fact that the macroscopic field inside the domain **B** cannot be calculated only on the basis of the equations of magnetostatics; in other words, they are confusing the concepts of **B** and $\langle\mathbf{B}\rangle$.

Generally speaking, both the magnitude and the direction of the field **B** can be influenced by the shape and location of the surrounding domains, as well as by the demagnetizing fields, which depend on the concentration and shape of random foreign inclusions and internal stresses, and for polycrystalline materials also on the location of the crystallites.[122-128] The contribution of the closest domains can be estimated roughly by assuming them to be uniformly magnetized spheres. Then it is easy to see that $\delta B \sim 0.1M \ll 4\pi M$, which is significantly less than the macroscopic field of the domain, but the spread in values of the field **b** arising from this may be one of the causes of damping of the polarization vector $\mathbf{P}(t)$.

Let us turn to calculation of the microscopic field of the dipoles $\mathbf{b}_r(\mathbf{x})$. We shall then assume that the field of the magnetic moment of each lattice site coincides with the field of a point dipole. This is equivalent to the statement that the density of magnetic moment is distributed with spherical symmetry with respect to a given site. According to the data of neutron diffraction experiments[124-135] in fcc and bcc lattices the symmetry of the distribution is cubic, while in hcp lattices it is close to spherical.

The components of the microfield from dipoles of a macroscopic region r are calculated from the formula

$$(b_r)_\alpha = M^a_\beta \sum_{l,k} \frac{\partial^2}{\partial x_\alpha \partial x_\beta} \frac{1}{\left|x - x\binom{l}{k}\right|}, \tag{4.2}$$

where l is the number of the unit cell, k is the number of the atom inside the cell, and M^a is the magnetic dipole moment of the atom; obviously $V^{-1}NM^a = M$.

A direct calculation on the basis of Eq. (4.2) is difficult, since the series in Eq. (4.2) converges conditionally. This series can be calculated by Ewald's method,[136-140] and here $\mathbf{b}_r$ can be represented as

$$(b_r)_\alpha = -4\pi n_{\alpha\beta} M_\beta + a_{\alpha\beta} M_\beta, \tag{4.3}$$

where the first term is the demagnetizing field of a uniformly magnetized region r in which the demagnetization coefficients $n_{\alpha\beta}$ take into account the geometry of the region r (for a sphere $n_{\alpha\beta} = \delta_{\alpha\beta}/3$). The second term in Eq. (4.3) $b'_\alpha = a_{\alpha\beta} M_\beta$—the so-called internal dipole field—does not depend either on the geometry or on the size of the region r, and the tensor $a_{\alpha\beta}$ depends only on what kind of site the muon is stopped in. As can be seen directly from Eq. (4.2) we have the fold $a_{\alpha\alpha} = 4\pi$, and therefore for interstitial sites in an fcc lattice which have three axes of fourth order in an octahedral site and of third order in a tetrahedral site (Fig. 4), this tensor is a multiple of the unit tensor $a_{\alpha\beta} = 4\pi\delta_{\alpha\beta}/3$. It is easy to show that in bcc and hcp lattices the two diagonal components coincide. According to Ewald the components of the tensor $a_{\alpha\beta}$ are calculated from the formula

$$a_{\alpha\beta} = VN^{-1} \sum_k \Big[\sum_l R^3 H_{\alpha\beta}\Big(R\Big|\mathbf{x} - \mathbf{x}\binom{l}{k}\Big|\Big) - 4\pi V^{-1} \sum_{h \neq 0} \frac{y_\alpha(h)\, y_\beta(h)}{R^2} \mathcal{G}\Big(\frac{\pi^2 |\mathbf{y}(h)|^2}{R^2}\Big) \exp\Big\{2\pi i \mathbf{y}(h)\Big[\mathbf{x} - \mathbf{x}\binom{0}{k}\Big]\Big\}\Big], \tag{4.4}$$

where N is the number of atoms in the unit cell, V is the volume of the unit cell, and R—an arbitrary positive number—is the series separation parameter for the direct lattice and the reciprocal lattice,

$$H_{\alpha\beta}(x) = \frac{\partial^2}{\partial x_\alpha \partial x_\beta} \frac{2}{\sqrt{\pi}|\mathbf{x}|} \int_{|\mathbf{x}|}^{\infty} \exp(-t^2)\, dt, \tag{4.5}$$

$$\mathcal{G}(u) = \frac{\exp(-u)}{u}, \tag{4.6}$$

$\mathbf{y}(\boldsymbol{h})$ is the reciprocal lattice vector, and $\boldsymbol{h}$ is the Miller index.

Equation (4.3) is applicable, strictly speaking, only for infinite size of the region r. However, the error in the calculations can be arbitrarily small. Corresponding estimates for a sphere, a flat disk, and a long cylinder were obtained in Refs. 136 and 137 and have the form

$$\delta b = O\left(\frac{a}{L}\right)^2 b, \tag{4.7}$$

where a is the lattice period and L is the characteristic dimension of the macroscopic region.

Calculation of the nonzero components of the tensor $a_{\alpha\beta}$ has been carried out by computer.[63,118-121] As usual, one of the coordinate axes was chosen parallel to the symmetry axis of the tensor. The results of this calculation for O_1 and T_1 sites are given in Table I.

Substituting Eq. (4.3) into Eq. (4.1), we write the field b acting on the muon as

$$b_\alpha = B_\alpha - 4\pi M_\alpha + (B_{\text{cont}})_\alpha + a_{\alpha\beta} M_\beta. \tag{4.8}$$

It should be noted that, as can be seen from Figs. 4–6, in a bcc lattice the crystallographically equivalent sites O_1, O_2, O_3 or T_1, T_2, T_3 in the presence of a distinguished direction given by the magnetization M become nonequivalent magnetically and the fields b at them are different. In an hcp lattice the fields b in crystallographically equivalent sites are equal, and in an fcc lattice the field b is identical at all sites.

From the data given in I it is evident that at interstitial sites of a nondeformed fcc lattice the field b′ is equal to $4\pi M/3$, and that at crystallographically equivalent sites of a nondeformed hcp lattice b′ has the same value but the magnitude and direction depend on the direction of magnetization.

