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Relaxation of μ^+ -meson spin in the crystal lattice of copper, vanadium, or niobium in weak magnetic fields

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We compare the experimental and calculated values of the relaxation rates Λ of the spin of a non-diffusing μ^+ meson in copper, vanadium, and niobium in the absence of an external magnetic field. The enlargement of the interstitial pores of the crystal lattices of these metals by the localization of the μ^+ meson in the pores is estimated. The $\Lambda(B)$ dependence is investigated for weak transverse magnetic fields B .

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1. INTRODUCTION

The spin of a μ^+ meson in a crystal lattice relaxes because of the magnetic dipole interactions with the magnetic moments of the surrounding nuclei.¹ Measurement of the relaxation rate Λ of the spin of the μ^+ meson makes it possible to determine the type of the interstitial pore in which the μ^+ meson is localized, and also find the deformation of this pore. The determination type of the pore and the degree of its deformation are determined by comparing the experimental and calculated values of Λ . This method is quite sensitive, since $\Lambda \sim r_i^{-3}$, where r_i is the distance between the μ^+ meson and the nuclei of the neighboring atoms of the metal. This comparison is possible only in the limiting cases of strong and weak (zero) external magnetic fields B , since the calculation of the values of Λ for an arbitrary field B entails very great difficulties.

The values of Λ should be measured at sufficiently low temperature, when the μ^+ meson hardly diffuses in the crystal. The diffusion of the μ^+ meson causes the local dipole magnetic field at the meson to become alternating in time, as a result of which the relaxation rate Λ decreases. In the present study we have measured the relaxation rate of the spin of a non-diffusing μ^+ meson in a crystal lattice of copper, vanadium, or niobium in

weak transverse magnetic fields B and at $B=0$. The measurement of the relaxation rate Λ at $B=0$ is in certain respects more convenient than at $B \rightarrow \infty$, inasmuch as to obtain in practice the asymptotic values of $\Lambda(B \rightarrow \infty)$ in these metals it is necessary to produce very strong magnetic fields. The magnetic fields B needed to measure the values of $\Lambda(B \rightarrow \infty)$ must lead to a practically complete suppression of the influence of the electric quadrupole interactions of the μ^+ meson with the neighboring metal-atom nuclei. Camani *et al.*² have shown experimentally that in copper this requires a field $B \sim 10$ kOe. Allowance for the quadrupole interactions at $B=0$, in the case of sufficiently large quadrupole moments Q of the nuclei, reduces to simple corrections to the calculated values of $\Lambda(B=0)$, and when these are calculated one can assume the limiting value $Q \rightarrow \infty$.

2. THEORY

The calculated value Λ_{calc} of the relaxation rate of the spin of a μ^+ meson in a metal is determined from the dependence of the polarization $P(t)$ of the μ^+ meson on the time. In the calculation of the $P(t)$ we represent this function as a series in even powers of t :

$$P(t) = 1 - \frac{1}{2}t^2 M_2 + \frac{1}{4}t^4 M_4 - \dots \quad (1)$$

There are no odd powers of t in the expression (1), in-

asmuch as $P(t)$ is an even function for a nondiffusing μ^+ meson. We confine ourselves hereafter to the calculation of only the coefficient M_2 , called the second moment in the theory of nuclear magnetic resonance. This procedure is justified in practice, since the experimental function $P(t)$ is well described by the expression $P(t) = \exp(-\Lambda^2 t^2)$ in the entire considered interval Δt , and consequently makes it possible to measure reliably the experimental value of the second moment $M_2 = 2\Lambda^2$ in the expansion (1).

The calculated value of M_2 is determined from the Hamiltonian where

$$H = \sum_i g_i(I_i, \sigma) + \sum_{i,k} G_{ik}(I_i, I_k) - \xi \sigma - \sum_i \beta_i I_i, \quad (2)$$

the first term is the energy of the dipole interaction of the magnetic moments of the μ^+ meson and the nuclei of the medium, and the second is the energy of the magnetic interaction of the nuclei of the medium, while the third and fourth are the Zeeman energies of the magnetic moments of the μ^+ meson and of the nuclei in the external magnetic field. The following notation is used in (2): σ and I_i are the spins of the μ^+ meson and the nuclei of the medium; g_i and G_{ik} are tensor quantities that determine the magnetic dipole interactions of the pair of particles; ξ and β are vector quantities that determine the Zeeman interaction of the particles in an external magnetic field B . Thus, for example, the tensor g_i takes the form

$$g_i = \frac{\gamma_\mu \gamma_I \hbar}{2r_i^3} [\delta_{\alpha\beta} - 3n_\alpha n_\beta]. \quad (3)$$

Here γ_μ and γ_I are the gyromagnetic ratios for the μ^+ meson and nuclei of the medium; r_i is the radius vector from the μ^+ meson to the i -th nucleus; n_α are the direction cosines of the radius vector r_i .

