Sub-barrier diffusion of μ^+ mesons in copper

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A method for determining the diffusion coefficient of μ^+ mesons in solids is proposed. It is based on measurement of the spin dipole relaxation rate of the diffusing μ^+ mesons. This method is used to study the diffusion of μ^+ meson in copper. It is shown that μ^+ meson diffusion in copper is a sub-barrier process at temperatures $T < 250$°K.

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1. INTRODUCTION

Diffusion of hydrogen and heavier atoms in metals has been intensively studied for many years. In this paper we describe for the first time[1)] the diffusion of a positive muon, which can be regarded in this process as a lighter isotope of hydrogen. The small mass of the positive muon has made it possible to observe a new process, wherein the muon diffuses by penetrating under the potential barrier separating the equivalent positions of this particle in neighboring unit cells of a crystal. The observation of the below-barrier diffusion of the positive muon in copper supplements the already available data on above-barrier diffusion of hydrogen in metals and greatly extends the possible region where the theory of this process can be compared with experiment. Let us consider briefly the main experimental results obtained in the study of the diffusion of hydrogen and the positive muon, and let us compare the diffusion of these particles.

In numerous works (see, e.g., the review by Wert[2] and a number of other papers,[3,4] where a rather detailed bibliography is given), it was found that the temperature dependence of the diffusion coefficient of hydrogen and heavier atoms is given by

$$D=D_0e^{-Q/T}. \quad (1)$$

Using the relation $D = a^2/\tau$ (here a is the dimension of the unit cell of the crystal in which the particles diffuse, and τ is the average time spent by the diffusing particle in one crystal cell), and also assuming that $D_0 = \nu a^2$, we can rewrite the experimental relation (1) in the form

$$1/\tau=\nu e^{-Q/T}. \quad (2)$$

In the study of the diffusion of hydrogen (protons) it was found that the parameter $\nu = \nu_p$ in (2) is the same for all the investigated metals and is equal to $\sim 10^{13}$ sec^{-1}. The same value of the parameter ν characterizes the diffusion of heavy hydrogen isotopes (D^2 and T^3) in metals, and also of atoms of heavier elements (C, N, O). The agreement between the experimental value of the parameter $\nu_p \sim 10^{13}$ sec^{-1} and the frequency ν_0 of the particle vibrations in the crystal lattice of the metal enables us to interpret the relation (2) obtained for the proton (and heavier particles) as above-barrier diffusion, where $1/\tau$ is the probability that the particle will jump from one unit cell to another, and Q is the height of the potential barrier. It was found that the barrier height Q_p for the proton in various metals fluctuates in the range $Q_p = 1000-6000$°K.

The temperature dependence of $1/\tau = f(T)$ measured in the present study for the diffusion of the positive muon in metal can also be described by formula (2), but the values (11) and (12) obtained in this case for the parameters ν_μ and Q_μ (see below) differ greatly from the corresponding values of ν_p and Q_p for the proton: $\nu_\mu = 10^{7.5}$ sec^{-1}, $Q_\nu = 560$°K (the parameter Q_p in copper is equal to $(Q_p)_{Cu} = 4600$°K[5–8]). The value $\nu_\mu = 10^{7.5}$ sec^{-1} is too low to be interpreted as the oscillation frequency of the positive muon in the copper lattice. It follows therefore that the diffusion of the positive muon in copper can not be above the barrier, and the parameter ν_μ, unlike the case of the proton, is not an oscillation frequency.

A simple calculation shows that the small muon mass should lead to the possibility of above-barrier diffusion of these particles, and this indeed explains the experimentally observed regularities in the diffusion of positive muons in copper. For under-barrier diffusion, the probability $1/\tau$ is given by

$$1/\tau=\nu_0 \exp\{-2\hbar^{-1}(2mU)^{1/2}b\}F(T). \quad (3)$$

Here U and b are respectively the height and width of the barrier, m is the muon mass, $\nu_0 = 10^{13}$ sec^{-1} is the frequency of the μ^+-meson oscillations in the copper lattice, and $F(T) = \exp(-Q_\mu/T)$ (see below). The low value of the experimental parameter $\nu_\mu \ll 10^{13}$ sec^{-1} is thus the result of the low transparency of the potential barrier. The parameter Q_μ, which determines the temperature dependence of $1/\tau$ in (3), can be interpreted[9] as the activation energy that must be consumed to expand the pore in the neighboring cell into which the μ^+-meson diffuses.

