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Studies of μ^+ Localization in Cu, Al, and Al Alloys in the Temperature Interval 0.03–100 K

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Muon spin-rotation studies on the purest available samples of Al and Cu and on $AlMn_x$ (x = 1300, 57, and 42 ppm) down to temperatures around 50 mK show that the muons are delocalized in pure Al but increasingly localized with increasing Mn content. The temperature variations are discussed in terms of strain localization and trapping in the vicinity of the impurities. For Cu, partial delocalization is observed below 2 K, which is probably limited by elastic strains from remaining impurities.

Two years ago it seemed to be true that muons are localized in all metals except aluminum at the lowest temperatures measured (2 K). For instance, the prototype experiment on copper[1] showed that muons are mobile above 100 K and at lower temperatures are localized at fixed interstitial sites, later identified as octahedral.[2] Experiments carried out a year ago demonstrated that muons can also be localized in aluminum by adding substitutional impurities[3,4] or by creating defects by irradiation[5] or thermal treatment.[6] One remaining question is whether the muon is "frozen in" at still lower temperatures in pure aluminum.

The above-mentioned investigations on aluminum and many other studies indicate the importance of any kind of defect in the question of the localization of the muons. Hence the question arises whether the localization of muons in other materials, notably copper, might be due to the impurity content of the samples investigated so far. We have tried to address ourselves to this question by investigating very pure polycrystalline copper, down to a temperature of about 0.05 K, in addition to aluminum. It turns out that muons still behave quite differently in these two metals.

The experiments were performed at the 600-MeV synchrocyclotron at CERN, using a conventional muon spin-rotation setup with a transverse magnetic field arrangement. Details of the layout can be found in Ref. 7.

Measurements down to 2 K were performed in a continuous-flow ^{4}He cryostat, and temperatures down to 30 mK were obtained with a ^{3}He-^{4}He dilution refrigerator. The sample temperature was

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measured using calibrated carbon resistors below about 30 K and platinum resistors above 30 K. The temperature was stabilized by feedback to a heater during the measurements.

The experimental muon spin-rotation data were analyzed with the usual time-dependent expression

$$N = N_0 \exp(-t/\tau_\mu)[1 + P(t)\cos(\omega t + \varphi)] + \text{background}.$$

The data were fitted with either a Gaussian damping, $P(t) = P_0 \exp(-\sigma^2 t^2)$, or an exponential damping, $P(t) = P_0 \exp(-\lambda t)$, but there was no preference for one or the other. In the figures of this Letter we have plotted the damping parameter (or linewidth) obtained from the Gaussian fit. The fit interval was 7 μsec, and each spectrum contained $(1-2)\times 10^6$ events. All spectra were corrected for background arising from muons stopping in the sample holder and the cryostat walls. This was done by running stainless steel dummy samples, of mass and size equal to the samples themselves, below 20 K, where the signal from the steel itself vanishes.

The starting material for the polycrystalline Al sample and the *Al*Mn alloys from Jülich was nominally 99.9999+% pure Al with resistivity ratio > 30 000 from VAW Leichtmetallwerke GMBH, Bonn. The *Al*Mn single crystals were grown from a melt with use of the Bridgman method. The purity and the Mn contents were checked with mass, emission, and atomic adsorption spectroscopy, which also showed the main residual impurities, Fe and Cu, to be < 1 ppm each.

The starting material for the polycrystalline Cu sample from Vitry was American Smelting and Refining Co. (ASARCO), Cu (99.9999+%). It was purified by electrolysis (soluble anode process) in a solution of concentrated copper nitrate.[8] The cathode was melted in a crucible of high-purity graphite in a vacuum of 10^{-6} Torr. The residual resistivity ratio was about 6600 and the content of (C, N, and O) interstitial impurities was estimated to be 20 at. ppm and the substitutional impurities less than 1 ppm.

The measurements of the linewidth in the pure polycrystalline aluminum sample are shown in Fig. 1. The damping of the muon spin-rotation precession signal is very small, and it is practically impossible to determine the specific shape of the line. Here we have plotted the linewidth parameter σ in order to compare with the results from polycrystalline copper included in the figure. The linewidth for aluminum is very close to zero at all temperatures, but a slight increase is seen below 100 mK. The data indicate a very mobile or extended muon in pure Al down to 30 mK, since the nuclear dipole interaction is averaged out.