The standard procedure for calculations with the Hamiltonian (2) at $B=0$ leads to the following expression for the second moment M_2 in a polycrystalline sample, i.e., after averaging over all the orientations of the crystal:

$$M_2 = \frac{4}{3} \gamma_\mu^2 \gamma_I^2 \hbar^2 I(I+1) \sum_i r_i^{-6}, \quad B=0. \quad (4)$$

Similar expressions for M_2 in a weak ($B \rightarrow 0$) magnetic field was obtained by Didyk and Yushankhaï.³ Analogous calculations lead to an expression for M_2 in a polycrystalline sample for the case $B \rightarrow \infty$ (Ref. 4), in the form

$$M_2 = \frac{4}{15} \gamma_\mu^2 \gamma_I^2 \hbar^2 I(I+1) \sum_i \frac{1}{r_i^6}, \quad B \rightarrow \infty. \quad (5)$$

It follows from (4) and (5) that

$$M_2(B=0) = 5M_2(B \rightarrow \infty). \quad (6)$$

It is interesting to note that expression (4) for the second moment M_2 at $B=0$ does not depend on G_{ik} , i.e., on the magnetic interactions of the nuclei with one another. The dipole interactions of the nuclear magnetic moments are present only in the expressions for the fourth and succeeding moments in the expansion (1) of $P(t)$. In this case, however, this property of the Hamiltonian (2) is of no practical significance, since the

frequencies connected with the nuclear dipole interactions are too small to exert a noticeable influence on the relaxation of the spin of the μ^+ meson in the metal.

Expression (4) was obtained without allowance for the electric quadrupole interactions, which, as shown by Camani *et al.*,² greatly influence the value of M_2 . The influence of the quadrupole interaction on the relaxation of the μ^+ meson spin in a medium manifests qualitatively in the fact that only the radial components of the dipole magnetic fields at the μ^+ meson remain effective. This means that in the expression for the energy of the dipole interaction of

$$\sum_i g_i I_i \sigma$$

the vectors I_i must be replaced by their radial components $(I_i)_r$, i.e., the tensor t_i [Eq. (3)] must be rewritten in the form

$$g_i = \frac{\gamma_\mu \gamma_I \hbar}{2r_i^3} (-2n_\alpha n_\beta). \quad (3')$$

In a rigorous quantum-mechanical analysis these classical arguments are valid only for integer values of the spin I , and, of course, in the classical limit $I \gg 1$. For half-integer nuclear spins the quadrupole interactions do not lead to a total annihilation of the transverse (with respect to r_i) components of the dipole magnetic field at the μ^+ meson. As a result, the following exact expressions are obtained for the second moment M_2 at $B=0$ in the limiting case $Q \rightarrow \infty$:

$$M_2(Q \rightarrow \infty) = \frac{4}{3} \gamma_\mu^2 \gamma_I^2 \hbar^2 I(I+1) \frac{2}{3} \sum_i \frac{1}{r_i^6} \quad (7)$$

for integer values of I and

$$M_2(Q \rightarrow \infty) = \frac{4}{3} \gamma_\mu^2 \gamma_I^2 \hbar^2 I(I+1) \left[\frac{2}{3} + \frac{1}{4I(I+1)} \right] \sum_i \frac{1}{r_i^6} \quad (8)$$

for half-integer values of I . Expression (8) for M_2 goes over at $I \gg 1$, as it should, into formula (2), and goes over into formula (4) at $I=1/2$, when there are no quadrupole interactions.