Of course, the positive muon is not the only object for the observation of below-barrier diffusion. In principle this process can be observed also for heavier particles. But the resultant very small value of the below-barrier diffusion makes such experiments extremely difficult.

The present work was performed with the JINR synchrocyclotron in Dubna.

2. EXPERIMENTAL STUDY OF THE μ^+-MESON DIFFUSION

The diffusion of the positive muon was revealed by the change of the relaxation rate of the muon spin due to dipole interaction with the magnetic moments of the nuclei of the material. The maximum relaxation rate should be observed at low temperature, when the time τ

that the muon stays in one unit cell is much longer than the observation time, i.e., when there is practically no diffusion. When the μ^+-meson diffuses over the crystal, the nuclear magnetic fields at the muon become variable in time and the relaxation rate of the muon spin decreases.

In the experiment, the rate of relaxation of the muon spin was determined from the damping of the muon precession amplitude in a transverse magnetic field. The damping of the μ^+-meson precession is due to the fact that the nuclear magnetic fields are different at different μ^+-mesons, and therefore the precession rates of the spins of the individual μ^+-mesons become unequal and the observable precession is damped.

The precession of the μ^+ meson was observed by registering the positrons from the $\mu^+ \to e^+$ decay, which are emitted predominantly in the direction of the μ^+-meson spin. The resultant time dependence of the counting rate of the positrons emitted in a direction opposite to that of the primary μ^+-meson polarization is then written in the form

$$dN/dt = N_0 e^{-\Lambda_0 t}[1 - P(t)\, a \cos \omega t]. \quad (4)$$

Here a is the experimental asymmetry coefficient of the angular distribution of the positrons from the $\mu^+ \to e^+$ decay at t = 0, $\omega = eB/mc$ is the frequency of the Larmor precession of the μ^+-meson in a transverse magnetic field B, $\Lambda_0 = 4.55 \times 10^5\ \text{sec}^{-1}$ is the rate of decay of the μ^+-meson, and P(t) is a function that determines the time dependence of the rate of muon spin relaxation due to dipole interaction and diffusion.

The expression for the function P(t) was derived by Abragam;[10]

$$P(t) = \exp\{-2\sigma^2\tau^2[e^{-t/\tau} - 1 + t/\tau]\}. \quad (5)$$

Formula (5) was obtained under the assumption that in the absence of diffusion ($\tau \gg t$) the distribution of the magnetic fields at the μ^+-mesons, and consequently also the function P(t), take a Gaussian form:

$$P(t) = e^{-\sigma^2 t^2} \quad \text{at} \quad \tau \gg t. \quad (6)$$

The parameter σ in (5) is thus the rate of the muon spin relaxation due to the dipole interactions when there is no diffusion. A calculation of the distribution of the nuclear magnetic fields at the μ^+ muon has shown that in the absence of diffusion the function P(t) is indeed well approximated by (6). Experiment has also confirmed the Gaussian form of P(t) at low temperatures, when there is no diffusion (see the table).

The parameter σ can in principle be obtained by calculation, but the result will depend on the location of the muon in the crystal cell. In our work, the value of σ was determined experimentally from the damped precession of the μ^+-meson in the absence of diffusion, i.e., when the rate of damping ceased to depend on the temperature. It should be noted that in the other limiting case, $\tau \ll t$, i.e., in fast diffusion, Eq. (5) for P(t) becomes exponential:

$$P(t) = e^{-2\sigma^2 \tau t} \quad \text{at} \quad \tau \ll t. \quad (7)$$

Observation of the damped precession $(dN/dt)_{exp}$ enables us to determine the time τ characterizing the μ^+-meson diffusion in a given substance. The time τ was determined by comparing the experimental spectrum $(dN/dt)_{exp}$, by least squares, with the expression (4), where the function P(t) was determined by (5) and the value of σ was obtained experimentally as indicated above.

