Title
ZERO-FIELD u SR AND LCW-TEMPERATURE u+ DIFFUSIVITY IN COPPER

Author
Clawson, C.W.

Publication Date
1983-04-01

LBL-16291

Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

Invited talk presented at the Yamada Conference on
Muon Spin Rotation and Associated Problems,
Shimoda, Japan, April 18-22, 1983

ZERO-FIELD μSR AND LOW-TEMPERATURE μ^+ DIFFUSIVITY IN COPPER

C.W. Clawson

April 1983

NUCLEAR SCIENCE DIVISION

Prepared for the U.S. Department of Energy under Contract DE-AC03-76SF00098

DISCLAIMER

LBL-16291

Zero-field μSR and Low-Temperature μ^+ Diffusivity in Copper

C.W. Clawson

Department of Physics and Lawrence Berkeley Laboratory,
University of California, Berkeley, Ca. 94720

ABSTRACT

In this paper I review the history of μ^+ diffusion studies in copper, with particular emphasis on the increased low-temperature diffusivity which has been known for several years now. I survey the theory and practice of the zero-field μSR method, which has come into increasing favor in the study of muon diffusion and trapping in metals, and discuss its application to the low-temperature copper problem.

Invited talk presented at the Yamada Conference on Muon Spin Rotation and Associated Problems, April 18-22 1983, Shimoda, Japan.

This work was supported by the Director, Office of Energy Research, Division of Nuclear Physics of the Office of High Energy and Nuclear Physics of the U.S. Department of Energy under Contract DE-AC03-76SF00098.

The study of muon motion in metals by the motional narrowing effect is one of the oldest problems in solid state μSR. A wealth of experimental data now exists, showing a rich variety of behavior. Karlsson[1] has given a recent review.

Some pure metals, notably aluminum, show little or no relaxation of the μ^+ spin precession[2] at most temperatures if at least moderately pure. Others, for example Bi,[3] Nb[4] and V,[5,6] show rather complicated structure of the relaxation rate as a function of temperature. It is now believed that in many, if not all, of these cases the complexity is due to the capture and release of the μ^+ by traps. In the case of Nb, the influence of impurity traps has been demonstrated[7-9] very clearly.

Thus it was serendipitous that Cu, the first metal to be used[10] in a μSR diffusion study, showed a very straightforward curve of relaxation rate against temperature,[11] fitting very well to an Arrhenius curve between about 90 K and 300 K. That the small deviations from classical thermally activated behavior, as well as the magnitudes of the activation energy and attempt frequency, were quantitatively explained[12] by considering a quantum mechanical diffusion mechanism further encouraged the belief that here was a system where the diffusion of the muon in a pure host crystal could be observed—a valuable proving ground for both quantum diffusion theory and the μSR motional narrowing technique.

Even more justification for the belief that muon diffusion in copper was characteristic of the pure material came from the success of the orientation dependence studies[13] of the linewidth in single crystals. This work clearly identified the position of the μ^+ at the octahedral interstitial site, surrounded by copper nearest-neighbor nuclei. The presence of an electric field gradient at the neigboring nuclei was also demonstrated, again indicating that the nearest neighbors are copper.

Still not understood, however, is the result obtained[2,14] when measurements were extended to below 5 K. Here the linewidth, being nearly constant between 10 K and 80 K, decreases by 30-40% over a range of only a few degrees K.

It is known that even in rather dirty copper[14] the relaxation still follows the same curve above 80 K as does that in the purest material, but that the decrease in linewidth at low temperatures is less pronounced. At this point one must begin to wonder: Is the behavior in copper really due to the pure material, or is it so completely dominated by impurities that even the purest material exhibits a diffusion curve characteristic of the capture and release of the μ^+ by traps? If the trapping site were not immediately adjacent to an impurity site, then all the work mentioned above would not discriminate between self-trapping in a pure Cu matrix and impurity-enhanced trapping. Indeed, in the case of muon trapping by Mn impurities in Al[15] the μ^+ is believed to be surrounded by Al nearest neighbors. This has been attributed to the presence of long-range strain fields caused by the impurities, creating energy shifts which cause localization by the Anderson mechanism.