Since even for rapid diffusion a muon should not go beyond the limits of a single domain, the behavior of the polarization in nickel and cobalt, where the field at crystallographically equivalent interstitial sites has a single value, will not depend greatly on the diffusion of the muons. In iron, where the microscopic field can take on several values, the diffusion of the muons will lead to a qualitative change of the precession pattern.

In fact, it is clear from simple qualitative consid-

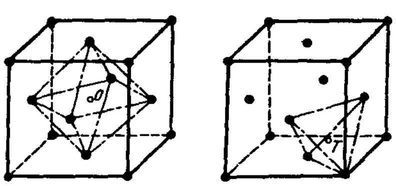

FIG. 4. Lattice of Ni with octahedral interstitial site O and tetrahedral interstitial site T.

TABLE I. Components of the tensor $a_{\alpha\beta}$ in various interstitial sites.

Type of lattice and site	a_{xx}	a_{yy}	a_{zz}	$a_{\alpha\beta}$ ($\alpha \neq \beta$)
bcc, octahedral	−1.165	−1.165	14.930	0
bcc, tetrahedral	5.707	5.707	1.152	0
fcc, octahedral	4.188	4.188	4.188	0
fcc, tetrahedral	4.188	4.188	4.188	0
hcp, octahedral	4.240	4.240	4.086	0
hcp, tetrahedral	4.082	4.082	4.402	0

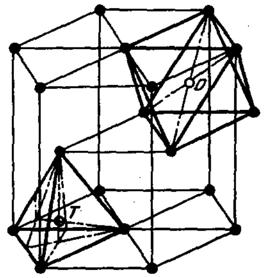

FIG. 5. Lattice of Co with octahedral site O and tetrahedral site T.

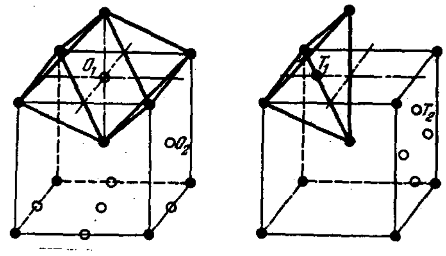

FIG. 6. Lattice of Fe with octahedral sites O_1 and O_2 and tetrahedral sites T_1 and T_2.

erations that, for rapid diffusion in a bcc lattice, single-frequency spin precession should be observed in a field averaged over all sites.[3)] As can be seen from Eqs. (4.2) and (4.3) and simple symmetry considerations, on averaging both over O sites and T sites the term $a_{\alpha\beta}M_\beta$ in Eq. (4.3) is constant and equal to $4\pi M_\alpha/3$ for an arbitrary direction (with respect to the crystallographic axes) of the magnetization. (This can be deduced directly from Table I.) Therefore for rapid diffusion even in a polycrystalline sample single-frequency precession will be observed.

Some refinement is necessary, however. At the initial moment of time the muon can both at O and T sites. One of the states is obviously metastable. Then two variants are possible: 1) outside of the dependence on the inital state, all muons diffuse over O and T sites, arbitrarily changing the type of site in the diffusion process. 2) All muons transfer to stable sites and diffuse only over them. 3) Each muon diffuses only over sites of its own type. In the latter variant there are two forms of muons in the sample: O muons and T muons. Since the contact fields at O and T sites should, generally speaking, differ substantially, in this case we must expect a two-frequency precession. Accordingly, the observation of single-frequency precession indicates that one of the first two possibilities is realized.

Let us turn now to discussion of the experimental data.

At room temperature all authors have observed only a single precession frequency in Fe and a relatively small relaxation rate ($\Lambda = 5$–$10\ \mu\text{sec}^{-1}$).[85] On reduction of the temperature to liquid-nitrogen temperature the relaxation rate rises rapidly.[93,141] In one of the latter experiments[141] observation of several muon-spin precession frequencies was reported in an Fe single crystal at $T = 10$ K. However, there is unfortunately no detailed description of the results available as yet. The data of another experimental group[94] seem also to indicate that at 23 K multifrequency precession is observed, although the spectrum of precession frequencies is not given. At higher temperatures, as in all other studies, single-frequency precession is observed. The fact that at $T = 300$ K only one frequency is observed and the precession signal disappears rapidly with reduction of the temperature indicates unambiguously a rapid diffusion of the muon on the basis of an activation mechanism. The authors' interpretation of the observed six frequencies[141] is somewhat arbitrary: one is completely ignored, three are compared with a tetrahedral site, and two with an octahedral site. However, the entire study has the nature of a preliminary report, and the main result must be considered that at $T = 10$ K multifrequency precession is observed. As follows from the discussion above,[63,121] on dropping of a muon into tetrahedral and octahedral sites only four frequencies should be observed in iron single crystals in the absence of diffusion and an external field. However that may be, the results of Refs. 94 and 141 apparently indicate the absence of quantum diffusion of the muon in Fe.

Interesting data have been obtained in study of the magnetization of Fe, Co, Ni, and Gd.[79,85,142] In iron and nickel the microscopic field at the muon remains unchanged until the sample is magnetized to saturation. Gurevich *et al.*[85] studied thin oblate ellipsoids of rotation magnetized along a major axis. After the sample becomes single-domained, an obvious effect is observed upon further increase of the external field: the change in the field at the muon was equal to the change of the external field. It is an interesting fact that in materials the field **B** inside a domain does not change with high magnetic permeability upon magnetization of the sample. Nevertheless, as can be seen from the well known formula of magnetostatics,

$$\mathfrak{B} = (1 - n)\,\langle \mathbf{H} \rangle + n_x\,\langle \mathbf{B} \rangle, \qquad (4.9)$$

where $\mathfrak{B}$ is the external field and n is the demagnetizing factor. For small n_s there should be an appreciable average field $\langle B \rangle$ inside the sample already in the initial state of magnetization[4)] (samples with $n = 0.11$ and $n = 0.04$ were used in Ref. 85).

Since $\langle \mathbf{B} \rangle = \langle \mathbf{H} \rangle + 4\pi \langle \mathbf{M} \rangle = \mu \langle \mathbf{H} \rangle$, in the initial stage for large $\mu \sim 10^3$–10^4 $(1 - n)\langle \mathbf{H} \rangle \ll n\langle \mathbf{B} \rangle$, in spite of the fact that $n \sim 0.1$. Therefore at the point of complete saturation of the sample the relation $\mathfrak{B}_{\text{sat}} \approx 4\pi n \mathbf{M}_{\text{sat}}$ is fulfilled approximately. It must be recalled, however,

[3)]We shall give a rigorous theoretical proof of this statement below.