The detailed experimental setup for the observation of the μ^+-meson precession in a transverse magnetic field and the corresponding electronic apparatus are described in [11].

3. DIFFUSION OF μ^+ MESON IN COPPER

We investigated the diffusion of a μ^+-meson in single-crystal and polycrystalline copper samples. The impurities in these samples did not exceed 10^{-3} %. The experimental spectra $(dN/dt)_{exp}$ illustrating the damped precession of the μ^+-meson in copper are shown in Fig. 1, from which it is seen that the undamped muon precession in copper, usually observed at room temperature, becomes damped with decreasing temperature. The temperature dependence of the precession damping rate $\Lambda(T)$ in the single-crystal sample of copper is shown in Fig. 2. It is seen from the presented data that Λ becomes constant at sufficiently low temperatures ($T \lesssim 70°K$), as it should when there is no diffusion. The lower experimental values of Λ (compared with the calculated $\Lambda(T)$ dependence) observed at $T \gtrsim 270°K$, are due to the influence of above-barrier diffusion, which becomes appreciable at these temperatures (see Sec. 4). The method of calculating the experimental and calculated values of Λ shown in Fig. 2 is described at the end of this section.

The data in the table confirm experimentally the Gaussian character (6) of P(t). The table lists the

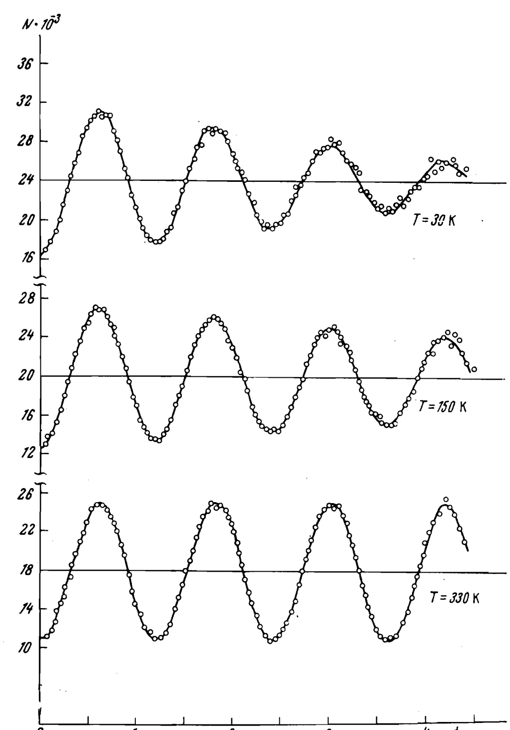

FIG. 1. Precession of μ^+-meson spin in single-crystal copper in a transverse magnetic field B = 62 Oe at three values of the temperature T. The width of the time-analyzer channel is 40 nsec. The smooth curves are plots of (4) with the parameters N_0, a, ω, and τ chosen by the maximum-likelihood method. The presented data have been corrected for the exponent of the μ^+-meson decay $[\exp(-\Delta_0 t)]$.

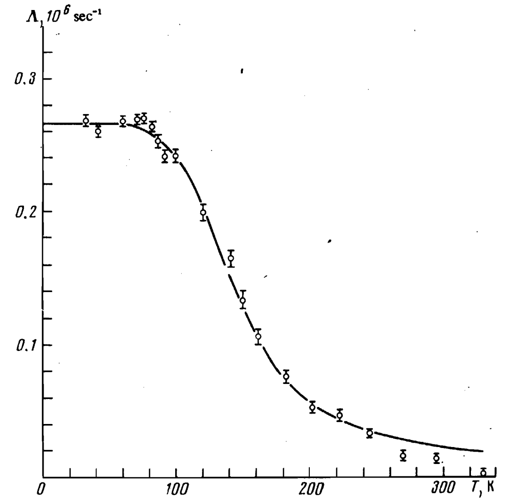

FIG. 2. Temperature dependence of the damping rate of the μ^+-meson precession rate Λ in single-crystal copper. The smooth curve is the calculated function $\Lambda(T)$ obtained under the assumption that the time τ, which determines the damping rate, depends on the temperature in accordance with (2) with the parameters ν_μ and Q_μ given in (11).

