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"SLOW" MUON DEPOLARIZATION IN A SINGLE CRYSTAL OF GYPSUM ($Ca\ SO_4 \cdot 2H_2O$)*

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Polarized muons were stopped in a single crystal of $Ca\ SO_4 \cdot 2H_2O$ placed in a magnetic field. Muon precession displayed a beat behavior and a "slow"-μsec-decrease of the muon spin polarization; these effects were found to be strongly dependent on crystal orientation in the field. Consistent explanation of the results was achieved by assuming that the muon occupied a normal proton site, and by considering the local field effects due to the dipole-dipole interaction of muon-proton pairs.

The depolarization phenomena of polarized positive muons stopping in nonmetallic solids has been the subject of many investigations.[1] This "fast" transverse depolarization occurs in times so short (< 1 nsec) that only the net result has been measured by observing the residual polarization. The dependence on the longitudinal magnetic field of this fast depolarization and rate can be understood by assuming that the muon exists part of the time as muonium during the early phase of its

life in the medium.

Measurements of the precession amplitude of polarized muons stopped in solids indicate that the residual polarization seemed to be constant in time which the exception of one measurement done in B_4C. Swanson[2] observed a decay of the precession amplitude with a time constant of ~ 6.5 μsec. For many years this was the only known example of a "slow" depolarization in a solid in a transverse magnetic field. In 1968 Gurevich et al.[3] found another example of slow decay of muon polarization in sulfur with a time constant of 30 ± 5 nsec. The recent observation of decay of the spin precession signal of polarized positive muons in paramagnetic solutions,[4] which established a close relationship to the relevant relaxation times in proton-NMR experiments, stimulated us to look again for "slow" depolarization in other solids. During the course of our experiment we learned of the measurements by E. V. Minaichev et al.,[5] which showed that indeed muons depolarize slowly in a wide class of solids in transverse magnetic fields. It should be pointed out that this "slow" depolarization is an additional process which occurs subsequent to the above mentioned "fast" depolarization. Attempts[3, 5] have been made to explain the "slow" depolarization in terms of the muonium mechanism of Ivanter and Smilga.[6] The effect of local magnetic field interactions due to the presence of paramagnetic impurities or neighboring nuclei has also been considered.[7] However, no quantitative calculations and comparisons have been performed.

In applying the NMR picture to the "slow" muon depolarization in

solids, one must consider the question of the chemical situation of the stopped muon. The success of interpreting the results in paramagnetic solutions under the assumption that the muon replaces a proton in a water molecule and then using the proton NMR analogy encouraged us to try the same approach in a solid. A single crystal of gypsum ($CaSO_4 \cdot 2H_2O$) seemed to be a quite suitable candidate. It is a diamagnetic salt, the proton-NMR spectrum is very simple,[8] and reorientation of the crystal leads to a significant change in the splitting of the NMR pattern. Furthermore, large single crystals are readily available.

The unit cell of single-crystal gypsum contains pairs of protons having two different orientations with respect to crystal axes, and therefore the proton-NMR pattern in an external field H_0 consists of four lines: ω_1 and ω_4 are resonance frequencies due to the first proton pair and ω_2 and ω_3 are resonance frequencies due to the second proton pair. As was shown by Pake,[8] these lines may partly coincide, depending on crystal orientation in the external field. The field that a proton experiences owing to the presence of its neighbor proton is given by

$$\delta H_p = \frac{3}{2} \frac{\mu_p}{r^3} (3 \cos^2\theta - 1) = \pm 5.4 \, (3 \cos^2\theta - 1) \qquad \text{(gauss)}. \qquad (1)$$

A muon in the place of a proton, however, would see a field that is smaller than that seen by a proton by a factor of 2/3, because the muon and proton have different precession rates. (Refer to Pake,[8] Sec. 7, for details.) Therefore:

$$\delta H_\mu = \pm \frac{\mu_p}{r^3} (3 \cos^2\theta - 1) = \pm 3.6 \, (3 \cos^2\theta - 1) \qquad \text{(gauss)}, \qquad (2)$$

where θ = angle between μ-p radius vector and $\vec{H}$, and r = 1.55 Å.