[4)]We everywhere call the magnetic induction $\langle \mathbf{B} \rangle$ since it is appropriate to call it the average field. Use of the generally accepted terminology would be extremely awkard in our article. It must be said that at present the historically justified and understandable confusion with the designation of $\langle \mathbf{B} \rangle$—magnetic induction, and of $\langle \mathbf{H} \rangle$—magnetic field, has been ratified by the SI system a century ago.

that $\mu(H)$ decreases on approach to saturation, approaching unity, and actually $\mathfrak{B}_{sat} < 4\pi n M_{sat}$. As can be seen from Eq. (4.8), the macroscopic part of the field acting on the muon is $\mathbf{B} - 4\pi\mathbf{M}_{sat} = \mathbf{H}$. The problem of determining the "field" H inside the domain, as we mentioned above, is a problem of the thermodynamic theory of ferromagnetic materials[124,143,144] and is directly related to calculation of the free energy of a ferromagnetic material. For a definite model of the ferromagnetic material it has been shown theoretically that $\mathbf{H} \approx 0$ for arbitrary magnetization of the sample.[144] As we have pointed out, it was established in Ref. 89, in the case of magnetization of a cobalt single crystal along the difficult axis it was established that the field H begins to change even in the initial stage of magnetization. For Fe and Ni near saturation the field H inside a domain also should differ somewhat from zero. This can be observed in a more detailed analysis of the transition region near saturation. Usually ellipsoids are used as samples, and therefore the macroscopic field of a domain is $\mathbf{B} \equiv \langle \mathbf{B} \rangle$ and therefore is determined by the demagnetizing coefficients of the ellipsoid.

The results of Refs. 79–95 have permitted determination of the contact field at the muon in Fe, Ni, and Co. This characteristic of a ferromagnetic material obviously is important in determination of the magnetizability of the conduction electrons. Attempts to calculate the contact field at the muon theoretically have been made in Refs. 102–107, 145, and 146. For a more accurate experimental determination of the contact field at the muon it is necessary to study carefully the behavior of the field at the muon near the point of complete magnetization. It is of considerable interest to conduct experiments on protons in ferromagnetic metals in parallel NMR.

Let us now consider the case of low (helium) temperatures, in which there is no diffusion. We shall use the results of calculation of the components of the tensor of the internal dipole field for construction of a theory of the muon method in uniformly magnetized single crystals of nickel, cobalt, and iron. Let $W(\mathbf{b}) = W(m)$—the probability that the muon be in a field $\mathbf{b}(m)$—take on a discrete set of values; then according to Eq. (1.7)

$$\mu_{\alpha\beta}(t) = \sum_m W(m)\left\{\frac{b_\alpha(m)\, b_\beta(m)}{b^2(m)} + \left[\delta_{\alpha\beta} - \frac{b_\alpha(m)\, b_\beta(m)}{b^2(m)}\right]\cos(\gamma_\mu b(m)\, t) + e_{\alpha\beta\delta}\frac{b_\delta(m)}{b(m)}\sin(\gamma_\mu b(m)\, t)\right\}. \tag{4.10}$$

In nickel and cobalt, as can be seen from Table I, the fields $\mathbf{b}'$ and, accordingly, b have only a single value if the muon occupies crystallographically equivalent interstitial sites (octahedral or tetrahedral). In cobalt the field values are different for tetrahedral and octahedral sites and differ from $4\pi\mathbf{M}/3$. In iron there are three values of the field $\mathbf{b}'$ in the general case and accordingly three frequencies each in octahedral and tethedral sites. If the crystal is magnetized along the easy axis [100] or along the face diagonal [110], the precession is two-frequency. Here and everywhere below we shall assume that the muon drops into sites of a single type; usually it is assumed that these are octahedral sites.

For magnetization along the [100] axis the field directions are collinear with the external field at all sites, and for magnetization along the [110] axis they are different. Finally, if the crystal is magnetized along the principal cube diagonal (the [111] axis), there is only one frequency. However, the directions of the field in different but crystallographically equivalent sites in this case are not the same. This circumstance leads to a very peculiar behavior of the muons. If the initial polarization P(0) is directed along the [111] axis, Eq. (4.8) takes the form

$$\mathbf{P}(t) = [\cos^2\theta + \sin^2\theta\cos(\gamma b t)]\,\mathbf{P}(0), \tag{4.11}$$

where θ is the angle between b and the [111] direction and

$$\operatorname{tg}^2\theta = 2\left(a_{xx} - \frac{4\pi}{3}\right)^2 M^2\left(\mathbf{B} - \frac{8\pi\mathbf{M}}{3} + \mathbf{B}_{\rm cont}\right)^{-2}. \tag{4.12}$$

Thus, in spite of the fact that the direction of polarization is always constant, its magnitude experiences oscillations. The relation (4.9) can be used for determination of $B_{\rm cont}$.

As can be seen from Table I, the dependence of the modulus of the field $\mathbf{b}'$ on the direction M in iron and cobalt is substantially different in tetrahedral and octahedral sites. Thus, the possibility appears of diagnosing the type of interstitial site on the basis of the change of the precession frequency of the muon polarization in magnetization in different directions. For example, in iron in magnetization along the [100] axis the field $\mathbf{b}'$ is directed along the vector M at two octahedral sites and opposite to it in four. The contact field is collinear with the vector M and in saturation is constant in modulus. Graf *et al.*[89] studied unmagnetized Co at $T = 4.2$ K. Here it was found that the relaxation rate is $\Lambda \sim 10^6$ sec^{-1}. Experiments with a sample completely magnetized along the easy axis have permitted elucidation of the causes of the relaxation. According to the data of Refs. 82 and 83, $B_{\rm cont} = -10.6$ kG. As is well known, in iron $4\pi M = 21.6$ kG. It is then easy to see that by increasing the field we can obtain a situation where the field turns out to be zero in four interstitial sites. In this case the amplitude of precession obviously decreases by a factor of three and only one frequency remains. If the muon has stopped in a tetrahedral site, this field also exists but, in addition, there is a field in which the precession amplitude decreases by a factor of one and one half.

