Possibility of identifying the regions of localization of positive muons in crystals

A. G. Lundin, O. V. Falaleev, and N. A. Sergeev

L. V. Kirenskiĭ Institute of Physics, Siberian Division, USSR Academy of Sciences
(Submitted December 1, 1976)
Pis'ma Zh. Eksp. Teor. Fiz. **25**, No. 2, 103–106 (20 January 1977)

With copper crystals as an example, we consider the possibility of using the method of moments to determine the regions where positive muons are localized in crystals.

PACS numbers: 76.90.+d

A new method of investigating the physical properties of a solid with the aid of polarized positive muons is being successfully developed of late.[1,2] The method is based on experimentally investigating the precession of the magnetic moment of positive muons that are initially polarized perpendicular to the external magnetic field. The damping of the precession is due to the interaction of the positive muon with the magnetic moments of the crystal nuclei. As noted in[1,2], the shape of the decay curve of the free precession of the positive muon, $P(t)$, depends on the location of the muon in the crystal lattice and on th character of the mobility.

We propose below to determine the region of localization of the positive muons in crystals by using the method of moments, which is extensively employed in solid-state microwave spectroscopy.

The Fourier transform of $P(t)$ is the distribution function of the local magnetic fields at the positive muons, and is in essence the line-shape function $g(\omega)$ of the magnetic resonance of the positive muons.[3] In the general case $P(t)$ seems to have a complicated form, but at sufficiently short times it can be approximated by the series

$$P(t) = P_0\left(1 - \frac{M_2}{2!}t^2 + \frac{M_4}{4!}t^4 - \ldots\right),$$

where

$$M_{2n} = \int_{-\infty}^{\infty} g(\omega)(\omega - \omega_0)^{2n}\,d\omega \Big/ \int_{-\infty}^{\infty} g(\omega)\,d\omega = \frac{(-1)^n}{P_0}\left(\frac{\partial^{2n}P(t)}{\partial t^{2n}}\right)_{t=0}$$

are the moments of the line shape $g(\omega)$.

In contrast to $P(t)$, the moments can be calculated exactly, and for magnetic dipole-dipole interactions they are relatively simply connected with the coordinates of the positive muons in the unit cell of the crystal.

By way of example, we consider the experimental damping rate of the spin-precession amplitude of positive muons in the crystal lattice of copper.[4] At low temperatures, when there is no diffusion of the positive muons the precession damping rate $P(t)$ does not depend on temperature and is well described by the Gaussian curve

$$P(t) = \exp(-\sigma^2 t^2).$$

In this case the experimentally measured parameter σ determines uniquely the entire $P(t)$ curve and is connected with the second moment in the following manner:

$$M_2 = 2\sigma^2.$$

In the case of polycrystalline samples, as shown by Van Vleck, the value of the second moment is determined by the expression[3]:

$$M_2 = \frac{4}{15}\gamma_\mu^2\gamma^2\hbar^2 S(S+1)\sum_i^{(R_i < R_0)} R_i^{-6}. \qquad (1)$$

In this formula, γ_μ is the muon gyromagnetic ratio, $\gamma = (f_1\gamma_1^2 + f_2\gamma_2^2)^{1/2}$, where γ_1, γ_2, and f_1, f_2 are the gyromagnetic ratios and the relative concentrations of the copper isotopes, $\hbar$ is Planck's constant, S is the spin of the copper nucleus, and R_i is the distance between the muon and the ith copper nucleus. The summation in (1) is over all the nuclei inside a sphere of radius R_0. For cubic lattices, an approximate accuracy of 0.1% in the calculation of ΣR_i^{-6} is ensured when R_0 exceeds the unit-cell parameter a by approximately five times. For copper at temperatures 20—60 °K, at which the employed experimental results were obtained, the parameter a will be assumed to be 3.594 Å, taking into account the temperature correction, which is taken from[6]. The contributions made to (1) by all the remaining magnetic moments in the crystal, as well as the corrections for the nuclear vibrations, is negligibly small.

Recognizing that the unit cell of copper is face-centered cubic and contains two types of voids, octahedral and tetrahedral, it is natural to assume that the positive muons can be at the centers of the voids. We assume also that the in-

troduction of the muons does not change the mutual placements of the surrounding copper nuclei.

If the positive muons occupy both possible positions, then the observed second moment is determined by the weighted sum

$$M_2 = p_T M_2^T + p_0 M_2^0,$$

where p_T and p_0 are the probabilities of the capture of the μ^+ mesons by the tetrahedral and octahedral voids, respectively ($p_T + p_0 = 1$); M_2^T and M_2^0 are the values of the second moments calculated for each of these positions.