If impurity trapping were invoked to explain the results on copper the interpretation would be that below 5 K the linewidth increases with temperature as the muons diffuse more rapidly into the traps, and that above 80 K the linewidth decreases as the muons are thermally activated out of the traps. It is very difficult to determine with transverse-field experiments whether this is the case, or whether the muon actually diffuses more slowly as the temperature is raised. Less difficult, but still somewhat problematic, is the question of whether the muons diffuse at all or whether the change in linewidth is mainly a static effect arising from a change in the trapping site, as has recently been suggested.[16] However, a zero-field experiment is ideally suited to discriminate among these alternatives.[17]

The reason for this sensitivity of the zero-field method is that the relaxation function for static random magnetic fields is qualitatively different than that for fluctuating fields. In transverse field the effect of a slight motion of the muon is to create a small change in the linewidth. An increase in the linewidth due to slower diffusion is indistinguishable from an increase due to a faster approach to the traps. In the zero-field case when the relaxation rate is limited by the approach of muons to the traps, the relaxation function must always approach 1/3 for long times because, once trapped, the muon is relaxed by a static random field. But if a slight motion decreasing with increasing temperature occurs, the first evidence of it is the loss of this 1/3 asymptotic polarization.[18] No such qualitative feature in the relaxation function is present in the transverse-field case.

That the polarization approaches 1/3 for relaxation by static fields is easily seen. If a muon precesses, describing a cone of half-angle ϑ about the local field, then there is a d.c. component of the spin along the field proportional to $\cos\vartheta$. Projecting back onto the original spin direction gives a constant component $\cos^2\vartheta$ which, averaged over directions of the local field, yields 1/3. As long as the random field is isotropic and static, and independently of its actual distribution in magnitude, the asymptotic polarization will be 1/3.

In the case of muons diffusing rapidly into the traps, i.e. in the region where the linewidth is increasing with temperature, such a result still holds under the assumption that the muons are not relaxed significantly until reaching a trap. This assumption should hold if one is not too close to the minimum of the linewidth. What does occur in this case is that the initial decay rate of the polarization is sensitive to the rate at which the muons approach the traps. Thus we can distinguish two regimes. If the muons are approaching the traps by diffusing rapidly enough, we expect a Petzinger[17] relaxation function with a variable initial decay rate and a long-time polarization of 1/3. If the muons are diffusing without trapping or diffusing rapidly enough out of the traps, we expect a Kubo-Toyabe[18] relaxation where the initial decay rate remains constant while the 1/3 long-time polarization is suppressed. These two regimes have been shown[9] to occur in the capture and release of muons from traps in Nb.

At TRIUMF we have performed zero-field and transverse-field experiments on copper for $0.5\ K < T < 5\ K$ and also near 20 K.[19,20] Because of the need for determining the long-time polarization accurately, a very "clean" experiment is needed. The experimental facilities at TRIUMF include a positron-free surface muon beam and a high-rate, low-background general purpose μSR spectrometer[21] equipped with a ^{3}He evaporation cryostat capable of reaching 0.5 K. We have used two copper samples, one being a slice of the polycrystal used in Ref. 2 and one being a very high purity single crystal. Details can be found in Ref. 20, but it suffices to say here that we observed no significant differences between the samples.

The results of the experiments are simple. The spectra are fit very well by a Kubo-Toyabe relaxation function with a temperature independent dipolar width and a hopping rate that decreases from $\sim 0.4\ \mu s^{-1}$ to $\sim 0.05\ \mu s^{-1}$ as the temperature is increased from 0.5 K to 5 K. The width was at first allowed to vary in the fits so that any change in the initial decay rate would be seen. As can be seen in Fig. 1, no systematic temperature dependence is apparent. These results show both that the muons are mobile at all temperatures from 5 K down to 0.5 K and that they are not approaching traps but rather diffusing more slowly as the temperature is raised. From the transverse-field work[2,14] it was rather apparent that the muons were mobile, so the elimination of trapping is the most important feature of the zero-field experiment.