The interaction with the proton neighbor in the same water molecule leads to the line structure, whereas the interaction with protons farther away is responsible for the shape and width of the individual lines, which can be described by a Gaussian distribution:

$$\exp[-4(H-H_i)^2/\Delta H^2] = \exp[-4(\omega-\omega_i)^2/\Delta\omega^2],$$

where $\omega_i = \mu(\text{muon}) \cdot H_i/\hbar$ is the frequency or field at the center of an individual line (i); $H_i = H_0 + \delta H_i$, where δH_i is as given in Eq. (2); and $\Delta\omega$ is the full width of individual lines.

By observing the decay of the muon precession signal we are measuring the Fourier transform of the NMR line pattern or the superposition of muon precession frequencies weighted by the shape of the NMR pattern. The magnitude of the polarization vector is

$$F(t) = \exp(-t^2/T_2^2)\cos(\frac{\Delta\omega_1}{2}t)\cos(\frac{\Delta\omega_2}{2}t), \tag{3}$$

where we have made use of the mirror symmetry of the lines with respect to the external field H_0 or the frequency ω_0 that would be observed with no dipole interaction.

The central frequency is

$$\omega_0 = \frac{1}{4}(\omega_1 + \omega_2 + \omega_3 + \omega_4),$$

and the difference frequencies are

$$\Delta\omega_1 = \omega_2 - \omega_1 = \omega_4 - \omega_3,$$

$$\Delta\omega_2 = \frac{\omega_4 - \omega_1}{2} + \frac{\omega_3 - \omega_2}{2}.$$

Transverse relaxation time, T_2, is equal to $4/\Delta\omega$.

The experiment was performed at the 184-in. cyclotron at the Lawrence Radiation Laboratory, Berkeley, using the same apparatus that was used by Hague et al.[9] in a recent measurement of the muon magnetic moment. The precession of stopped polarized muons in a magnetic field of 1.0 and 4.5 kG, transverse to the axis of polarization (beam direction), was observed by means of the rotating asymmetric μ-decay pattern ($\mu^+ \rightarrow e^+ + \nu + \bar{\nu}$). Electrons from this decay, detected by a counter in the plane of the precession, were recorded as a function of lifetime of their parent muon and were formed in an elapsed-time histogram. This distribution can be described by

$$N = N_0 e^{-t/\tau_\mu} [1 + AF(t) \cos(\omega_0 t + \phi)] + BG, \qquad (4)$$

where τ_μ = 2.2 μsec, muon mean lifetime; A = initial decay asymmetry ($t \leq 10^{-9}$ sec); and BG = constant background.

Equation (4) can be used to determine the parameters T_2, A, ω_i's, N_0, ϕ, and BG by fitting it to the histogram. The first 10 μsec of the histogram, consisting of 600 cycles (at 4.5 kG), was used for this analysis.

One striking feature of Eqs. (3) and (4) is the prediction of occurrences of beats. However, even for the most favorable case (position 3, Table I) the first zero crossing of the asymmetry will not occur before 3.5 μsec. Because the signal at 1.6 mean lives is rather small, it is difficult to detect the beat behavior by means of fitting Eq. (4). Therefore we have used a simplified form of Eq. (4), which is a good approximation for $t < \tau_\mu$, the statistically most significant part of the histogram:

$$N_e(t) = N_0 \exp(-t/\tau_\mu)[1 + A \exp(-t^2/T_2^{*2}) \cos(\omega_0 t + \phi)] + BG. \quad (5)$$

The effective relaxation time, T_2^*, is related to the width of the complete "muon-NMR" pattern.

Table I summarizes the crystal orientations used, the calculated "muon-NMR" line pattern, the corresponding T_2^*, and the T_2^* obtained by fitting the data to Eq. (5). The uncertainty assigned to the measured T_2^* includes the statistical error as well as possible errors due to inaccuracies of the crystal orientation. The calculated T_2^* are seen to be in good agreement with the experimental ones.

The NMR line pattern was calculated for each crystal orientation by means of Eq. (2).

The individual line width, T_2, can now be directly obtained from the results of position 3, Table I, where the NMR splitting is almost zero and consequently $T_2^* = T_2 = 10.50$ μsec.

The relaxation time for polycrystalline powder is consistent with the foregoing analysis. The fit for the anhydrous powder gives a relaxation time that is very much longer and is consistent with no depolarization.