Expressions (7) and (8) for M_2 are the limiting ones for $Q \rightarrow \infty$, consequently correspond to the maximum possible decrease of the second moment on account of the quadrupole interactions. The calculation of the $M_2(Q)$ in the general case entails very serious difficulties, since it is necessary to take into account the deformation of the internal electron shells of the atom under the influence of the electric field of the μ^+ meson. It must be noted, however, that for nuclei with relatively large values of Q , the limiting formula (7) and (8) are a good practical approximation, for in this case the energy of the quadrupole interactions exceeds substantially the energy of the magnetic dipole interactions, which is determined by the Hamiltonian (2) at $B=0$.

We consider now the $M_2(B)$ dependence in weak external fields B . The exact expression for this dependence is very difficult to derive. It can be stated, however, that

$$M_2(B=0) = 2M_2(B \gg B_{\text{dip}}), \quad (9)$$

where $B_{\text{dip}} \approx 10$ Oe is the dipole magnetic field at the

μ^+ meson, produced by the magnetic moments of the nuclei of the metal. Relation (9) can be obtained from the Hamiltonian (2),³ as well as from the following simple geometrical considerations. At $B=0$ the relaxation of the μ^+ -meson spin is determined by two projections of the magnetic field B_{d1p} that are transverse to the μ^+ -meson spin. In an external transverse magnetic field $B \geq B_{d1p}$ the relaxation of the μ^+ -meson spin is influenced in practice only by one projection of the field B_{d1p} , namely the one which is collinear with the external field B . It is this which decreases the value of M_2 by one half at $B \geq B_{d1p}$.

3. EXPERIMENT AND RESULTS

The relaxation rates Λ of the μ^+ -meson spin in weak transverse fields B and at $B=0$ in copper, vanadium, in niobium were measured with the JINR synchrocyclotron in Dubna. The value of Λ in a transverse magnetic field B was determined from a comparison, by the maximum-likelihood method, of the experimental precession curve with the corresponding theoretical relation

$$N_1(t) = N_0 e^{-t/\tau} (1 - C e^{-\Lambda^2 t^2} \cos \omega t). \quad (10)$$

Here $\tau = 2.2 \times 10^{-6}$ sec is the lifetime of the μ^+ meson, C is the experimental asymmetry coefficient of the angular distribution of the positrons from the $\mu^+ \rightarrow e^+$ decay, and $\omega = eB/mc$ is the frequency of the Larmor precession of the μ^+ meson in the field B . At $B=0$ the value of Λ was determined by comparing the experimental time spectrum $N_{exp}(t)$ with the theoretical expression

$$N_2(t) = N_0 e^{-t/\tau} (1 - C e^{-\Lambda^2 t^2}), \quad (11)$$

which differs from (10) only in the absence of the oscillating factors $\cos \omega t$, which is due to the precession of the μ^+ -meson spin in the external field B . A detailed description of the corresponding experimental apparatus is given in Ref. 5.

In expressions (10) and (11), the function (1) has the Gaussian form $P(t) = \exp(-\Lambda^2 t^2)$. As noted in Sec. 2, it is precisely this form of $P(t)$ which corresponds to a low sample temperature, i.e., to a nondiffusing μ^+

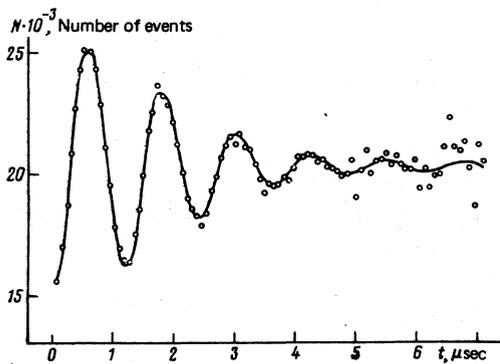


FIG. 1. Precession of the μ^+ -meson spin in vanadium $T=18$ K and $B=70$ Oe. The width of the time-analyzer channel is $\delta t = 80$ nsec. The solid curve is the theoretical relation (10). The experimental and theoretical functions $N(t)$ were corrected for the exponential character of the decay of the μ^+ meson.

