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AUTHOR(S) J. H. Brewer, M. Celio, D. R. Harshman, R. Keitel, S. R. Kreitzman, G. M. Luke, D. R. Noakes, R. E. Turner, E. J. Ensaldo, C. W. Clawson, K. M. Crowe, and C. Y. Huang

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DETERMINATION OF VERY SLOW μ^+ HOP RATES IN Cu BY LLF-μSR

J.H. Brewer, M. Celio, D.R. Harshman, R. Keitel, S.R. Kreitzman, G.M. Luke, D.R. Noakes and R.E. Turner, TRIUMF, 4004 Wesbrook Mall, Vancouver, B.C., Canada V6T 2A3

E.J. Ansaldo, Dept. of Physics, University of Saskatchewan, Saskatoon, Saskatchewan, Canada S7N 0W0

C.W. Clawson and K.M. Crowe*, Lawrence Berkeley Laboratory, University of California, Berkeley, California, USA 94720

C.Y. Huang, Los Alamos National Laboratory, Los Alamos, NM, USA 87545

Muon spin relaxation in low (weak) longitudinal magnetic field (LLF-μSR) provides a means of independently determining the static dipolar width Δ characterizing the μ^+ lattice site and the correlation time τ_c for μ^+ hopping, in a manner that is nearly model-independent for τ_c and especially accurate in the near-static limit ($\tau_c > \tau_\mu$). The advantages of this method are illustrated by its application to muon hopping in Cu near the τ_c maximum around 50 K.

Positive muon spin relaxation (μ^+SR) has found one of its main applications in the investigation of μ^+ diffusion (analogous to hydrogen diffusion) in metals, by virtue of the "motional narrowing" effect of muon "hopping" on the relaxation rate./1/ The classic example of this application is μ^+ diffusion in copper, where the temperature dependence of the μ^+ motion above 80 K is well understood in terms of thermally activated muon tunneling between octahedrally coordinated interstitial sites (o-sites) where the muon is localized by self-trapping [see summary of various authors' work in Ref. 1]. In 1979, transverse-field (TF)-μSR experiments at CERN/2/ indicated that the correlation time τ_c for μ^+ hopping in Cu decreases with decreasing T below about 50 K, levelling off below about 0.3 K to a value approximately equal to that at 130 K. This surprising result stimulated considerable theoretical speculation about the mechanisms responsible./3,4/

Although the field-dependence of the effective relaxation rate was consistent with μ^+ hopping between o-sites,/2/ the TF-μSR time spectra themselves did not offer much model-specific information. Zero-field (ZF)-μSR experiments were therefore performed at TRIUMF/5/ and BOOM/6/ to take advantage of the superior sensitivity of ZF-μSR time spectra to the details of the relaxation mechanism -- particularly to the decay of the "$\frac{1}{3}$ tail" of the Kubo-Toyabe (KT) relaxation function/7/ due to slow hopping of the μ^+ between o-sites. The results were entirely consistent with the previous interpretation of the TF-μSR data; however, some controversy remained because of the conjecture/8/ that the μ^+ might temporarily occupy a metastable tetrahedrally coordinated interstitial site (t-site) with a static dipolar width Δ_t different from that of the more stable o-site, Δ_o. It appeared possible to find a suitable combination of such effects that would generate ZF relaxation functions consistent, within statistical and systematic uncertainties, with existing ZF-μSR data.

Subsequently, several new developments have effectively laid this controversy to rest. First, a high-precision ZF-μSR measurement at BOOM/9/ was able to detect the deviation of the ZF relaxation function $G_{zz}(t)$ from the simple gaussian Kubo-Toyabe function $g_{zz}^{KT}(t)$/7/ as predicted by the quantum mechanical calculations of Holzschuh and Meier/10/ in the static (no μ^+ hopping) limit. The precision of this measurement left little room for alternate interpretations. Second, an efficient method was found for calculating precise quantum mechanical relaxation functions for the μ^+ in an o-site in Cu in the static limit ($\tau_c \rightarrow \infty$) including both electric quadrupole and longitudinal field effects, thus providing tables of rigourous theoretical functions with which to compare $G_{zz}(t)$ data. [See M. Celio, these Proceedings.] Third, level-crossing resonance muon spin relaxation (LCR-μSR) measurements of the Cu quadrupole coupling to the EFG of the muon/11/ [see also S.R. Kreitzman, these Proceedings] have left no doubt that the μ^+ continues to occupy the o-site down to 5 K where τ_c is markedly decreased from its maximum near 50 K. Finally, the use of low (weak) longitudinal magnetic field (LLF)-μSR to independently determine the static width Δ and the correlation time τ_c has confirmed that Δ does not change appreciably at low T./12/ In developing LLF-μSR for this application, we realized that a combination of ZF- and LLF-μSR could also provide more precise and reliable values of τ_c near its maximum at around 50 K. In this paper we explain why this is so and present the results of LLF-μSR measurements of τ_c in copper.

