Low-Temperature Mobility of Positive Muons in Copper

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The spin relaxation of the positive muon in copper has been measured below 5 K in zero applied magnetic field. The results are well described by the theory of Kubo and Toyabe with a temperature-independent dipolar width. It is concluded that neither trapping nor changes in the muon site with temperature explain the increased mobility below 5 K.

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The positive-muon spin-relaxation (μ^+SR) technique has been used for over ten years to study the diffusion of the μ^+ in metals and alloys through the motional narrowing effect on the muon spin precession signal.[1] At high enough temperatures the μ^+ spin relaxation rate yields a diffusivity which follows the Arrhenius-law temperature dependence expected for a thermally activated diffusion process.[1] As the temperature is reduced, the linewidth increases as the slower hopping of the μ^+ is less efficient at averaging the random local magnetic fields in the metal. But at low enough temperatures complicated structure often appears,[1,2] with regions where the linewidth *decreases* with decreasing temperature, an effect which is believed to be associated with diffusion of muons into impurity traps.[3]

There remains an interesting possibility that for the μ^+ in metals there exists a diffusion process with a diffusivity decreasing with increasing temperature, which could arise from tunneling of the μ^+ at low temperatures. In this Letter we present experimental evidence for such a process in high-purity copper below 5 K, using the unique ability of zero-field μ^+SR (Ref. 4) to study the single-impurity mobility. Such a mechanism would help explain the results on some pure metals—most notably aluminum, where there is little evidence for μ^+ localization in the pure material even at temperatures as low as 30 mK,[5] and where dilute alloy studies[6] indicate a diffusivity proportional to $T^{-0.6}$ below 1 K. Because the muon is intermediate in mass between the electron and the proton, it may be able to offer insights into the onset of quantum behavior in diffusion.

Early transverse-field studies[7] of μ^+ diffusion in copper have yielded results which are consistent with a thermally activated diffusion process whereby the μ^+ tunnels between octahedral interstitial sites with weak lattice activation.[8] The linewidth increases monotonically as the temperature is lowered from 300 K and reaches a plateau below about 80 K, indicating that the μ^+ is stationary below that temperature. However, later work[5] showed a decrease of the linewidth by about 30% as the temperature was reduced from ~5 K to ~0.7 K, with a plateau below 0.7 K down to the lowest temperature (50 mK) studied.

In the transverse-field geometry[1] a magnetic field $\vec{H}_0$ is applied perpendicular to the initial μ^+ spin. Nuclear dipole fields $\vec{H}_d$ create random shifts in the Larmor frequency; for a stationary μ^+ this results in a Gaussian damping of the precession described by the transverse relaxation function $G_x(t) = \exp(-\sigma^2 t^2)$. Diffusion of the μ^+ with a mean time between hops $\tau \equiv \nu^{-1}$ will cause a reduction in the effective width of the random field distribution and, for $\nu \gg \sigma$, a change from Gaussian to Lorentzian of the precession line shape.

In the present work we use the zero-field μ^+SR technique. The μ^+ spin relaxes solely under the influence of $\vec{H}_d$, and the line shape for a stationary μ^+ is no longer simply an image of the dipolar field distribution. For a time-independent Gaussian field distribution, the relaxation function is[9]

$$G_z(t) = \langle \cos^2\theta + \sin^2\theta \cos(\gamma_\mu H_d t)\rangle$$
$$= \tfrac{1}{3} + \tfrac{2}{3}(1-\Delta^2 t^2)\exp(-\tfrac{1}{2}\Delta^2 t^2),$$

where $\gamma_\mu = 2\pi \times 13.55$ kHz/Oe is the μ^+ gyromagnetic ratio, Δ/γ_μ is the width of a single component of $\vec{H}_d$, and θ is the angle between $\vec{H}_d$ and the muon spin. This function initially decays as $\exp(-\Delta^2 t^2)$, reaches a minimum at $t_{min} \equiv \sqrt{3}\Delta^{-1}$, and recovers to $\frac{1}{3}$ as $t\to\infty$. The polycrystalline averages of the zero-field width Δ and the high-transverse-field width $\sqrt{2}\sigma$ are related by[4] $\Delta = \sqrt{5}\sigma$ or, including the quadrupole effect appropriate for copper,[4,10] $\Delta = 2\sigma$.