In single crystals of cobalt and iron magnetized to saturation there is the possibility of measuring B and $\mathbf{b}'$. As an example let us consider the case in which a single crystal of iron is magnetized along the [100] axis. As was shown previously, the vector b in this case takes on two values in crystallographically equivalent sites, and the directions of b at the sites are collinear. Then for determination of $\mathbf{b}'$ we have

$$b'(1) - b'(2) = b(1) - b(2), \quad b'(1) + 2b'(2) = 4\pi M. \tag{4.13}$$

In derivation of the system of equations (4.13) we utilized the property $a_{\alpha\alpha} = 4\pi$.

The vector of the microscopic field b is known from experiment. The macroscopic field is known as soon as the demagnetizing factor of the ellipsoid is known.

From the system (4.13) and Eq. (4.8) it is easy to obtain

$$\left.\begin{aligned} b'(1) &= \frac{2}{3}[b(1) - b(2) + 2\pi M], \\ b'(2) &= \frac{1}{3}[4\pi M - b(1) + b(2)], \\ B_{\text{cont}} &= -B + \frac{1}{3}[b(1) + 2b(2) + 8\pi M]. \end{aligned}\right\} \quad (4.14)$$

We note that determination of the internal dipole field can be carried out by magnetizing a single crystal in any direction, for example [110]. To determine the contact field in single crystals of cobalt it is necessary to choose two directions of magnetization: along the axis of the hexagonal lattice and an arbitrary direction in the basal plane.

Comparing the quantities $b'(1)$ and $b'(2)$ with the theoretical values, we can estimate the degree of distortion of an interstitial site on implantation of a muon.

b) Possibilities of the method in study of deformations of crystal lattices

We shall show that the muon method gives the possibility of studying deformations of the crystal lattices of nickel, cobalt, and iron. Study of the deformations and the state of stress of lattices is based on the dependence of the field $\mathbf{b}'$ and the precession frequency of the muon polarization on the deformation of the lattice (Table II). The change of the internal field on deformation of the lattice can be described as follows. The components of the tensor $\tilde{a}_{\alpha\beta}$ for the deformed lattices can be represented in the form of a sum of the components of the tensor $a_{\alpha\beta}$ for undeformed lattices and the fold of some tensor of fourth rank $\Gamma_{\alpha\beta\gamma\delta}$ and a tensor of second rank of the relative deformations $\varepsilon_{\gamma\delta}$:

$$\tilde{a}_{\alpha\beta} = a_{\alpha\beta} + \Gamma_{\alpha\beta\gamma\delta}\varepsilon_{\gamma\delta}. \quad (4.15)$$

Using Eqs. (4.7), (4.10), and (4.15) for the muon spin precession frequency ω, we have

$$\omega^2 = \gamma^2 [B_\alpha - 4\pi M_\alpha + (B_{\text{cont}})_\alpha + a_{\alpha\beta} M_\beta + \Gamma_{\alpha\beta\gamma\delta} M_\beta \varepsilon_{\gamma\delta}]^2. \quad (4.16)$$

The components of the tensor $\Gamma_{\alpha\beta\gamma\delta}$ can be found from the equation

$$\Gamma_{\alpha\beta\gamma\delta} = \sum_{l,k} \frac{\partial^4 \left| \mathbf{x} - \mathbf{x}\binom{l}{k} \right|}{\partial x_\alpha\, \partial x_\beta\, \partial x_\gamma\, \partial x_\delta}. \quad (4.17)$$

Here it is essential that the following condition be satisfied:

$$\Gamma_{\alpha\alpha\gamma\delta}\varepsilon_{\gamma\delta} = 0. \quad (4.18)$$

In undeformed nickel the symmetry of the tensor $a_{\alpha\beta}$ is cubic. If a single crystal of nickel is deformed along the z axis, the cubic symmetry is destroyed and the field acting on the muon depends on the angle between $\mathbf{M}$ and the z axis for a given value of $\mathbf{M}$. In the nickel lattice the tensor $\Gamma_{\alpha\beta\gamma\delta}$ is characterized by axial symmetry, i.e., the following properties exist:

$$\Gamma_{xxxx} = \Gamma_{yyyy} = \Gamma_{zzzz},\ \Gamma_{xxyy} = \Gamma_{xxzz} = \Gamma_{yyzz},\ \Gamma_{xxxx} = -2\Gamma_{xxyy}. \quad (4.19)$$

We shall show that in the lattices of cobalt and iron, deformation can lead to a change of the number of precession frequencies. In fact, in these metals the tensor $a_{\alpha\beta}$ has axial symmetry. On deformation the axial symmetry may be destroyed, since the components of the tensor $\Gamma_{\alpha\beta\gamma\delta}$ have the following properties:

$$\begin{aligned} &\Gamma_{xxxx} = \Gamma_{yyyy} \neq \Gamma_{zzzz},\ \Gamma_{xxzz} = \Gamma_{yyzz} \neq \Gamma_{xxyy}, \\ &\qquad \Gamma_{xxxx} = -(\Gamma_{xxyy} + \Gamma_{xxzz}),\ \Gamma_{zzzz} = -2\Gamma_{xxzz}. \end{aligned} \quad (4.20)$$

It follows from Eqs. (4.12) that in a deformed lattice of cobalt the number of precession frequencies can change when the muon is in an octahedral site for any direction of the deformation axis not coinciding with the axis of the hexagonal lattice, while deformation of an iron crystal produces a change of the number of precession frequencies of the muon polarization when they are in both octahedral and tetrahedral sites. The components of the tensor $\Gamma_{\alpha\beta\gamma\delta}$ for the interstitial sites of fcc, hcp, and bcc lattices are given in Table II.

In iron magnetized along the [100] axis and deformed along the [010] or [001] axes, instead of two-frequency precession there will appear three-frequency precession and beats will be observed. Thus, if the vector $\mathbf{P}(0)$ is perpendicular to the field $\mathbf{b} = b\mathbf{n}$, we find according to Eqs. (4.10) and (1.5) that

$$\begin{aligned} 3\mathbf{P}(t) = \mathbf{P}(0)\cos\omega(1)t &+ [\mathbf{P}(0)\,\mathbf{n}(1)]\sin\omega(1)t \\ &+ 2(\mathbf{P}(0)\cos\omega(2)t \\ &\qquad + [\mathbf{P}(0)\,\mathbf{n}(2)]\sin\omega(2)t)\cos\Omega t, \end{aligned} \quad (4.21)$$

where

$$\omega(1) = \gamma_\mu [b(1) + \Gamma_{xxzz} M\varepsilon], \quad (4.22)$$

$$\omega(2) = \gamma_\mu \left[b(2) + \frac{1}{2} M\varepsilon (\Gamma_{xxzz} + \Gamma_{zzzz})\right], \quad (4.23)$$

$$\Omega = \frac{1}{2}\gamma_\mu M\varepsilon(\Gamma_{xxzz} - \Gamma_{zzzz}). \quad (4.24)$$

As was pointed out above, $b(1)$ and $b(2)$ can be determined from experiments on the precession of muons in undeformed lattices.