The existence of a process which yields a diffusivity decreasing as the temperature increases would be interesting indeed. Such a process is indicated both by the copper experiments and, somewhat more indirectly, by the recent impurity-trapping studies in doped Al,[22] which imply a diffusivity in pure Al proportional to $T^{-0.6}$ below 1 K.

It is impossible on purely empirical grounds to state what the mechanism for such diffusion is. The only candidate which seems credible theoretically is coherent diffusion, where rapid tunneling of the μ^+ is broken up by some sort of scattering mechanism or inhibited by crystalline disorder. The original prediction[23] that coherent diffusion limited by thermal phonon scattering would lead to a T^{-9} temperature dependence caused some doubt as to whether it could really explain our results and those of Ref. 22, neither of which show such a strong temperature dependence. Recent work[24] has shown, however, that a weaker ($D_{coh} \propto T^{-1}$) temperature dependence results if electron-muon scattering is assumed to be the influence which interrupts the coherent transport. This is encouraging in itself, but the absolute magnitude obtained for the diffusivity is several orders of magnitude different from the value obtained in Ref. 22. I am not aware of an attempt to quantitatively evaluate this theory in the case of copper. Here it should be noted that the possibility of coherent diffusion in copper has itself been called into question,[25] based on the expected strains due to residual interstitial impurities at the few dozen ppm level.

Faced with such a disparity between reasonable theoretical efforts and the available data, it is perhaps best to take a step back from the details and ask some questions about the general physics of the problem. What other physics happens in copper at these temperatures? What types of processes could lead to a diffusion of the kind we seem to see? What types of processes could inhibit this diffusion?

Thinking in this way, we note that we are again encouraged in considering the electron-muon scattering because the neighborhood of a few degrees K is where the electronic processes begin to dominate over thermal phonon processes. For example, the specific heat due to electrons is near that due to phonons in this regime.[26] Generally, a phonon assisted process would have to yield a diffusion rate that increases with temperature, so such a mechanism would be most attractive to explain the data above the 10-80 K plateau in the linewidth. If the diffusion mechanism is assumed to be limited, through the Anderson mechanism or otherwise, by disorder (e.g. random strain) then such disorder will be temperature independent in the case of impurity strain, but will increase with temperature for thermal phonon strain. The zero-field data imply that whatever the actual mechanism of diffusion is below 5 K, it yields faster hopping at lower temperatures. In the absence of inibiting effects such a trend may continue to very low temperatures, but it seems that it must become inhibited by either static disorder or electron scattering below 0.5 K, and possibly by thermal disorder above about 5 K.

The case of copper is by no means closed, although it may have appeared that way three or four years ago. More experiments must be done, especially the zero-field measurements below 0.5 K and between 10 K and 80 K. It is hoped that improvements to the experimental techniques, e.g. the use of wire chamber tracking to reduce the background, will allow much better data to be obtained than we have so far. But at present the theory does not even explain the data qualitatively, so there is no lack of room for effort in that domain either.

REFERENCES

[1] E. Karlsson, Phys. Repts. **82** (1982) 271.

[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** (1980) 337.

[3] V.G. Grebinnik, I.I. Gurevich, V.A. Zhukov, A.I. Klimov, V.N. Majorov, A.P. Manych, E. V. Mel'nikov, B.A. Nikol'skii, A.V. Pirogov, A.N. Ponomarev, V.I. Selivanov and V.A. Suetin, Pisma Zh. Eksp. Teor. Fiz. **25** (1977) 322.

[4] H.K. Birnbaum, M. Camani, A.T. Fiory, F.N. Gygax, W.J. Kossler, W. Rüegg, A. Schenck, and H. Schilling, Phys. Lett. **65A** (1978) 435, and Phys. Rev. **B17** (1978) 4143.