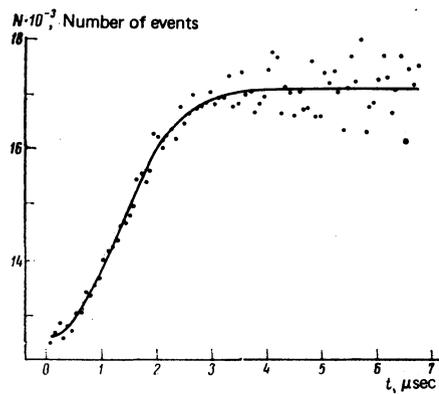


FIG. 2. Relaxation of the μ^+ meson spin in vanadium in $T=18$ K and $B=0$. The width of the time-analyzer channel is $\delta t = 80$ nsec. The solid curve is the theoretical function (11). The experimental and theoretical $N(t)$ plots are corrected for the experimental μ^+ -meson decay.

meson. Figures 1 and 2 show, by way of illustration, plots of $N(t)$ in vanadium at $T=18$ K for $B=70$ Oe and $B=0$. It is seen from these figures that the theoretical expressions (10) and (11), which correspond to a Gaussian $P(T)$, describe well the corresponding experimental spectra $N_{exp}(t)$.

The interval of the temperatures at which the diffusion of the μ^+ meson stops is determined from the experimental $\Lambda(T)$ relations. These are shown in Fig. 3 for the investigated metals, and it is seen from them that at sufficiently low temperatures (T lower than 60, 35, and 40 K for copper, vanadium, and niobium, respectively) the values of Λ are maximal and do not depend on the temperature. At these temperatures, the μ^+ meson can be regarded in practice as nondiffusing, i.e., localized in one crystal cell of the metal.² The decrease of Λ with increasing temperature is due to the diffusion of the μ^+ meson when the magnetic dipole fields at the meson become alternating in time. All the subsequent measurements of Λ in the investigated metals pertain only to the indicated region of low temperatures, when the μ^+ meson can be regarded as localized in one crystal cell during the entire observation time.

Table I illustrates the experimental reliability of the Gaussian relation (1) $P(t) = \exp(-\Lambda^2 t^2)$ at low temperatures for vanadium and niobium. Analogous data for copper are given in Ref. 6. Table I shows the values of the Pearson parameter χ^2 , obtained by comparing the experimental $N_{exp}(t)$ relations in a field $B=76$ Oe with expression (10), in which $P(t)$ was assumed to be Gaussian [$P(t) = \exp(-\Lambda^2 t^2)$] or exponential [$P(t) = \exp(-\lambda t)$].

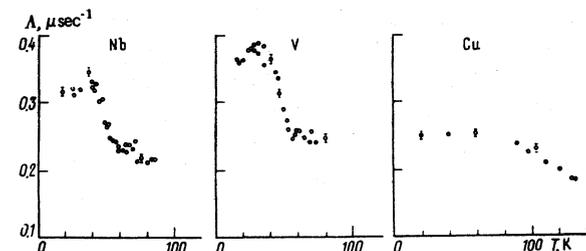


FIG. 3. Experimental plots of $\Lambda(T)$ at $B=70$ Oe for niobium, vanadium, and copper.

TABLE I. Values of the Pearson parameter χ^2 for Gaussian and experimental $P(t)$ and of the relaxation rate Λ for two Δt intervals (number of degrees of freedom $\chi^2=180$).

Metal	T, K	χ^2 ($\Delta t=7 \mu\text{sec}$)		Λ , μsec^{-1}	
		$P = e^{-\Lambda^2 t^2/2}$	$P = e^{-\Lambda t}$	$\Delta t=2 \mu\text{sec}$	$\Delta t=7 \mu\text{sec}$
V	15	188	219	0.40±0.02	0.363±0.009
	17	162	300	0.36±0.01	0.359±0.006
	19	165	218	0.37±0.02	0.361±0.009
	24	168	299	0.36±0.01	0.376±0.007
	25	175	358	0.37±0.01	0.378±0.007
	27	181	272	0.39±0.01	0.376±0.007
	28	195	321	0.39±0.01	0.386±0.007
Nb	17	163	255	0.34±0.01	0.317±0.005
	24	184	260	0.34±0.02	0.321±0.007
	25	200	316	0.31±0.01	0.312±0.006
	30	163	295	0.32±0.02	0.320±0.007
	35	182	296	0.34±0.02	0.347±0.007

The table lists also the values of Λ for $P(t) = \exp(-\Lambda^2 t^2)$ obtained by comparing the experimental and theoretical plots of $N(t)$ for small ($\Delta t = 2 \mu\text{sec}$) and large ($\Delta t = 7 \mu\text{sec}$) time intervals.