The important aspect of this work is the effect of μ^+ motion on $G_z(t)$. The theory was worked out initially by Kubo and Toyabe,[9] and subsequently applied to μ^+SR by Hayano *et al.*[4] For slow enough hopping ($\nu < \Delta$) the initial decay remains as $\exp(-\Delta^2 t^2)$, while the asymptotic value $G_z(t \to \infty) = \frac{1}{3}$ is rapidly suppressed as the hopping rate increases to a value $\nu \simeq \Delta$. Only for $\nu \gtrsim \Delta$ is there a narrowing and a change from Gaussian to Lorentzian line shape similar to that in the transverse-field case. If the long-time polarization can be determined, the zero-field method is much more sensitive to slow hopping of the μ^+ than is the transverse-field method.

Apart from the sensitivity per se the parameters ν and Δ are, for $\nu < \Delta$, "uncoupled" in that Δ governs the behavior of $G_z(t)$ for $t \lesssim t_{min}$, while ν determines the later behavior for $t \gtrsim t_{min}$. Thus the static (Δ) and dynamic (ν) aspects of the relaxation are clearly separated. This is in strong contrast to slow hopping in the transverse-field case, where there occurs only a small change in the width which can be attributed to a change in either ν or σ.

We have performed a zero-field experiment in copper[10,11] on the positron-free M9-W3 surface muon beam line at TRIUMF. Two high-purity samples were used, a slice of the same polycrystal sample used in Ref. 5 and an oxygen-annealed single crystal. Within the uncertainties, no differences are seen between them, and so we do not discriminate between them here. Further information about the samples and experimental techniques can be found in Refs. 5, 10, and 12.

Positron spectra were taken in the forward and backward directions at each temperature, and were each analyzed by a least-squares fit by the theory of Ref. 4 for $G_z(\nu, \Delta, t)$. No background corrections were made except for the subtraction of a small time-independent term due to accidental events. The data were analyzed in two passes. First, Δ and ν were both allowed to vary independently, giving the result for $\Delta(T)$ shown in Fig. 1(a). From this we conclude $\Delta = 0.389 \pm 0.003$ μs^{-1}, independent of temperature. Then Δ was fixed to this value, and the fits were done with only ν varying, with the results shown in Fig. 1(b). On one axis of Fig. 1(b) is indicated the diffusivity for octahedral occupancy, $D = (\frac{1}{12})\alpha_0^2\nu$, where $\alpha_0 = 3.61$ Å is the cubic lattice constant of Cu. A measurement at 21.9 K yielded essentially the same results as the 5.15-K data.

In Fig. 2 we show the experimental points for

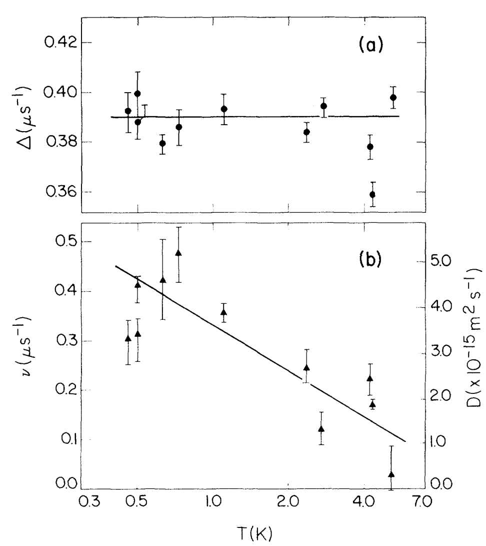

FIG. 1. Temperature dependence of (a) zero-field dipolar width and (b) hopping rate and diffusivity. The lines are drawn only to guide the eye.