A search for the beating of the precession amplitude was done in the following way. Each histogram for positions 4 and 5 was divided into 500-nsec sections. In each section, a Fourier analysis was performed that led to values for the amplitude and the phase of the precession at the mean frequency ω_0. Equation (3) predicts the disappearance of the amplitude for position 4 at 7.0 μsec and for position 3 at 3.6 μsec and at 9.5 μsec. The experimental results, together with a curve

calculated from Eq. (3), are shown in Figs. 1 and 2.

The analyses of these results have been performed on the basis of assuming the muon to take the place of a proton, and the consistency of the explanation confirms the basic validity of this assumption. The results of the Fourier analysis with the demonstration of the agreement between the function [Eq. (3)] and the data implies, furthermore, that the muons we observed are found in the different sites of a proton with equal probabilities and in no other places.

As far as muonium formation is concerned, the muons that we observed could only have been in the muonium state for less than half a muonium hyperfine precession period ($\approx 10^{-10}$ sec). The most reasonable assumption is that these muons were placed into the site of a proton in the hot-atom reactions of muonium atoms. Muons emerging from that fraction of muonium atoms that were thermalized probably stay long enough as muonium to be completely depolarized. The observed asymmetry ($A = 0.15$) is about half that found in nondepolarizing targets (metals), indicating that about 50% of the muons have been depolarized by a "fast" mechanism. The site and fate of these muons is not known. A future publication on a variety of solids and liquids will deal in detail with this subject, i.e., fast depolarization mechanisms.

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FOOTNOTES AND REFERENCES

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Table I. Comparison between relaxation measurements and calculations for different orientations of the gypsum crystal. The crystal axes are given in the Onorato convention: (100) = x, (010) = y, z ⊥ (x, y). The measured T_2^* are independent of magnetic field strength.