T, K	χ^2 $P(t)=e^{-\sigma^2t^2}$	χ^2 $P(t)=e^{-\lambda t}$	T, K	χ^2 $P(t)=e^{-\sigma^2t^2}$	χ^2 $P(t)=e^{-\lambda t}$
33	299	408	82	259	368
42	242	382	86	276	364
60	243	415	91	266	324
71	221	342	100	275	334
76	253	399	120	278	309

values of the Pearson parameter χ^2, obtained from a least-squares comparison of the experimental spectra $(dN/dt)_{exp}$ in single-crystal copper with expression (4), where the function P(t) is given by

$$P(t)=e^{-\sigma^2t^2} \text{ or } P(t)=e^{-\lambda t}. \tag{8}$$

The number of degrees of freedom is $\overline{\chi}^2 = 246$.

An experimental observation of the plateau of the function $\Lambda(T)$ at low temperatures makes it possible to determine the parameter σ in (6), which describes the behavior of P(t) in the absence of diffusion, i.e., as $T \to 0$. It follows from Fig. 2 that for single-crystal copper we can assume σ, with good accuracy, to be equal to

$$\sigma^{Cu}_{sing}=0.266\cdot10^6 \text{ sec}^{-1}. \tag{9}$$

Similarly, the experimental plot of $\Lambda(T \to o)$ (see Fig. 3) was used to determine the parameter σ for a polycrystalline copper sample:

$$\sigma^{Cu}_{poly}=0.252\cdot10^6 \text{ sec}^{-1}. \tag{10}$$

The obtained values (9) and (10) for the parameter σ enable us to use Eq. (5) to determine the time τ. The values of τ for single-crystal and polycrystalline copper samples at various temperatures were determined, as indicated in Sec. 2, by least-squares comparison of the experimental spectra $(dN/dt)_{exp}$ with Eq. (4), where P(t) was determined by (5). Examples of such a comparison are shown in Fig. 1, which demonstrates the good agreement between them.

The temperature dependence of the obtained values of τ in units of $1/\tau = f(1/T)$ is shown in Figs. 4 and 5. It follows from these figures that the experimental

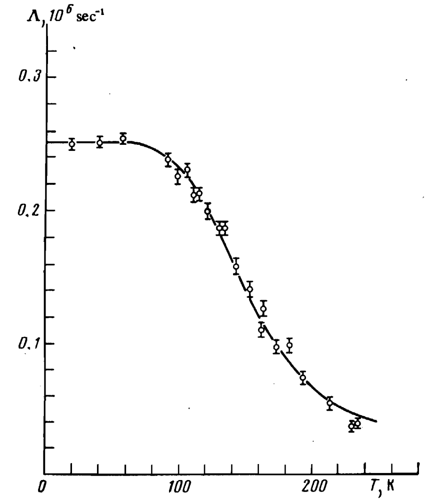

FIG. 3. Temperature dependence of the damping rate of the μ^+-meson precession amplitude in polycrystalline copper. The smooth curve is the calculated function obtained in analogy with the similar curve of Fig. 2.