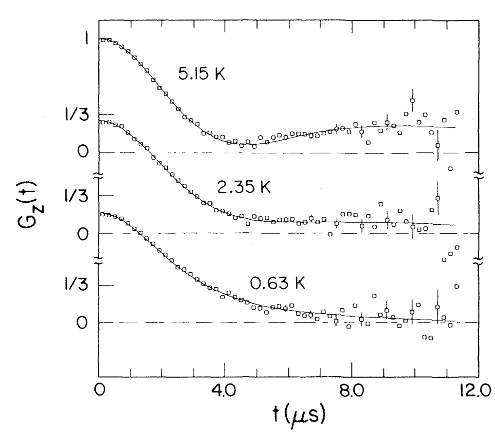

FIG. 2. Experimental data for $G_z(t)$ at three representative temperatures. The lines are the theory of Ref. 4 fitted to the data.

$G_z(t)$ derived from the raw data using fitted values for the normalization and background. The decay and recovery of the polarization is clearly seen at 5 K while the recovery is completely suppressed at 2.35 K. At 0.6 K the motional narrowing is visible. The solid curves are the theoretical fits to the data.

The good agreement of our data with the Kubo-Toyabe model is evidence against trapping as the cause of the anomalous temperature dependence in this temperature region, as has recently been suggested by Chappert *et al.*[13] The zero-field relaxation function for motion with trapping has been calculated by Petzinger,[14] who assumed that the relaxation occurs only when the μ^+ is trapped and that the temperature is low enough such that no detrapping occurs. These assumptions are valid in the temperature regime where $\sigma(T)$ is increasing due to the muons diffusing into the traps, which, if one invokes trapping to explain the results, is just the regime of our experiment.

Rather than a hopping rate, this model is parametrized by a trapping rate governing the approach of the μ^+ distribution to the traps. Because all relaxation is due to static fields, a recovery to $\frac{1}{3}$ is always expected in this model. Furthermore, the initial decay is not Gaussian and shows large changes with the trapping rate. This model has been shown to describe the zero-field μ^+SR experiments for Nb,[15] where the transverse-field work[3] had already established the influence of impurities.

Our data cannot be fitted by Petzinger's model—they clearly show that the relaxation is described by a local field distribution having a constant width, with a hopping rate that decreases with increasing temperature. The initial Gaussian decay is unchanged until sufficient motion is taking place that the recovery to $\frac{1}{3}$ is completely suppressed. However, we cannot rule out the possibility that a more complicated trapping mechanism may give the observed type of relaxation. If there existed a distribution of trap energies and concentrations, the muons could reach successively deeper traps as the temperature is raised. Because all traps would need to exhibit closely similar dipolar widths in order to have $\Delta(T)$ remain constant we consider this unlikely.

Another explanation which can be eliminated is that of Seeger,[16] who has proposed a metastable state at the tetrahedral site with a thermally activated transition to the octahedral site beginning at 0.7 K. This model cannot be correct as it predicts a temperature-dependent Δ and a stationary μ^+ below 0.7 K. Both of these implications are contrary to our data. Instead, we suggest that the μ^+ site does not vary with temperature between 0.7 and 5 K, but that a diffusion process occurs which is limited by static disorder (e.g., Anderson localization by impurity strains[17]) below 0.7 K, and by thermal disorder above 5 K. An extension of the present experiment to temperatures below 0.5 K would be very valuable to show whether the μ^+ does in fact remain mobile at the lowest temperatures, as is indicated by the transverse-field studies.[5]

We comment briefly on the magnitude of Δ. From the known lattice expansion[7] about the μ^+ of 4.9% we can calculate the expected value[10] $\Delta = 0.350\ \mu s^{-1}$, independent of crystal orientation. The deviation of ~10% from the measured value may indicate that the lattice expansion is not perfectly symmetric, or that the simple way in which the quadrupole interaction was treated[10] was inadequate.