The location of a muon in the nickel lattice can be determined if we take into account that the components of the tensor $\Gamma_{\alpha\beta\gamma\delta}$ have different signs in octahedral and tetrahedral sites. For example, if we magnetize nickel along the z axis and deform it along the same axis, then the field at an octahedral site falls off and that at a tetrahedral site increases. To determine the type of interstitial site in cobalt, the crystal must be deformed along a lattice axis not coinciding with the z axis. Then if beats are observed on deformation the interstitial site is octahedral, and if the precession for any direction of deformation is single-frequency the site is tetrahedral. In particular, if a cobalt single

TABLE II. Components of the tensor $\Gamma_{\alpha\beta\gamma\delta}$ at various sites.

Type of lattice and site	Γ_{xxxx}	Γ_{xxyy}	Γ_{xxzz}	Γ_{zzzz}
bcc, octahedral	−13.8	−5.60	19.40	−38.8
bcc, tetrahedral	−14.2	16.90	−2.70	5.4
fcc, octahedral	−23.8	11.90	11.90	−23.8
fcc, tetrahedral	12.0	−6.00	−6.00	12.0
hcp, octahedral	2.16	3.57	−5.73	11.5
hcp, tetrahedral	−7.60	0.222	7.38	−14.8

crystal is deformed along an axis lying in the basal plane (the x axis) and magnetized along the same axis and the polarization $\mathbf{P}(0)$ is directed along the z axis, then when a muon falls into an octahedral site we have

$$3\mathbf{P}(t) = (1 + 2\cos\Omega t)\,[\mathbf{P}(0)\cos\omega t + [\mathbf{P}(0)\,\mathbf{n}]\sin\omega t], \quad (4.25)$$

where

$$\omega = \gamma_\mu b,\ \mathbf{n} = \mathbf{b}/b,\ 8\Omega = 3\gamma_\mu(\Gamma_{xxxx} - \Gamma_{xxyy})\,M\varepsilon.$$

c) Determination of magnetic grain orientation (texture)

We shall now show that the muon method permits study of the question, which is extremely important for practical purposes, of the magnetic grain orientation of transformer steels.

In unoriented unmagnetized materials the spread of the magnetic field modulus does not depend on the direction. If the grain orientation of the material has cubic symmetry, the symmetry of the easy-plane—square type, the field distribution as a rule has similar symmetry. Therefore in such materials we can assume $W(\mathbf{b}) = W(b)W(\mathbf{n})$ and the vector $\mathbf{M}$ is collinear with the axes of easy magnetization. It follows from geometrical considerations that the directions of the principal axes of the tensor $\langle n_\alpha n_\beta\rangle$ in nickel and iron coincide with the [100], [010], and [001] axes. In single crystals of cobalt one of the principal axes coincides with the axis of the hexagonal lattice, and the two others can be chosen arbitrarily. For nonzero components of the tensor $\mu_{\alpha\beta}(t)$ in these axes we obtain [see Eq. (1.7)] we have

$$2\mu_{xx}(t) = 2\mu_{yy}(t) = 1 - \langle n_z^2\rangle + (1 + \langle n_z^2\rangle)\,\mu(t),$$
$$\mu_{zz}(t) = \langle n_z^2\rangle + (1 - \langle n_z^2\rangle)\,\mu(t), \quad (4.26)$$

where

$$\mu(t) = \langle\cos\omega t\rangle. \quad (4.27)$$

If we assume that

$$W(b) = \sum_m \frac{W(m)\,b^2}{(2\pi)^{3/2}} \exp\left[-\frac{(b - \langle b(m)\rangle)^2}{2\delta b^2}\right], \quad (4.28)$$

where $W(m)$ is the probability of occurrence of a muon in an interstitial site with an average value of the field modulus $\langle b(m)\rangle$, then the function $\mu(t)$ will have the form

$$\mu(t) = \sum_m W(m)\cos\gamma_\mu\langle b(m)\rangle\,t\,\exp\left(-\frac{1}{2}\gamma_\mu^2\delta b^2 t^2\right). \quad (4.29)$$

In nickel and cobalt the modulus of the field b takes on a single value and therefore $W = 1$; in unmagnetized iron $W(1) = 1/3$ and $W(2) = 2/3$.

The muon polarization $\mathbf{P}(t)$ for unoriented single crystals of Ni and Co, coincides in direction with the initial polarization for materials possessing cubic symmetry of magnetic orientation, and also for polycrystalline samples $\langle n_z^2\rangle = 1/3$, in accord with Eqs. (4.26), (1.5), and (1.9) and

$$\mathbf{P}^c = \frac{1}{3}\mathbf{P}(0), \quad \mathbf{P}^v(t) = \frac{2}{3}\mathbf{P}(0)\,\mu(t). \quad (4.30)$$

In the case when symmetry of the "easy-plane" type or square symmetry occurs, $\langle n_z^2\rangle = 0$, and the initial polarization lies in the xy plane, we have

$$\mathbf{P}^c = \frac{1}{2}\mathbf{P}(0), \quad \mathbf{P}^v(t) = \frac{1}{2}\mathbf{P}(0)\,\mu(t), \quad (4.31)$$

and when the initial polarization is parallel to the z axis:

$$\mathbf{P}^c = 0, \quad \mathbf{P}^v(t) = \mathbf{P}(0)\,\mu(t). \quad (4.32)$$

The domain structure with square symmetry has been observed by neutron diffraction in unmagnetized single crystals of silicon iron having the shape of a cylinder cut along the [100] axis, according to Elyutin *et al.*,[147] who also showed that a rotation of the magnetization vector of the domains occurs upon application of an external field along the axis of the cylinder. It should be noted that the value of $\langle n_z^2\rangle$ and the muon spin precession frequency change upon rotation of the magnetization vector; here if the magnetization is parallel to the [111] axis, all three frequencies coincide and one should observe single-frequency precession, and for magnetization along the [110] axis—two-frequency precession.