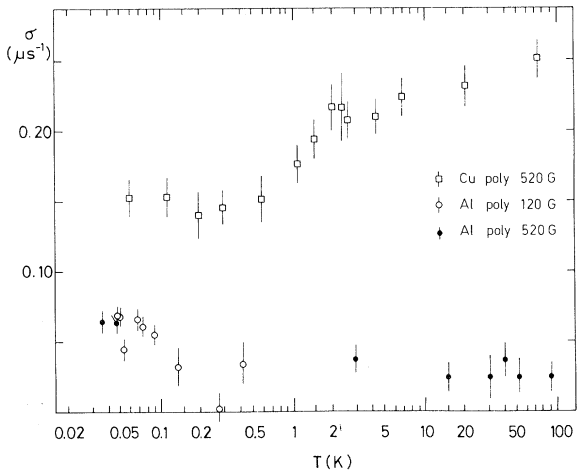

FIG. 1. Damping parameter σ for polycrystalline Al and Cu as function of temperature.

The results for the purified copper polycrystalline sample are also shown in Fig. 1. There is a significant decrease of the linewidth below 2 K, indicating a possible delocalization of the muon. All temperature points were taken in the same dilution refrigerator. Since the necessary background corrections were quite large, the possibility of a systematic error in the magnitude of σ cannot be ruled out. This would not, however, affect the general trend of the curve, which is certainly not a flat plateau. Our σ values in the region 10–75 K are also in good agreement with other data on copper.[1,2,9]

A decrease of σ at low temperatures was also observed in a measurement at 1170 G on a large Cu single crystal previously used as a neutron monochromator.[10] Here the [100] linewidth drops from 0.253(5) μs^{-1} in the 40–80-K region to 0.217(6) μs^{-1} at 2 K, while in the [111] direction σ changes from 0.124(5) to 0.108(6) μs^{-1} at 2 K.

The temperature dependence of the linewidth in Al samples doped with Mn shows a peak around 15 K. Results from a single-crystal sample with 1300 ppm Mn measured with the magnetic field in the [111] direction, and from a polycrystalline 42 ppm sample, are shown in Fig. 2. In the 1300-ppm sample the linewidth stays fairly constant below 5 K, while the 42-ppm sample has a very

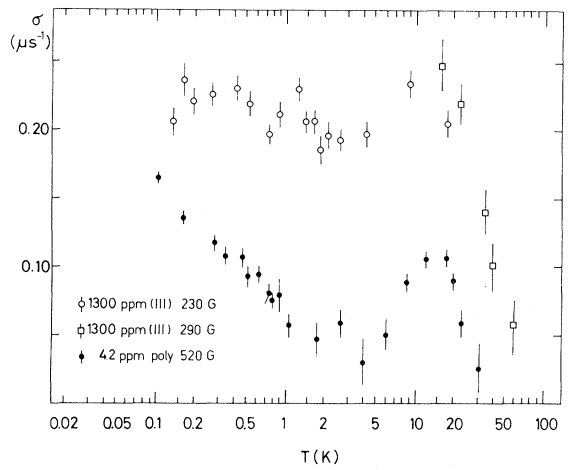

FIG. 2. Damping parameter σ for Mn-doped Al samples as function of temperature.

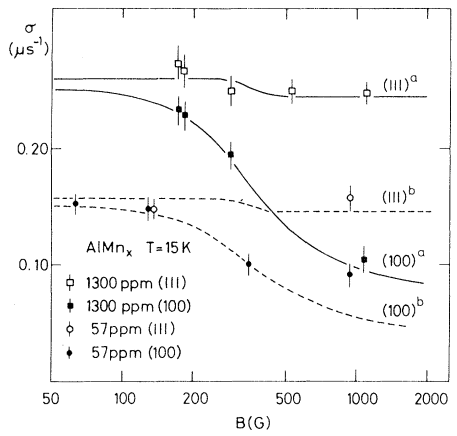

FIG. 3. Damping parameter σ as function of magnetic field. The lines are theoretical curves for tetrahedral muon sites. The full lines are reduced by a scale factor taking into account a lattice dilation of 7%. The dashed lines are reduced by a 50% scale factor.

pronounced minimum around 3 K. The maximum σ at 15 K is also much smaller in the 42-ppm sample, 0.11 μs^{-1} at 520 G and 0.13 μs^{-1} at 130 G external field. A similar σ value (0.14–0.15 μs^{-1}) was also obtained at 15 K and 130 G field for a single crystal sample containing 57 ppm Mn.

The field dependence of the muon spin-rotation linewidth in *Al*Mn single-crystal measurements is shown in Fig. 3. The data from the 1300-ppm sample and the corresponding theoretical curves were published previously.[11] It is clear that the muons occupy tetrahedral sites at temperatures around 15 K, but a new result is that the [111] linewidth is field dependent at 130 mK [510 G: 0.170(7) μs^{-1}; 230 G: 0.206(10) μs^{-1}] and at 250 mK [510 G: 0.171(11) μs^{-1}; 230 G: 0.200(6) μs^{-1}].