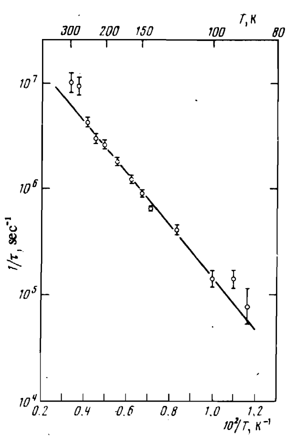

FIG. 4

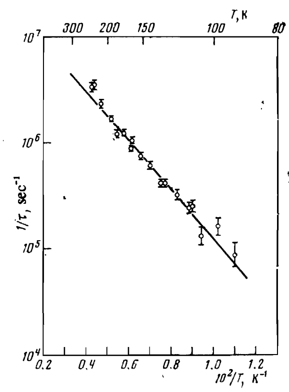

FIG. 5

FIG. 4. Temperature dependence of the time τ in single-crystal copper, in units of $1/\tau = f(1/T)$. The straight line is a plot of (2) $1/\tau = \nu_\mu \exp\{-Q_\mu/T\}$ with parameters ν_μ and Q_μ chosen by the maximum-likelihood method.

FIG. 5. Plot of $1/\tau = f(1/T)$ for polycrystalline copper sample. The straight line is drawn in the same manner as in Fig. 4.

$1/\tau = f(1/T)$ can be set in correspondence with the function (2) $1/\tau = \nu_\mu \exp\{-Q_\mu/T\}$ with the parameters

$$\nu_\mu=10^{(7.61\pm0.04)} \text{ sec}^{-1}, \quad Q_\mu=(562\pm17) \text{ K} \tag{11}$$

for the single-crystal sample and

$$\nu_\mu=10^{(7.46\pm0.04)} \text{ sec}^{-1}, \quad Q_\mu=(551\pm15) \text{ K} \tag{12}$$

for the polycrystalline sample of copper. These values of the parameters ν_μ and Q_μ, as indicated in Sec. 1, contradict above-barrier diffusion of the μ^+-meson in copper and can be explained if it is assumed that the μ^+-meson diffusion proceeds via tunneling under the barrier.

The comparison of the relation (2) with experiment is also illustrated in Figs. 2 and 3, which show the experimental and calculated values of the relaxation rate $\Lambda = 1/t_e$ as functions of the temperature (t_e is the time required for the muon precession amplitude to decrease by a factor e). The values of t_e were calculated in the following manner: For the experimental points at T

> 90°K, when the parameter τ could be determined from experiment, we obtained the time t_e from the relation

$$P(t_e)=e^{-1}, \tag{13}$$

where P(t) is given by (5). For the experimental points at T < 90°K, i.e., on the plateau of the $\Lambda(T)$ plot, the function P(t) in (4) was assumed in the form (6): $P(t) = e\exp(-\sigma^2t^2)$ and $t_e = 1/\sigma$. For the calculated (smooth) curves on Figs. 2 and 3, the time t_e was determined from relation (13), and the parameter $\tau(T)$ in formula (5) for P(t) was calculated using formula (2) at the chosen values of ν_μ and Q_μ (see (11) and (12)). It follows from Figs. 2 and 3 that the saturation of the experimental plot of $\Lambda(T)$ is also in good agreement with the relation (2) $1/\tau = \nu_\mu \exp(-Q_\mu/T)$.

4. DISCUSSION

As indicated in Sec. 1, the experimental plots of $1/\tau = f(1/T)$ shown in Figs. 4 and 5 are attributed to under-barrier diffusion of the μ^+-meson. Let us obtain the resultant barrier transparency coefficient for the μ^+-meson in the copper lattice:

$$\beta=\exp\{-2\hbar^{-1}(2mU)^{1/2}b\}. \tag{14}$$

From a comparison of the experimental plot of $1/\tau = f(1/T)$ and expression (3) it follows that

$$\nu_0\beta=\nu_\mu=10^{7.5}\ \text{sec}^{-1}.$$

We have assumed here $\nu_\mu = 10^{7.5}$ sec^{-1}, which is the average for the single-crystal (11) and polycrystalline (12) copper samples. Putting $\nu_0 = 10^{13}$ sec^{-1}, we obtain $\beta = 10^{-5.5}$.