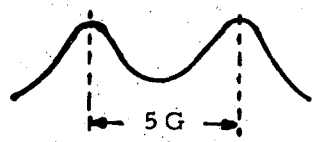
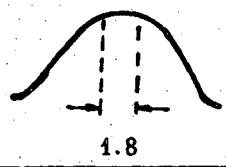
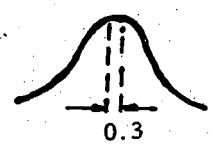
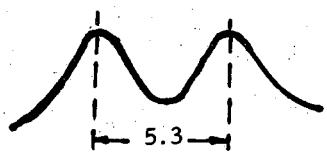
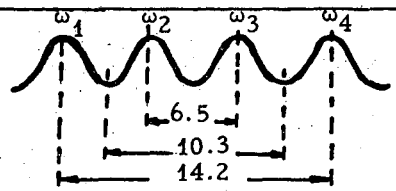
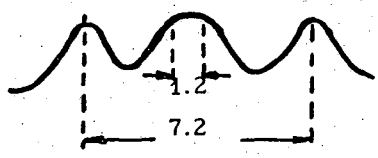
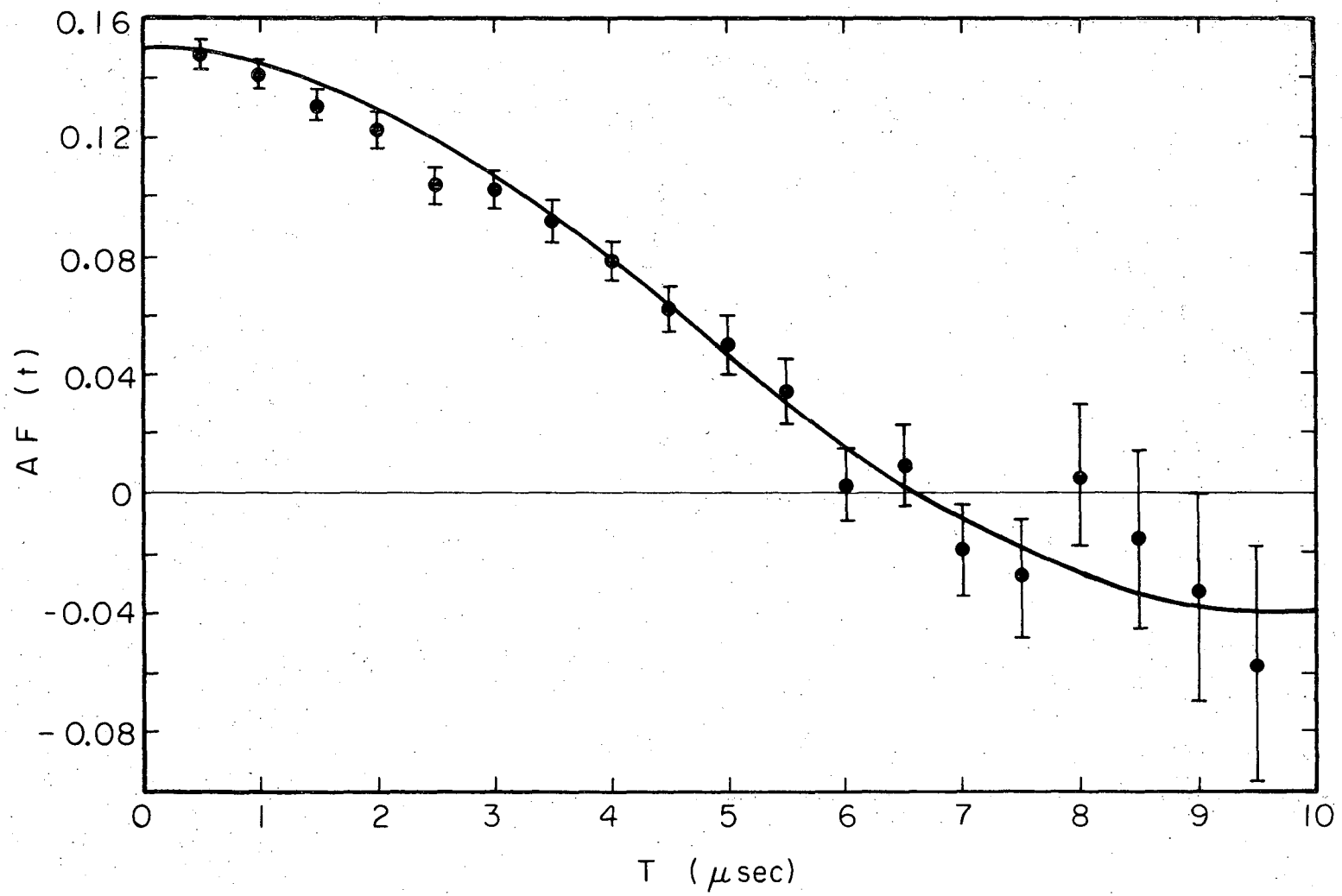
Position	Crystal orientation	Muon - NMR spectrum [scale = gauss]	Calculated T_2^* from NMR spectrum	Measured T_2^* (μsec)
1	$\vec{H}$, z = 90° $\vec{H}$, x = 94° $\vec{H}$, y = 57.5°		5.6	5.50 ±.50
2	$\vec{H}$, z = 90° $\vec{H}$, x = 146° $\vec{H}$, y = 62.5°		9.0	10.45 1.00
3	$\vec{H}$, z = 90° $\vec{H}$, x = 176° $\vec{H}$, y = 32.5°		10.50	10.50 1.00
4	$\vec{H}$, z = 0 $\vec{H}$, x = 90° $\vec{H}$, y = 90°		5.3	4.80 ±.30
5	$\vec{H}$, z = 40° $\vec{H}$, x = 142° $\vec{H}$, y = 111°		2.9	3.09 ±.20
6	$\vec{H}$, z = 24° $\vec{H}$, x = 90° $\vec{H}$, y = 66°		5.60	5.50 ±.40
Polycrystal, powder			5.3	5.30 ±.20
Polycrystal, anhydrous			---	60. ±20.

FIGURE CAPTIONS

Fig. 1. The observed asymmetry, AF(t) in Eq. (4) of text, for the gypsum crystal orientation having two NMR lines, position 4 in Table I. The asymmetry is calculated for 0.5-μsec intervals versus muon lifetime. The constant background is approximately 2% at t = 0. The solid curve slows the theoretical A·F(t) calculated from Eq. (3).

Fig. 2. The asymmetry for crystal position 5. The four lines are seen to interfere at about 4 μsec and the asymmetry reverses sign between 5 and 8 μsec before the statistics and background obscure the signal. The solid curve for A·F(t) was calculated from Eq. (3). The detailed agreement is marginal, suggesting that the actual crystal orientation was slightly different from the angles used. These angles were uncertain to ~ 10°.