Therefore, the muon site is not tetrahedral at these low temperatures.[12] An indication of this fact was found also in Ref. 3, where the pure tetrahedral behavior was seen to partly disappear on decreasing the temperature from 15 K to 5 K. In the 57-ppm sample the field dependence at 15 K indicates a site with tetrahedral features, although no quantitative agreement exists. The small values of σ show that either several sites are involved or only a fraction of the muons find these sites.

In ideal crystals where the potential seen by a muon is perfectly periodic, the propagation of the muon is described by the quantum theory of diffusion.[13-15] At low temperatures coherent diffusion is expected to dominate, similar to band propagation, although the mean free path need not be large compared to the lattice constant.[15] At higher temperature incoherent hopping sets in. There are essentially three parameters (tunneling matrix element J, activation energy E_α for incoherent hopping, and decay rate Ω of extended states) which are not well known for muons in Al. Hence the contributions from the two processes and the transition temperature between them is difficult to estimate. If the magnitude of the diffusion constant at the transition temperature is sufficiently large, the transition from coherent to incoherent diffusion with increasing temperature cannot be detected in a muon spin-rotation experiment.

We consider the observation of very low damping parameters in our purest Al sample as proof of the existence of the coherent process at low temperatures. As discussed earlier,[3] this conclusion is based on a comparison of the observed correlation times τ_c (now down to 0.03 K) with those expected from the incoherent diffusion mechanism.[14] Since thermal activation is necessary for these processes, they cannot explain values of τ_c of about 10^{-8} s at these temperatures without the assumption of unrealistically large tunneling matrix elements. Considerations of a one-phonon process[16] also lead to inconsistencies. For this process, as for coherent diffusion, the effective tunneling matrix element, $J_{\mathrm{eff}} = J \exp(-s)$, has to be taken into account, where the exponential corrects for polaron effects.[17] Following Ref. 16, we find at 30 mK that $J_{\mathrm{eff}} = 110$ μeV, which is much bigger than the

mean disturbances due to strains [$W_{Al} \approx 10^{-2}$ μeV (Ref. 3)]. Thus the one-phonon process is not relevant. We note that recent experiments[18] in very pure Nb also show too small a damping at low temperatures to be explained by incoherent processes only.

The Cu data indicate a transition from a localized to a more extended or mobile state of the muon when the temperature goes below 5 K, but an explanation of the reduced linewidth in terms of motional narrowing in the range 0.05–0.5 K seems unlikely since the linewidth is approximately temperature independent. It is known[2] that the muons localize at octahedral sites in the Cu lattice at 20 and 80 K. Our single-crystal experiments at 2 K show essentially the same field dependence as at 80 K, but with a reduced linewidth.

For the discussion of the localized character of the muon wave functions in our Cu and Al samples, we can make the following observations: The tunneling matrix element J and the activation energy E_a for Cu can be estimated by fitting the theory for incoherent hopping to muon spin-rotation data, as has been done for Cu by Teichler.[19] Using new data for our Cu sample above 100 K (not shown), we find $J \approx 32$ μeV and $E_a \approx 87$ meV. With these values, the effective J_{eff} is 0.01 μeV, which is rather small compared to the average energy shift $W \approx 5$ μeV produced by strain effects of the residual impurities in our Cu sample. Thus we expect to be in the region of localized behavior, using Anderson's criterion.[20] In addition, isotope effects (Cu is an isotope mixture while Al is not) may further suppress coherent processes, since the lattice can no longer be considered as ideally periodic. In the case of Al, none of the required parameters is known, but it is likely that J is considerably larger and/or E_a smaller than for Cu so that $J_{eff} > W_{Al}$.

For the *Al*Mn alloys, one of our main observations is that the maximum of σ appears at the same temperature ($\approx$ 15 K) for different concentrations (see Fig. 2), whereas the height is concentration dependent. A comparison with other experiments[4,21] and preliminary data from a 100-ppm *Al*Mg sample of our own show that for each different substitutional impurity the maximum occurs at a characteristic temperature. These observations are consistent with a picture of trapping of muons on definite sites, characterized by distinct binding energies for each kind of defect, but are less consistent with the picture of strain-induced localization. Further information on the trapping sites has been provided by the field dependence discussed in the last section, namely that these are tetrahedral sites, not too near to the Mn atoms.[3] Adopting the trapping picture, we explain the decrease of damping above 15 K by detrapping, whereas below 15 K we see motion towards the traps in the weakly doped samples. The observed tendency of a transition from octahedral to tetrahedral sites for the muons on increasing the temperature is probably connected with this motion. The low-T behavior of the 1300-ppm sample is most likely due to strain-induced localization, as suggested earlier.[3] For this sample most of the muons experience long-range strain fields of the type considered by Leibfried,[22] which are strong enough to prevent coherent motion.

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