Let us compare the obtained value of β with the expected value of the potential-barrier transparency coefficient for μ^+-mesons in copper. To this end we determine from the experimental value $\beta = 10^{-5.5}$ and formula (14) the width b of the barrier, putting U = 4000°K:[2)]

$$b=-\ln\beta\cdot\hbar/2(2mU)^{1/2}=1.5\cdot10^{-8}\ \text{cm}. \tag{15}$$

This value of b agrees fully with the possible width of the barrier in the copper lattice (face-centered cube, lattice parameter $d = 3.6 \times 10^{-8}$ cm), and thus confirms the considered model of the below-barrier diffusion of the μ^+-mesons.

The below-barrier diffusion of the μ^+ meson in copper, which predominates in the considered temperature interval, does not exclude, of course, the possibility that some contribution is made by above-barrier diffusion. Let us estimate this contribution. The value of $1/\tau$ for the above-barrier (a.b.) diffusion of the μ^+-meson is written in the form

$$(1/\tau)_{\text{a.b.}}=\nu_0e^{-U/T}=10^{13}e^{-4000/T}\ \text{sec}^{-1}. \tag{16}$$

Here, just as the calculation of b in (15), we assumed $\nu_0 = 10^{13}$ sec^{-1} and U = 4000°K. The values of $\tau_{\text{a.b.}}$ obtained from (16) and a comparison with the time $\tau_{\text{b.b.}}$ for the below-barrier diffusion in copper at various temperatures

$$\left(\frac{1}{\tau}\right)_{\text{b.b.}}=\nu_\mu\exp\left(-\frac{Q_\mu}{T}\right)=10^{7.5}e^{-560/T} \tag{17}$$

are given below:

T, K:	180	200	230	270
$\tau_{\text{b.b.}}/\tau_{\text{a.b.}}$	10^{-3}	10^{-2}	10^{-1}	1

It follows therefore that the above-barrier diffusion of μ^+-meson in copper becomes appreciable only at $T \gtrsim 250$°K, i.e., at the very boundary of the considered temperature interval. Therefore allowance for the above-barrier diffusion hardly changes the presented values (11) and (12) of the parameters ν_μ and Q_μ, which characterize the below-barrier diffusion of the μ^+-meson.

Allowance for the above-barrier diffusion explains the difference, seen in Figs. 2 and 4, between the experimental and calculated plots of $\Lambda(T)$ and $\tau(T)$ at $T \gtrsim 270$°K. Indeed, the ratio $\tau_{\text{a.b.}}/\tau_{\text{b.b.}} \sim 1$, obtained at these temperatures, leads to an appreciable decrease of the observed time τ:

$$1/\tau=1/\tau_{\text{b.b.}}+1/\tau_{\text{a.b.}} \tag{18}$$

and by the same token, according to (7), to an appreciable decrease of the damping rate Λ. Unfortunately, we cannot use relation (18) to refute quantitatively the presence of a contribution of above-barrier diffusion at high temperatures, since the parameters ν_0 and U in (16) are not known exactly for the μ^+-meson in copper.

We note, finally, the work by Chapman and Seymour,[12] who investigated the temperature dependence of the rate of spin relaxation $\Lambda_{Cu} = 0.016 \times 10^6$ sec^{-1} and remains constant in the temperature interval T = 20 to 290°K. The obtained value of Λ_{Cu} is attributed to dipole relaxation of the spin of the copper nuclei, and the independence of the relaxation rate of the temperature means the absence of self-diffusion of the copper atoms at T = 20–290°K. The result of [12] shows thus that the observed temperature dependence of the μ^+-meson spin relaxation rate in copper is due only to μ^+-meson diffusion.

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1)Preliminary results on the diffusion of the positive muon in copper were published elsewhere. [1]

2)We recall that the height of the potential barrier for the proton in copper is Q_p = 4600°K. [5-8] The potential barrier for the μ^+ meson should be somewhat lower, since the energy of the ground state of the μ^+ meson is three times higher than the energy of the ground state of the proton. An estimate shows, however, that on the basis of the indicated value of Q_p we can assume for the μ^+ meson in copper that U = 4000°K.

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