It is seen from Table I that the parameter χ^2 is much smaller for a Gaussian $P(t)$ than for an exponential one, and agrees with its mean value $\chi^2 = 180$. The agreement between the values of Λ for $\Delta t = 2 \mu\text{sec}$ and $\Delta t = 7 \mu\text{sec}$ also confirms the Gaussian form of the function $P(t)$.

We proceed now to a description of the results of the measurement of the relaxation rate of the spin of a non-diffusing μ^+ meson in a weak magnetic field B and as $B \rightarrow 0$. Table II lists the values of Λ for copper, vanadium, and niobium measured in transverse magnetic fields $B \rightarrow 0$. It is seen from Table II, in particular, that the fields $B = 0.1$ and $B = 0.25$ Oe correspond to the same values of Λ within the limits of errors (measurements at the same temperature were made for copper and vanadium). It follows therefore that at these values of B the limit $B \rightarrow 0$ has been reached in practice.

The experimental plots of $\Lambda(B)$ for copper, vanadium, and niobium are shown in Fig. 4. It is seen from the figure that for all these metals the value of Λ increases sharply in the region of weak fields B . This is particularly clearly seen for copper and vanadium, whose $\Lambda(B)$ was measured in sufficient detail, and it can be concluded that an abrupt increase of Λ takes place in fields $B \lesssim 15$ Oe. The quantitative relation $\Lambda(B=0) \approx 2^{1/2} \Lambda(B=15)$ that follows from Fig. 4 confirms formula (9) (see also Table IV). At the same time it is seen from Fig. 4 that at $B = 15-300$ Oe the experimental plots of $\Lambda(B)$

TABLE II. Relaxation rate Λ of μ^+ -meson spin as $B \rightarrow 0$ (the errors are statistical).

Metal	T, K	Λ , μsec^{-1}
V	21	0.1
	21	0.25
	47	0.2
	47	0.3
Average:		0.372±0.004
Cu	18	0.1
	18	0.25
	29	0.3
Average:		0.600±0.008
Nb	17	0.1
	29	0.3
Average:		0.509±0.008

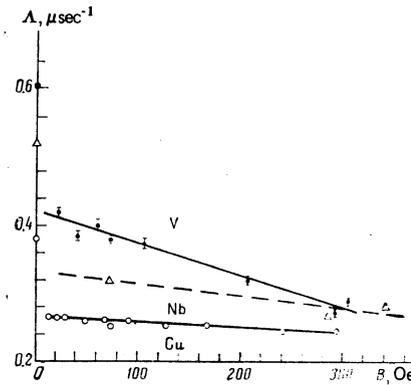


FIG. 4. Experimental plots of $\Lambda(B)$ for niobium, vanadium, and copper at the temperatures $T_{\text{Nb}} = 25$ K, $T_{\text{V}} = 30$ K, $T_{\text{Cu}} = 40$ K. The statistical errors $\delta\Lambda$ for niobium and copper do not exceed the dimensions of the experimental points. The straight plots of $\Lambda(B)$ at $B > 10$ Oe were drawn for the sake of clarity.

of vanadium and copper are substantially different. This difference can be attributed to the strong quadrupole interaction of the μ^+ meson in copper,² which leads in fact to the weak $\Lambda(B)$ dependence in this metal. The nuclear quadrupole moments for all the investigated metals are given in Table III.

4. DISCUSSION

Table III gives the calculated and experimental values of Λ for copper, vanadium, and niobium. The values Λ_{calc} were calculated for the limiting cases $Q \rightarrow 0$ and $Q \rightarrow \infty$, and for two types of pores of the undeformed metal lattice—octahedral and tetrahedral. At $B = 0$, we calculated Λ_{calc} from formulas (4) and (8). The values of Λ_{calc} given in Table III for $B = 15$ Oe were determined from the corresponding values of Λ_{calc} ($B = 0$) in accordance with the relation (9). The experimental relaxation rates Λ_{exp} are given in Table II for $B = 0$ and were determined for $B = 15$ Oe from Fig. 4 by extrapolating the corresponding experimental $\Lambda(B)$. Since $\Lambda(B)$ was not measured in sufficient detail for niobium, the value of $\Lambda(B = 15)$ for this metal was determined only approximately.

Table IV shows a comparison of the calculated and experimental values of $\Lambda(B = 0)$, which are given in

TABLE III. Calculated Λ_{calc} and experimental Λ_{exp} values of the relaxation rate of the spin of the μ^+ meson in Cu, V, and Nb.