In the case of uniaxial oriented unmagnetized materials the behavior of the muon polarization is described by the formula

$$\mathbf{P}^c = \langle n_z^2\rangle\,\mathbf{P}(0), \quad \mathbf{P}^v(t) = \mathbf{P}(0)\,(1 - \langle n_z^2\rangle)\,\mu(t) \quad (4.33)$$

when the initial polarization is parallel to the grain orientation axis z, and by the formula

$$\mathbf{P}^c = \frac{1}{2}(1 - \langle n_z^2\rangle)\,\mathbf{P}(0), \quad \mathbf{P}^v(t) = \frac{1}{2}(1 + \langle n_z^2\rangle)\,\mathbf{P}(0)\,\mu(t) \quad (4.34)$$

when the initial polarization is perpendicular to the grain orientation axis.

Thus, the muon method gives a simple means of determining the grain orientation axes and the distribution of internal fields in ferromagnetic materials. This prospect is particularly real for the analysis of magnetic grain orientation in transformer steels.

d) Diffusion of muons in iron single crystals

Let us consider the diffusion of muons in single crystals of iron magnetized along the [001] axis and along the principal cube diagonal [111]. For magnetization along the easy axis [001] the field $\mathbf{b}$ takes on two different values, both in octahedral sites and tetrahedral sites. In what follows we shall assume for definiteness that the muons are located in and diffused over octahedral sites.

First consider the case in which the magnetization is directed along the [001] axis. Here the internal fields have two different values: $\mathbf{b}(1)$ at a O_1 site and $\mathbf{b}(2)$ at O_2 and O_3 sites (see Fig. 6). It is evident that $W(1) = 1/3$ and $W(2) = 2/3$. If we assume that the muon "jumps" only to the closest interstitial site, then $\lambda_{12} = \lambda/2$, $\lambda_{21} = \lambda$, where λ is the frequency of jumps over interstitial sites.

The fields $\mathbf{b}(1)$ and $\mathbf{b}(2)$ are directed along the z axis, and therefore P_z does not change, and for the transverse component P_+ we obtain from the system of equations (2.2)[119]

$$P_+(t) = (A_+ e^{-(\nu_+ - i\omega_+)t} + A_- e^{-(\nu_- - i\omega_-)t})\,P_+(0); \quad (4.35)$$

here

$$
\left.\begin{aligned}
A_{\pm} &= \frac{1}{2}\{1 \pm [x + i(W(2) - W(1))]z^{-1}\},\\
z &= z' + iz'' = \sqrt{x^2 - 1 + 2ix\,\frac{\lambda_{21} - \lambda_{12}}{\lambda_{21} + \lambda_{12}}}, \quad z' > 0,\\
x &= (\lambda_{12} + \lambda_{21})[\omega(1) - \omega(2)]^{-1} = [\omega(1) - \omega(2)]^{-1}\,3\lambda/2,\\
\omega(1) &= \gamma_\mu b_z(1), \quad \omega(2) = \gamma_\mu b_z(2),\\
\nu_{\pm} &= -\frac{1}{2}[\omega(1) - \omega(2)](x \mp z'),\\
\omega_{\pm} &= -\frac{1}{2}[\omega(1) + \omega(2)] \pm \frac{1}{2}[\omega(1) - \omega(2)]z''.
\end{aligned}\right\} \quad (4.36)
$$

The relaxation rates are given in Fig. 7. It can be seen that for all x the value of ν_- is several times larger than ν_+, and therefore the principal contribution to the polarization is from the first term in Eq. (4.35). For $x \approx 0.9$, ν_+ has a maximum, i.e., the most rapid depolarization should be observed for $\lambda \approx 0.6[\omega(1) - \omega(2)]$.

In the limiting cases of fast diffusion ($x \gg 1$) and slow diffusion the expressions (4.35) and (4.36) are simplified and simple answers are obtained for the polarization. In the first case

$$P_+(t) = \exp(-i\langle\omega\rangle t)\exp(-\Lambda_2 t)\,P_+(0), \quad (4.37)$$

where

$$
\begin{aligned}
\langle\omega\rangle &= \frac{\omega(1)\lambda_{12} + \omega(2)\lambda_{21}}{\lambda_{12} + \lambda_{21}} = W(1)\omega(1) + W(2)\omega(2),\\
\Lambda_2 &= [\omega(1) - \omega(2)]^2\,\frac{\lambda_{12}\lambda_{21}}{(\lambda_{12} + \lambda_{21})^2}\,\frac{1}{\lambda} \approx \frac{4}{27}[\omega(1) - \omega(2)]^2\lambda^{-1}. \quad (4.38)
\end{aligned}
$$

For slow diffusion ($x \ll 1$)

$$
P_+(t) = \left\{\frac{1}{3}\exp\left[-\left(i\omega(1) + \frac{2}{3}\lambda\right)t\right] + 2\exp\left[-\left(i\omega(2) + \frac{\lambda}{3}\right)t\right]\right\}P_+(0), \quad (4.39)
$$

i.e., two-frequency precession should be observed with relaxation times proportional to the diffusion rate.

In the case in which the magnetization is directed along the [111] axis, in octahedral sites there are three values of the microfield different in direction but identical in modulus. For $\lambda \gg \omega$ (fast diffusion, $\omega = \gamma_\mu b$) in the coordinate system whose Z axis is directed along the [111] axis we find

$$P_z(t) = P_z(0)\exp(-\Lambda_1 t), \quad (4.40)$$

$$P_+(t) = P_+(0)\exp\left[-\left(i\langle\omega\rangle + \frac{1}{2}\Lambda_1\right)t\right], \quad (4.41)$$

where

$$\langle\omega\rangle = \frac{1}{3}\gamma_\mu|\mathbf{b}(1) + \mathbf{b}(2) + \mathbf{b}(3)| = \gamma_\mu b_z, \quad \Lambda_1 = \gamma_\mu^2 b_\perp^2\lambda^{-1}. \quad (4.42)$$

For slow diffusion

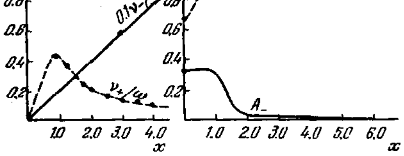

FIG. 7. Dependence of $\nu_\pm$ and $A_\pm$ on x ($\omega = \omega(1) - \omega(2)/2$).

$$
P_z(t) = \left\{\cos^2\theta\exp\left(-\sin^2\theta\cdot\frac{3}{2}\lambda t\right) + \sin^2\theta\cos\omega t\cdot\exp\left[-\frac{3}{4}(1 + \cos^2\theta)\lambda t\right]\right\}P_z(0). \quad (4.43)
$$

Here θ is the angle between the [111] direction and the microfield, which is determined by Eq. (4.12). Naturally as $\lambda \to 0$ Eq. (4.43) goes over into Eq. (4.11).