In summary, we have shown with zero-field μ^+SR that the low-temperature spin relaxation in copper is well described by the Kubo-Toyabe model with a temperature-independent static dipolar width and a hopping rate which decreases as the temperature is increased from 0.7 to 5 K. We have concluded that neither the conventional model for trap-limited diffusion nor models based on a change in trapping site with temperature are capable of explaining our results. Further theoretical work to clarify the diffusion mechanism would complement the present understanding[8] of the behavior at higher temperatures.

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[1]E. Karlsson, Phys. Rep. 82, 271 (1982); J. H. Brewer and K. M. Crowe, Annu. Rev. Nucl. Part. Sci. 28, 239 (1978).

[2]K. G. Petzinger, R. L. Munjal, and E. Zaremba, Hyperfine Interact. 6, 223 (1979).

[3]T. O. Niinikoski, O. Hartmann, E. Karlsson, L.-O. Norlin, K. Pernestal, K. W. Kehr, D. Richter, E. Walk-

er, and K. Schulze, Hyperfine Interact. 6, 229 (1979).
[4]R. S. Hayano, Y. J. Uemura, J. Imazato, N. Nishida, T. Yamazaki, and R. Kubo, Phys. Rev. B 20, 850 (1979).
[5]O. Hartmann, E. Karlsson, L.-O. Norlin, T. O. Niinikoski, K. W. Kehr, D. Richter, J.-M. Welter, A. Yaouanc, and J. Le Hericy, Phys. Rev. Lett. 44, 337 (1980).
[6]K. W. Kehr, D. Richter, J.-M. Welter, O. Hartmann, E. Karlsson, L.-O. Norlin, T. O. Niinikoski, and A. Yaouanc, Phys. Rev. B 26, 567 (1982).
[7]I. I. Gurevich, E. A. Mel'eshko, I. A. Muratova, B. A. Nikol'skii, V. S. Roganov, V. I. Selivanov, and B. V. Sokolov, Phys. Lett. 40A, 143 (1972); H. Schilling, M. Camani, F. N. Gygax, W. Ruegg, and A. Schenck, Hyperfine Interact. 8, 675 (1981).
[8]H. Teichler, in *Exotic Atoms '79, Fundamental Interactions and Structure of Matter*, edited by K. Crowe, J. Duclos, G. Fiorentini, and G. Torelli (Plenum, New York, 1980), p. 283.
[9]R. Kubo and T. Toyabe, in *Magnetic Resonance and Relaxation*, edited by R. Blinc (North-Holland, Amsterdam, 1967).
[10]C. W. Clawson, Ph.D. thesis, Lawrence Berkeley Laboratory Nuclear Science Division Report No. LBL-14489, 1982 (unpublished).
[11]C. W. Clawson, K. M. Crowe, S. E. Kohn, S. S. Rosenblum, C. Y. Huang, J. L. Smith, and J. H. Brewer, Physica (Utrecht) 109&110B, 2164 (1982).
[12]F. Fickett, Mater. Sci. Eng. 14, 199 (1974).
[13]J. Chappert, A. Yaouanc, O. Hartmann, E. Karlsson, L.-O. Norlin, and T. O. Niinikoski, Solid State Commun. 44, 13 (1982).
[14]K. G. Petzinger, Phys. Lett. 75A, 225 (1980), and Hyperfine Interact. 8, 639 (1981).
[15]C. Boekema, R. H. Heffner, R. L. Hutson, M. Leon, M. E. Schillaci, W. J. Kossler, M. Numan, and S. A. Dodds, Phys. Rev. B 26, 2341 (1982).
[16]A. Seeger, Phys. Lett. 93A, 33 (1982).
[17]O. Hartmann, E. Karlsson, L.-O. Norlin, D. Richter, and T. O. Niinikoski, Phys. Rev. Lett. 41, 1055 (1978).