[5] A.T. Fiory, K.G. Lynn, D.M. Parkin, W.J. Kossler, W.F. Lankford and C.E. Stronach, Phys. Rev. Lett. **40** (1978) 968.

[6] R.H. Heffner, J.A. Brown, R.L. Hutson, M. Leon, D.M. Parkin, M.E. Schillaci, W.B. Gauster, O.N. Carlson, D.K. Rehbein and A.T. Fiory, Hyp. Int. **6** (1979) 237.

[7] M. Borghini, T.O. Niinikoski, J.C. Soulie, O. Hartmann, E. Karlsson, L.O. Norlin, K. Pernestål, K.W. Kehr, D. Richter, and E. Walker, Phys. Rev. Lett. **40** (1978) 1723.

[8] T.O. Niinikoski, O. Hartmann, E. Karlsson, L.-O. Norlin, K. Pernestål, K.W. Kehr, D. Richter, E. Walker, and K. Schulze, Hyp. Int. **6** (1979) 229.

[9] C. Boekema, R.H. Heffner, R.L. Hutson, M. Leon, M.E. Schillaci, W.J. Kossler, M. Numan and S.A. Dodds, Phys. Rev. **B26** (1982) 2341.

[10] 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** (1972) 143.

[11] V.G. Grebinnik, I.I. Gurevich, V.A. Zhukov, A.P. Manych, E.A. Mel'eshko, I.A. Muratova, B.A. Nikol'skii, V.I. Selivanov and V.A. Suetin, Sov. Phys.-JETP **41** (1976) 777 [Zh. Eksp. Teor. Fiz. **68** (1975) 1548].

[12] H. Teichler, Phys. Lett. **64A** (1977) 78.

[13] M. Camani, F.N. Gygax, W. Rüegg, A. Schenck and H. Schilling, Phys. Rev. Lett. **39** (1977) 836.

[14] O. Hartmann, L.-O. Norlin, A. Yaouanc, J. Le Hericy, E. Karlsson and T.O. Niinikoski, Hyp. Int. **8** (1981) 533.

[15] O. Hartmann, E. Karlsson, L.-O. Norlin, D. Richter and T.O. Niinikoski, Phys. Rev. Lett. **41** (1978) 1055.

[16] A. Seeger, Phys. Lett. **93A** (1982) 33.

[17] K.G. Petzinger, Phys. Lett. **75A** (1980) 225.

[18] R.S. Hayano, Y.J. Uemura, J. Imazato, N. Nishida, T. Yamazaki and R. Kubo, Phys. Rev. **B20** (1979) 850.

[19] C.W. Clawson, K.M. Crowe, S.E. Kohn, S.S. Rosenblum, C.Y. Huang, J.L. Smith and J.H. Brewer, Proceedings of the 16th Low Temperature

Physics Conference, Los Angeles, Ca., August 19-26, 1981, Physica **109 & 110B** (1982) 2164.

[20] C.W. Clawson, Ph.D. thesis, U.C. Berkeley, 1982. Lawrence Berkeley Laboratory Nuclear Science Division report LBL-14489, July, 1982.

[21] J.H. Brewer, Hyp. Int. **8** (1981) 831.

[22] K.W. Kehr, D. Richter, J.-M. Welter, O. Hartmann, E. Karlsson, L.O. Norlin, T.O. Niinikoski and A. Yaouanc, Phys. Rev. **B26** (1982) 567.

[23] Yu. Kagan and M.I. Klinger, J. Phys. **C7** (1974) 2791.