In iron the characteristic microfields are of the order of several kilogauss and accordingly the maximum relaxation can be expected for $\lambda \sim 10^9\ \mathrm{sec}^{-1}$. Data on diffusion of hydrogen in iron give $\lambda = \lambda_0\exp(-\varphi/T)$, where $\lambda_0 = 10^{11}$–$10^{13}\ \mathrm{sec}^{-1}$ and the activation barrier is $\varphi = 500 - 1000$ K. Therefore at room temperature $\lambda \sim 10^{11}\ \mathrm{sec}^{-1}$ (Ref. 148). It follows from this that at low temperatures (of the order of helium temperatures) superbarrier diffusion can be suppressed practically completely and the only possible form remaining is so-called quantum diffusion, the theory of which has been developed in Refs. 149–151. Since the internal magnetic fields in completely magnetized single crystals of iron can be accurately measured,[85] and there is no stochastic spread either in magnitude or in direction, they represent an ideal object for investigation of muon diffusion, particularly for the study of quantum diffusion. The existence of a maximum in the relaxation rate also greatly facilitates the analysis, interpretation, and identification of the causes of damping.

e) Use of the method for study of rare earth metals

We now turn to a brief description of the possibilities of the muon method for investigation of rare earth metals (REM). The muon method is evoking special interest in this connection. In fact, although there is a direct method of study of the magnetic structures of REM—neutron diffraction, nevertheless, in view of the complicated nature of these structures, the deciphering of neutron diffraction data for many cases has been the subject of discussion.[152] A detailed analysis of the entire problem is given in the book by Taylor and Darby.[152]

It appears that the muon method is the most direct means of identification of the magnetic structures of REM. Naturally the experiments are best carried out in single-crystal samples but, as experiments show,[153] interesting data can be obtained for the simplest structures even in polycrystalline samples. At the present time several experimental studies have been published[90,92,93,96–101] and a purely qualitative explanation has been given for the behavior of the precession frequency as a function of temperature in REM. As yet no calculation has been made of the dipole fields at crystallographically possible interstitial sites even for the simplest magnetic structures and especially for the complicated antiferromagnetic structures of REM. For the simplest structures with magnetic moments of all lattice atoms constant both in magnitude and direction, the dipole field is easily calculated by means of the theory presented above. Such structures are probably to be found, in Dy at $T < 85$ K, in Gd at temperatures below 293 K, and in Tb at temperatures below 221 K. These metals have a somewhat distorted hcp lattice, and for calculation of the dipole fields one can use the results of calculation of the tensor $a_{\alpha\beta}$ given in Table I. For hcp lattices the components of the tensor are determined only by the ratio c/a; the maximum deviation of this ratio from $c_0/a_0=1.633$ in an ideal hcp lattice in all REM is less than 4%, i.e., they are all described as a hcp lattice weakly deformed along the C axis: $\varepsilon_{zz}=(c-c_0)/c_0=0.6124(c/a-1.633)$. Using Eq. (4.13) and the values of $a_{\alpha\beta}$ and Γ_{zzzz} from Table II, we have for the quantity Δ

$$\Delta=a_{xx}-\frac{4\pi}{3}=-\frac{1}{2}\left(a_{zz}-\frac{4\pi}{3}\right)$$
$$=\begin{cases}0.0514+3.509\left(1.633-\frac{c}{a}\right) & \text{for octahedral site},\\ -0.107-4.53\left(1.633-\frac{c}{a}\right) & \text{for tetrahedral site}.\end{cases}\quad(4.44)$$

We note that the value of Δ given in Refs. 90 and 92 is incorrect. Accordingly, the plots for the dipole fields at the muon in octahedral and tetrahedral sites there are also erroneous.

Knowing the dipole fields and the magnetization of the domain, one can evaluate the contact fields at the muon. As has been shown by experiments in Gd, the resulting field at the muon is of the order of 1 kG. For an accurate determination of the contact fields and elucidation of the location of the muon in the lattice, it is necessary to make measurements in uniformly magnetized single crystals and in deformed single crystals. The possibilities of similar experiments have already been discussed in detail for Fe. In REM, as in Co, the field H from neighboring domains in an unmagnetized sample can penetrate most rapidly into the domain where the muon has stopped, which leads to a spread of the fields in the domain and to damping of the amplitude of the polarization oscillations. For this reason a high rate of damping should be observed in polycrystalline samples. As a result of the comparatively large magnetic anisotropy constants, the relaxation rate will increase with increase in the external field, which has also been observed experimentally.[96]

Let us illustrate briefly the possibilities of the method, discussing the qualitative features of the behavior of the dipole fields in REM (with hcp structure), in which, in the transition from one hexagonal plane to another, the magnetic-moment vectors of the atoms are rotated by an angle φ around the hexagonal axis (the z axis), remaining constant in modulus. Of the ferromagnetic materials this structure (a conical spiral) is found in Ho and Er for $T<20$ K and also Tb, Dy, and Ho in the antiferromagnetic state (spiral in basal plane).[152] Since the magnetic moment of the atoms varies within a single crystallographic unit cell, Eq. (4.2) for the dipole field must be written in the form

$$(b_r)_\alpha=\sum_{l,k}\frac{\partial^2}{\partial x_\alpha\,\partial x_\beta}\frac{1}{\left|\mathbf{x}-\mathbf{x}\binom{l}{k}\right|}M^{a}_{\beta}\binom{l}{k}. \quad (4.45)$$