In the limit of slow hopping ($\tau_c > \tau_\mu$), μ^+ motion in Cu should be a good example of stochastic "strong-collision" dynamics. The dynamic relaxation function $G_{zz}(t,\nu)$ can thus be obtained from the static relaxation function $g_{zz}(t)$ for any value of the hop rate $\nu = 1/\tau_c$ by numerical integration (using the Trapezoidal rule) of the resulting Volterra equation of the second kind,/13/

$$G_{zz}(t,\nu) = \exp(-\nu t)\; g_{zz}(t) + \nu \int_0^t dt'\; G_{zz}(t-t',\nu)\; \exp(-\nu t')\; g_{zz}(t') \qquad [1]$$

as prescribed by Holzschuh and Meier,/10/ thus producing a tabulated dynamic function $G_{zz}(t,\nu)$ that can be compared with experimental data to determine $\nu = 1/\tau_c$. Examples of the results of such numerical "dynamicizations" of Celio's static $g_{zz}(t)$ functions for Cu in ZF and with LLF along the <111> crystal axis are displayed in Figs. 1 and 2. The function $G_{zz}(t,\nu,\omega_{LF})$ is actually stored in a 3-dimensional array which is interpolated by a generalized table-lookup function for fitting. [Available on request for VAX VMS installations.]

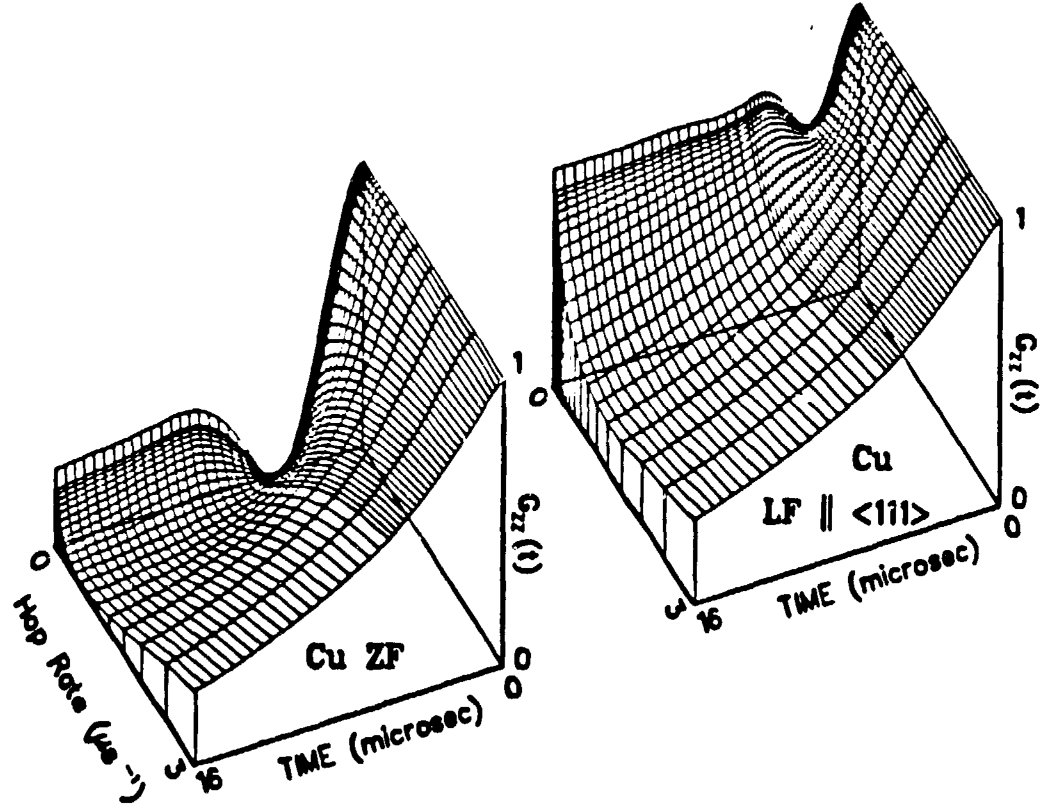

Figure 1. Dynamic relaxation functions $G_{zz}(t,\nu)$ for (a) ZF and (b) LLF (12 G) with hop rate ν ranging from 0 to 3 μs^{-1}. The static function $g_{zz}(t)$ is calculated for the Cu o-site with [in (b)] the field along the <111> axis.