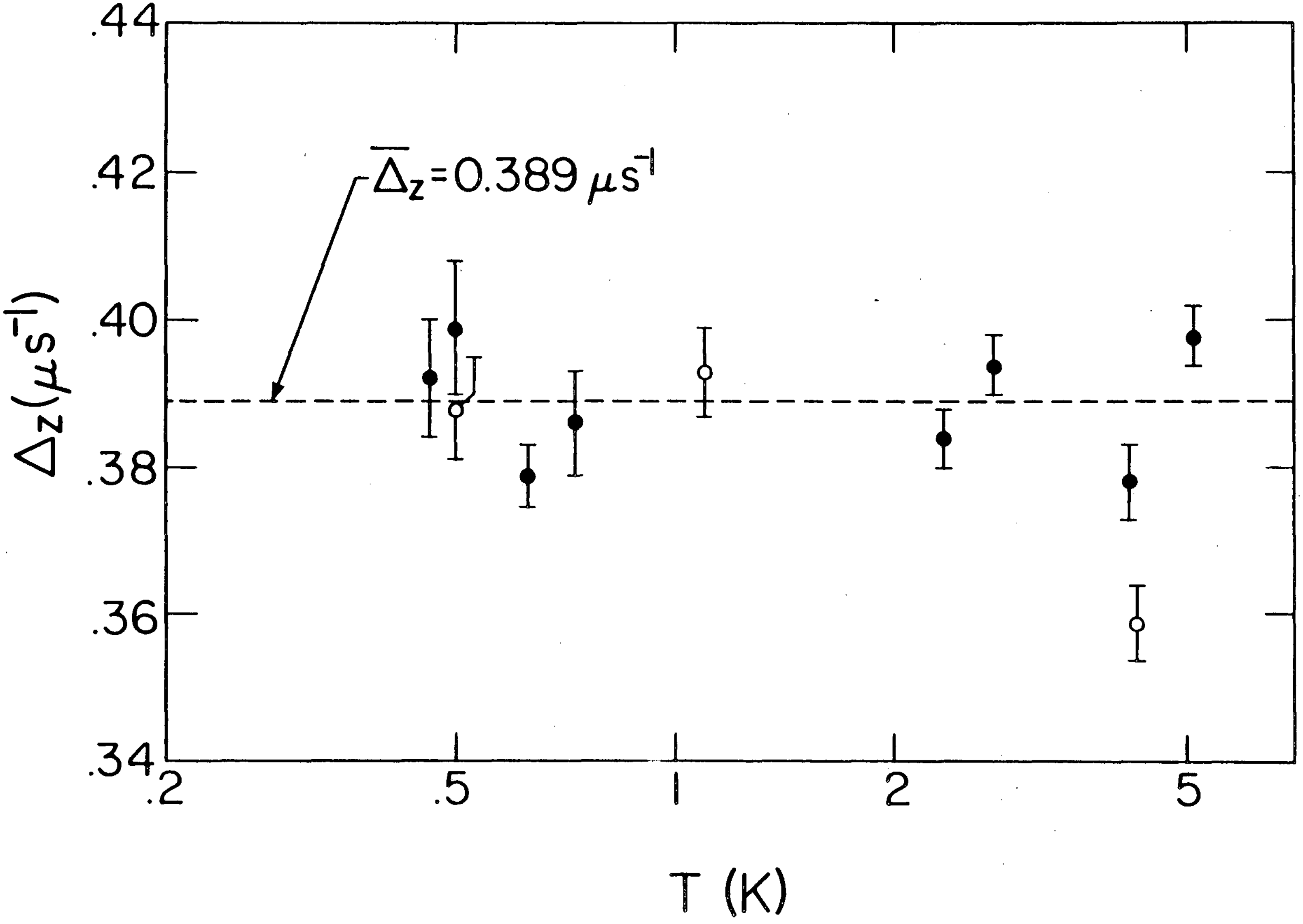
[24] J. Jäckle and K.W. Kehr, preprint, 1982.

[25] J. Chappert, A. Yaouanc, O. Hartmann, E. Karlsson, L.O. Norlin and T.O. Niinikoski, Solid State Commun. **44** (1982) 13.

[26] C. Kittel, *Introduction to Solid State Physics*, 4th ed. (Wiley, New York, 1971) chapters 6 and 7.

Figure Caption

(1) Zero-field dipolar width vs. temperature. The open points are polycrystalline copper, and the filled points are single crystal copper.



XBL 824-529

Fig. 1

TECHNICAL INFORMATION DEPARTMENT
LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720