In the case discussed, the vector $M^{a}_{\beta}\binom{l}{k}$ is constant in each hexagonal plane and therefore its direction will depend only on the index h that determines the number of the plane. Therefore it is convenient in Eq. (4.45) to change the order of summation, carrying it out first over each plane. Obviously the component M^{a}_{z} does not depend on h at all. The transverse components are conveniently written in the form

$$M^{a}_{+}(h)=M^{a}_{x}(h)+iM^{a}_{y}(h)=M^{a}_{+}(0)\,e^{i\varphi}. \quad (4.46)$$

From Eq. (4.45) it is easy to see that at interstitial sites located on the hexagonal symmetry axes, the only component of the dipole field that is different from zero, and that is proportional to M^{a}_{z}, will be $b'_z=a_{zz}M^{a}_{z}\nu$. Here ν is the specific volume and a_{zz} is determined by Eq. (4.44). From Eqs. (4.45) and (4.46) it follows that

$$b_+(h)=\nu a(\varphi)\,M^{a}_{+}(0)\,e^{ih\varphi}. \quad (4.47)$$

For a conical spiral structure in which the magnetic moment of the atom forms an angle θ with the hexagonal axis, the magnetic field at an interstitial site is inclined to this axis at an angle θ': $\cos\theta'=b_z/b$, $b=\sqrt{b_z^2+a^2(\varphi)M_+^{a2}\nu^2}$; after averaging over all sites of a single type (for simplicity, as usual, we assume that the muon sticks in sites of a single type), we have the following, in accord with Eq. (1.7), for the nonzero components of the tensor $\mu_{\alpha\beta}(t)$ which determines the behavior of the polarization, and without account of damping

$$\mu_{xx}(t)=\mu_{yy}(t)=1/2\,[\sin^2\theta'+(1+\cos^2\theta')\cos\omega t],$$
$$\mu_{zz}(t)=\cos^2\theta'+\sin^2\theta'\cos\omega t. \quad (4.48)$$

In the particular case of a spiral structure in the basal plane, the polarization is given by formulas similar to (4.31) and (4.32), which describe the magnetic grain orientation of the easy-plane type. Thus, on change of the direction of initial polarization of the muon $P(0)$ with respect to the C axis, the ratio of the constant and oscillating parts of the muon spin polarization will change, and in this way we have the possibility of measuring the angle θ.

A different situation exists in the antiferromagnetic material Cr, in which the magnetic structure is of the spin density wave type with a period of 58 lattice constants. In the absence of diffusion, complete depolarization should be observed as a result of the large choice of precession frequencies. However, diffusion plays an important role both in Fe and Cr with a bcc structure, right down to a few degrees K, and precession is observed[98] in a field close to the external field. The dipole fields produce rather rapid damping: the relaxation rate is $\Lambda\sim10^7$ sec^{-1} and drops rapidly in the transition of Cr to the paramagnetic state.

It should be emphasized particularly that the relaxation rate Λ in the muon method is extremely sensitive to magnetic phase transitions. For example, in REM antiferromagnetic transitions have been observed at the Néel temperature, and in Cr it has been possible to trace clearly the phase transition between states with two types of spin density waves.

f) Muonium in ferromagnetic metals

There are at present no direct indications that the Mu atom exists in ferromagnetic metals. We note, however, that the results of Refs. 92, 94, and 109 can in principle be interpreted by means of the hypothesis of formation of the Mu atom in iron. The observed dependence $b(T)$ of the field at the muon on the temperature did not agree with the well known Brillouin function $B_S(T)$, i.e, the contact field at the muon varied with T and not in proportion to the magnetization. For Fe, for example, the deviation reaches 30%. Generally speaking, this is just what should occur if the Mu atom is formed. The electron of muonium in a ferromagnetic material, as the result of strong exchange interactions with the electrons of the metal, can turn out to be highly polarized. In accordance with the discussion in Chapter 2, an additional field $\Delta B = -\hbar\omega_{0m}P_e/2\mu_\mu$ is then induced at the muon. The electron polarization is $P_e = \text{th}(\mu_e H_{ex}/T)$, where H_{ex} is the effective resultant exchange field produced by the s and d electrons of the ferromagnetic material at the muonium electron. Obviously we can assume that $H_{ex} \sim M(T)$. We note that a similar discussion for Mn ions in Fe has been given in Refs. 154 and 155.

The contact field of the s electrons at the muon is also determined by the Brillouin function $B_K(T) = B_K(0)B_S(T)$, and we finally obtain for the field $b(T)$ at the muon

$$b(T)=\left[\frac{4\pi}{3}M(0)+B_K(0)\right]B_S(T)-\frac{\hbar\omega_{0m}}{2\mu_\mu}\,\text{th}\frac{T_{ex}}{TB_S(T)}. \qquad (4.49)$$

Here T_{ex} is determined by the exchange integrals. An illustrative evaluation of ω_{0m} and T_{ex} from the data of Ref. 94 leads to reasonable values $\hbar\omega_{0m}/2\mu_\mu \approx 10$ kG, $T_{ex} \sim 3000$ K. It is also of interest to carry out experiments in external fields greater than the saturation field. Then an additional term appears in the right-hand side of Eq. (4.49).

The deviation from the Brillouin law in the contact fields is observed also in other ferromagnetic materials, but as yet there is no serious reason to relate these deviations to the existence of Mu in these metals, although one must not forget this possibility. We note in conclusion that special interest is presented by the search for muonium in ferrites and antiferromagnetic dielectrics.

5. CONCLUSION

In this paper we have discussed only the group of problems related to use of the muon method for study of metals, leaving aside numerous other possibilities. The first results of the muon method in the study of metals already show that experimental physics has obtained a new tool which permits study of the internal magnetic fields in metals, various phase transitions, the properties of the superconducting state, quadrupole interactions, deformations of a crystal lattice on arrival of a muon at an interstitial site, quantum and classical diffusion of a muon over the lattice, the charge state of the muon (or proton), and a number of other questions of metal physics.

We note particularly that for ferromagnetic metals the muon method appears to have possibilities greater than the existing methods.

These studies have only begun, and a wide range of problems accessible by the muon method have not yet been touched. We can point out, for example, questions related to the De Haas-van Alphen effect, the topology of the Fermi surface, the potential relief of crystal fields, the nature of radiation damage and effects in metals, including the processes of catalysis and adsorption.

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