Metal	a , Å	μ_I , μ_B	Q, b	B, Oe	Λ_{calc} , μsec^{-1}			Λ_{exp} , μsec^{-1}
					Calculation variant	tetrapore	octapore	
Cu	3.61	2.27	0.21 [7]	0	$Q \rightarrow 0$	0.564	0.451	0.372±0.004
				15	$Q \rightarrow \infty$	0.483	0.386	
V	3.03	5.15	0.05 [9]	0	$Q \rightarrow 0$	0.399	0.319	0.600±0.008
				15	$Q \rightarrow \infty$	0.342	0.273	
Nb	3.29	6.17	0.32 [9]	0	$Q \rightarrow 0$	0.892	0.960	0.509±0.008
				15	$Q \rightarrow \infty$	0.737	0.793	
Nb	3.29	6.17	0.32 [9]	0	$Q \rightarrow 0$	0.631	0.678	0.414±0.006
				15	$Q \rightarrow \infty$	0.521	0.561	
Nb	3.29	6.17	0.32 [9]	0	$Q \rightarrow 0$	0.813	0.875	0.509±0.008
				15	$Q \rightarrow \infty$	0.669	0.720	
Nb	3.29	6.17	0.32 [9]	0	$Q \rightarrow 0$	0.575	0.619	0.33
				15	$Q \rightarrow \infty$	0.473	0.509	

Note. a —lattice parameter, μ_I and Q —magnetic dipole and electric quadrupole moments of the nuclei.

TABLE IV. Ratios $\alpha = \Lambda_{\text{calc}}(B=0)/\Lambda_{\text{exp}}(B=0)$ and $A = [\Lambda_{\text{exp}}(B=0)/\Lambda_{\text{exp}}(B=15)]^2$ for copper, vanadium, and niobium.

Metal	Type of lattice	Pore	α	A
Cu	fcc	octahedral	1.04±0.01	1.99±0.04
V	bcc	tetrahedral	1.23±0.01	2.10±0.10
Nb	bcc	tetrahedral	1.31±0.02	-

Table III, and also the experimental ratio $\Lambda(B=0)/\Lambda(B=15)$, which illustrates the validity of formula (9). In comparison of the calculated and experimental values of Λ in Table IV, it was assumed that in a metal with an fcc lattice the μ^* meson is localized in an octapore, while in a metal with a bcc lattice it is localized in a tetrapore. The localization of μ^* mesons in octapores of an fcc copper lattice was determined experimentally.² The localization of μ^* mesons in the tetrapores of bcc lattices of vanadium and niobium was assumed in analogy with the localization of an impurity hydrogen atom.¹⁰

It is seen from Table IV that $\alpha > 1$. This ratio increases even more if we assume that the μ^* meson is localized in other pores of the crystal or that the limiting case $Q \rightarrow \infty$, used to calculate $\Lambda_{\text{calc}}(B=0)$, is not realized. In other words, the ratios α given in Table IV are the smallest possible.

Experimental values $\alpha > 1$ are natural, since a μ^* meson localized in an interstitial pore enlarges the pore, leading to an increase of the distances r_i in expressions (4) and (8) for Λ_{calc} . The quantity α is obviously the relative volume expansion of the pore, primarily of its first coordination sphere, since this sphere, i.e., the metal nuclei closest to the μ^* meson, makes more than 90% of the contribution to the Λ_{calc} .

Because of this large difference between the values of α for copper ($\alpha_{\text{Cu}} = 1.04$) and for vanadium or niobium ($\alpha_{\text{V}} = 1.23$, $\alpha_{\text{Nb}} = 1.31$) is not clear. A possible explanation is the assumption that in vanadium and niobium the

μ^* meson is trapped in somewhat larger pores.

It follows also from Table IV that the experimental ratio A for copper and vanadium agrees fully with the theoretical value (9) $A = 2$. This agreement affords an additional possibility of determining the volume expansion α of the pore from a comparison of the experimental and calculated values of $\Lambda(B \approx B_{\text{dip}})$. The experimental $\Lambda(B \approx B_{\text{dip}})$ is more accurate the larger the quadrupole interaction of the μ^* meson with the metal nuclei, since the function $\Lambda(B)$ is almost constant in the case of a strong quadrupole interaction (see Fig. 4).

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