A computer calculation has shown that

$$M_2^T = 176.1\times10^{48}\frac{\gamma_\mu^2\gamma^2\hbar^2 S(S+1)}{a^6} = 13.2\times10^{10}\ \text{sec}^{-2};$$

$$M_2^0 = 112.6\times10^{48}\frac{\gamma_\mu^2\gamma^2\hbar^2 S(S+1)}{a^6} = 8.5\times10^{10}\ \text{sec}^{-2}.$$

The experimental value of σ obtained in[4] is $(0.252 \pm 0.06)\times10^6$ sec^{-1}. (The random error is estimated from Fig. 3 of that reference). From this we have the following value of the second moment:

$$M_2^e = (12.70 \pm 0.60)\times10^{10}\ \text{sec}^{-2}.$$

Comparison of the theoretical and experimental data demonstrates convincingly that the positive muons in copper crystals occupy predominantly tetrahedral positions. We note that, for example, in a nickel crystal, which has the same structure as copper, the positive muons occupy according to the data of[6] the octahedral voids.

The validity of the initial assumptions concerning the localization of the positive muons at the centers of undistorted voids might be verified by measuring in experiment the second moments of single-crystal samples oriented in a magnetic field in a special manner. We note in this connection that the general properties of the orientation dependence of the moments, for crystals of all syngonies, as well as the procedure of determining the coordinates of magnetic nuclei in crystals on the basis of the theory of the optimal experiment, have been considered by us in[7,8]. In particular, for cubic-symmetry crystals the orientational dependence of M_2 is determined completely by two structure parameters, and to determine them most accurately in experiment it is necessary to measure second moments at two orientations of the crystal in a magnetic field. One of these orientations corresponds to the magnetic field vector directed along a fourfold symmetry axis of the crystal ($H_0 \parallel C_4$), and the other along a threefold axis ($H \parallel C_3$). The corresponding values of the moments, calculated for copper single crystals are

$$H_0 \parallel C_4: \quad M_2^T = 1.24\times10^{10}\ \text{sec}^{-2}; \quad M_2^0 = 19.25\times10^{10}\ \text{sec}^{-2},$$

$$H_0 \parallel C_3: \quad M_2^T = 20.75\times10^{10}\ \text{sec}^{-2}; \quad M_2^0 = 0.95\times10^{10}\ \text{sec}^{-2}.$$

We see that the maximum value of the second moment occurs for the tetrahedral position at the minimum value for the octahedral position and vice versa. This should ensure a rather high accuracy when the theoretical and experimental data are compared.

In conclusion, we are indebted to Dr. Patterson (Physics Institute of the University of Zurich, Switzerland), who called our attention to the interesting use of our procedure to determine the coordinates of spins in crystals by the method of moments.

I. I. Gurevich, A. I. Klimov, V. N. Maĭorov, E. A. Meleshko, B. A. Nikol'skiĭ, V. I. Selivanov, and V. A. Suetin, Zh. Eksp. Teor. Fiz. **69**, 439 (1975) [Sov. Phys. JETP **42**, 222 (1976)].
B. D. Patterson, H. Graf, W. Hofman, and W. Kündig, Abstracts of XIX Congress AMPERE, Heidelberg, 1976, p. 222.
A. A. Abragam, Principles of Nuclear Magnetism, Oxford, 1961.
V. G. Grebinnik, I. I. Gurevich, V. A. Zhukov, A. P. Manych, E. A. Meleshko, I. A. Muratova, B. A. Nikol'skiĭ, V. I. Selivanov, and V. A. Suetin, Zh. Eksp. Teor. Fiz. **68**, 1548 (1975) [Sov. Phys. JETP **41**, 777 (1975)].
M. P. Malkov, I. B. Danilov, A. G. Zel'dovich, and A. B. Fradkov, Spravochnik po fiziko-tekhnicheskim osnovam gluobokogo okhlazhdeniya (Handbook of Physical and Technical Principles of Deep Freezing) Moscow-Leningrad, Gosenergoizdat, 1963, p. 79.
I. I. Gurevich, A. N. Klimov, V. N. Maĭorov, E. A. Meleshko, B. A. Nikol'skiĭ, V. S. Roganov, V. I. Selivanov, and V. A. Suetin, Pis'ma Zh. Eksp. Teor. Fiz. **18**, 564 (1973) [JETP Lett. **18**, 332 (1973)].
O. V. Falaleev, N. A. Sergeev, and A. G. Lundin, Kristallografiya **19**, 560 (1974) [Sov. Phys. Crystallogr. **19**, 347 (1974)].
A. G. Lundin, O. V. Falaleev, and N. A. Sergeev, Abstracts of XIX Congress AMPERE, Heidelberg, 1976, p. 33.