XBL7011-4115

Fig. 1

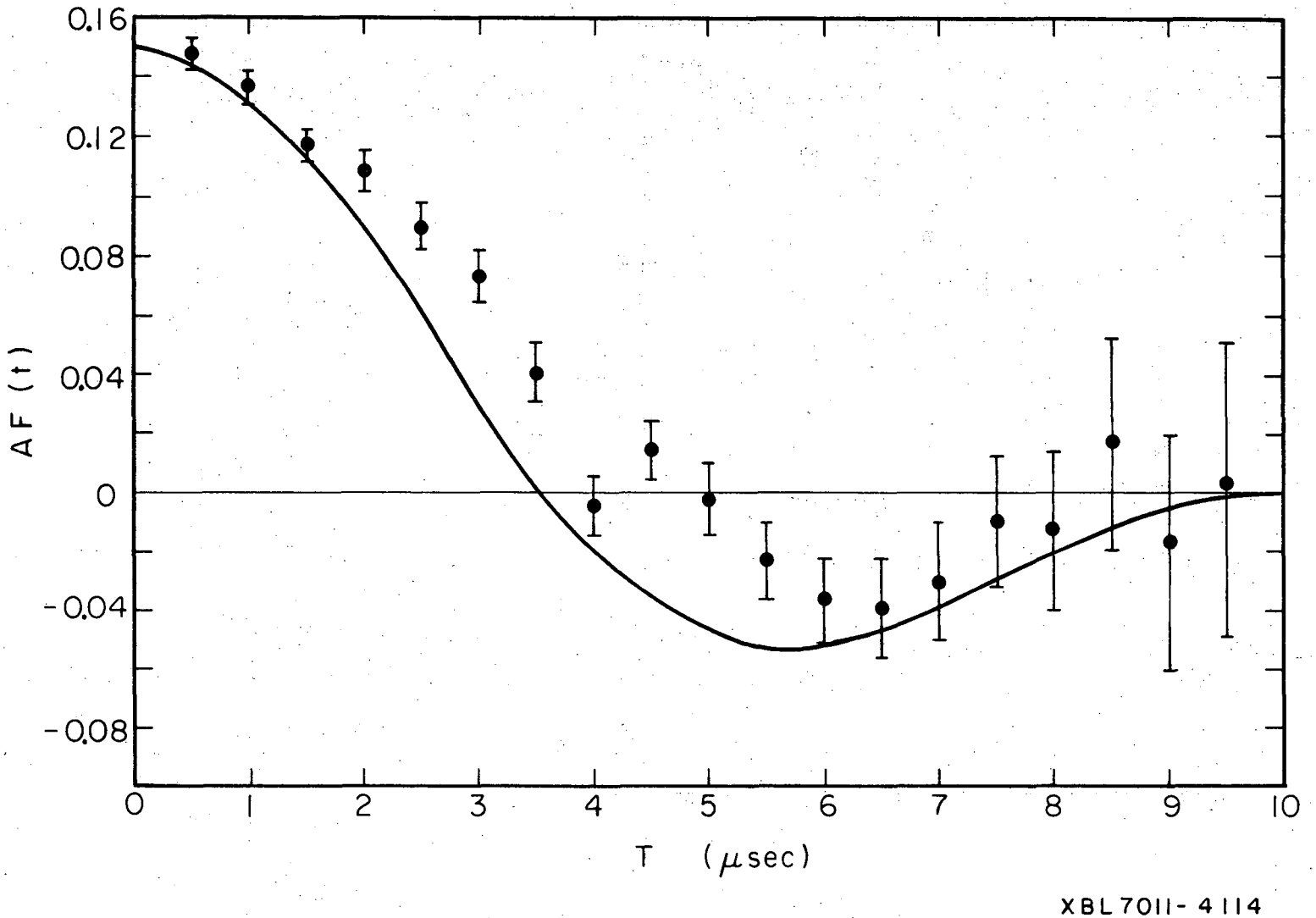

0.16
0.12
0.08
0.04
0
-0.04
-0.08
AF (t)
0
1
2
3
4
5
6
7
8
9
10
T (μsec)
XBL 7011-4114

Fig. 2

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