Inspection of Fig. 1 will reveal several important advantages of the LLF-μSR method: First, there is a "T_1 minimum" effect in both ZF and LLF cases where the relaxation function is least sensitive to small changes in ν; however, for ZF this flat region occurs for hop rates of about 1 μs^{-1}, whereas for a LF of 12 G it moves to $\nu \approx 2$ μs^{-1}. It is thus possible to "tune" the LF to an optimum value for any given ν. Second, the gross difference between the static and dynamic relaxation functions is roughly twice as large for the LLF case as for the ZF case, giving correspondingly more "leverage" on ν, over all but the earliest time range. Third, the time range over which the effects of slow hopping are most pronounced starts at earlier times (4 μs) for the LLF case, while accurate measurements of the decay of the "$\frac{1}{3}$ tail" in the ZF case depend on the data beyond 6 μs. This is illustrated clearly in Fig. 2, as is the final (and perhaps most important) distinction between ZF and LLF spectra: In Cu, owing to the complexity of the spin system in the o-site, the ZF "$\frac{1}{3}$ tail" is neither time-independent nor equal to $\frac{1}{3}$ at long times. This no longer presents a problem for Cu now that the exact static $g_{zz}(t)$ is known, but it illustrates the *model dependence* of precise interpretations of ZF data in terms of hopping.

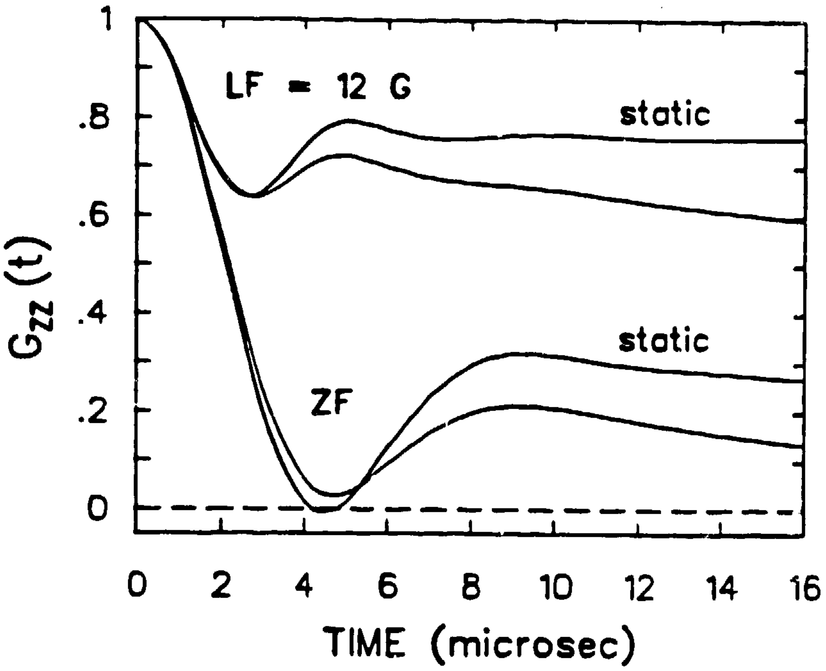

Figure 2. Detail of static and selected dynamic cases of the same $G_{zz}(t,\nu)$ functions shown in Fig. 1.

By contrast, the LLF static function has a flat long-time asymptote; it also has the (model-dependent) shape of the initial decay decreased in relative magnitude and compressed to earlier times, and more closely resembles the gaussian Kubo-Toyabe shape than does the ZF function. It thus provides a more model-independent and precise method for determining ν.

The experiment consisted of typical ZF- and LF-μ^+SR measurements./1/ A beam of 4.1 MeV "surface muons" was stopped in several single crystals of Cu, mounted inside a ^{4}He flow cryostat (with a ^{3}He evaporation insert for the 0.7 K measurement/5/). Positrons from μ^+ decay were detected in "forward" (F) and "backward" (B) scintillation counters and the time distributions $F(t)$ and $B(t)$ of such events relative to the muon's entry into the target at t=0 were collected in a computer in the conventional way./1/ These time spectra were later combined and corrected according to standard practice/12/ to form the asymmetry spectrum $A(t) = A_0 G_{zz}(t)$. The empirical maximum asymmetry factor A_0 was determined as usual from TF measurements/1/ and subsequently held fixed for ZF- and LF-μ^+SR fits.

The four time spectra taken with longitudinal fields of 0 (ZF) and 9 G (LLF) at 0.7 K and 4.2 K are plotted together in Fig. 3 for illustrative purposes. The results of these and other fits are summarized in Fig. 4. These high-precision measurements of τ_c in the nearly-static region are expected to play a crucial role in the ultimate explication of the mysterious hopping mechanism at lower temperatures.

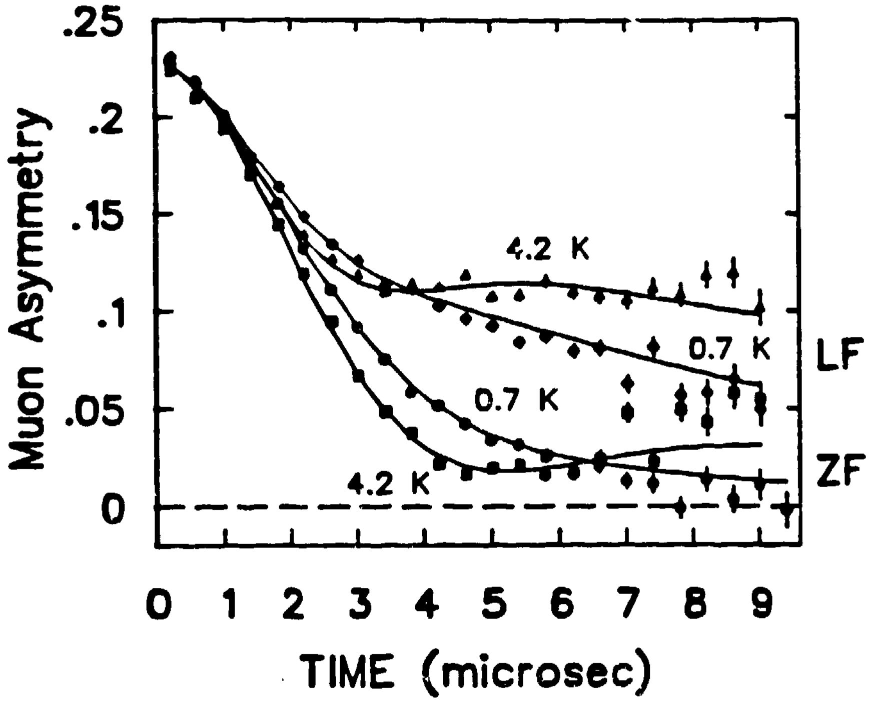

Figure 3. Positive muon relaxation functions in Cu crystal for zero applied field (ZF) at 4.2 K (squares) and 0.7 K (circles) and for 9 G longitudinal field (LF) at 4.2 K (triangles) and 0.7 K (diamonds). The data were fitted globally to a common scale factor relative to the undistorted o-site (consistent with unity) and common hop rates at 0.7 and 4.2 K.

Although the calculational methods developed by Celio and the LLF-μSR experimental technique have brought us much closer to a rigourous understanding of the empirical behaviour of the μ^+ in Cu, several pitfalls await the incautious interpreter of these results: First, when ν becomes comparable to the frequency of the electric quadrupole splitting of the Cu spins (1 Mz), it is no longer reasonable to use the present method of "dynamicization", which convolutes "static" functions in which the oscillating components of the Cu dipolar fields have already been averaged out. In effect, at higher hop rates the muon must seem to see an increased static dipolar width. We therefore have not included our data above about 160 K, in which region $\nu > 1$ MHz. Second, we have made no attempt to account for the fact that when a muon hops to a new site, it is probably adjacent to the old site, which therefore shares several Cu spins in common. This amounts to a partial breakdown of the strong collision approximation and should have measurable effects.

Nevertheless, we are confident that the numbers shown in Fig. 4 differ from the "true" μ^+ hop rates in Cu by at most a common factor very close to unity.

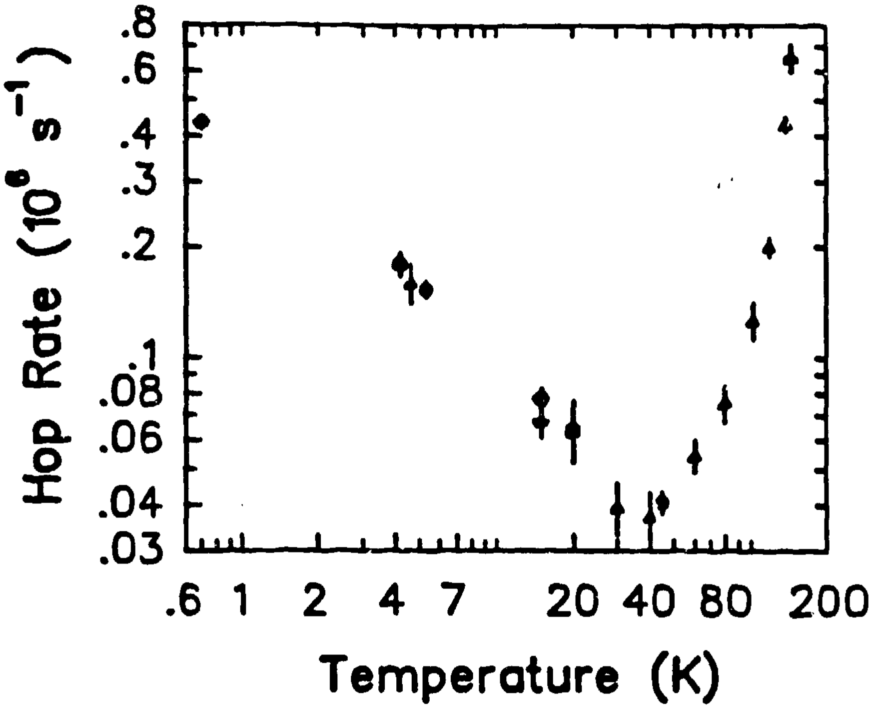

Figure 4. Muon hop rates extracted from LLF-μSR time spectra in Cu. Triangles: global fit to spectra at various temperatures with the same value of the LF (14.3 G); squares and circles: global fits to several spectra at the same temperature with various values of LF. Note very small statistical errors on the latter.

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REFERENCES

* *Current address:* Tektronix Corp., Beaverton, Oregon, USA 97124.

/1/ A. Schenck, *Muon Spin Rotation Spectroscopy -Principles and Applications in Solid State Physics*, (Adam Hilger Ltd., Bristol and Boston, 1985).
/2/ 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); O. Hartmann, Hyperfine Interactions 8, 525 (1981).
/3/ J. Kondo, Physica 125B, 279 and 126B, 377 (1984).
/4/ K. Yamada, Prog. Theor. Phys. 72, 195 (1984).
/5/ C.W. Clawson, K.M. Crowe, S.E. Kohn, S.S. Rosenblum, C.Y. Huang, J.L. Smith and J.H. Brewer, Physica 109 B and 110 B, 2164 (1982).
/6/ R. Kadono, J. Imazato, K. Nishiyama, K. Nagamine, T. Yamazaki, D. Richter and J.M. Welter, Hyperfine Interactions 17-19, 109 (1984).
/7/ R.S. Hayano, Y.J. Uemura, J. Imazato, N. Nishida, T. Yamazaki and R. Kubo, Phys. Rev. B20, 850 (1979).

/8/ A. Seeger and L. Schimmele, Hyperfine Interactions 17-19, 133-138 (1984).
/9/ R. Kadono, J. Imazato, K. Nishiyama, K. Nagamine, T. Yamazaki, D. Richter and J.M. Welter, Phys. Lett. 107A, 279 and 109A, 61 (1985).
/10/ E. Holzschuh and P.F. Meier, Phys. Rev. B29, 1129 (1984).
/11/ A.D. Booth, *Numerical Methods*, 3rd Ed. (Butterworths, London 1966).
/12/ S.R. Kreitzman, J.H. Brewer, D.R. Harshman, R. Keitel, D.Ll. Williams, K.M. Crowe, E.J. Ansaldo, Phys. Rev. Lett. 56, 181 (1986).
/13/ J.H. Brewer, S.R. Kreitzman, K.M. Crowe, C.W. Clawson, S.S. Rosenblum and C.Y. Huang, submitted to Phys. Lett. A (1986).