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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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ABSTRACT

μSR 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 can be understood as combinations of strain localization and trapping in the vicinity of the impurities. For Cu, we observe partial delocalization below 2K, which is probably limited by elastic strains from remaining impurities.

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Two years ago it seemed to be true that muons are localized in all metals except aluminum at the lowest temperatures measured (2K). 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 the 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, Geneva using a conventional μSR 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 continous-flow ^{4}He cryostat, and temperatures down to 30 mK were obtained with a ^{3}He-^{4}He dilution refrigerator. The sample temperature was 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 μSR data were analyzed with the usual time-dependent expression $N = N_o \exp(-t/\tau_\mu) \left[1 + P(t)\cos(\omega t+\phi)\right]$ + background. The data were fitted with either a Gaussian damping $P(t) = P_o \exp(-\sigma^2 t^2)$ or an exponential damping $P(t) = P_o \exp(-\lambda t)$. In this letter we have used extensively the damping parameter (or linewidth) obtained from the Gaussian fit. The fit interval was 7 μsec, and each spectrum contained $(1 - 2)\cdot 10^6$ events.

All spectra were corrected for background arising from muons stopping in the sample holder and the cryostat walls. This was done by running dummy samples of stainless steel below 20 K, where the signal from the steel itself vanishes.

The starting material for the polycrystalline Al sample and the AlMn alloys from Jülich was nominally 6 N pure Al with resistivity ratio >30 000 from VAW. The AlMn single crystals were grown from a melt using 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 ASARCO Cu (5N). 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 interstitial impurities was estimated to be 20 ppm at.

The measurements of the linewidth in the pure (6N) polycrystalline aluminum sample are shown in Fig. 1. The damping of the μSR 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.

The results for the purified copper polycrystalline sample are also shown in Fig. 1. There is a clear decrease of the linewidth below 2 K, indicating a possible delocalization of the muon. 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 1100 G on a Cu single crystal previously used in other studies /10/. Here the [100] linewidth drops from 0.25 μs^{-1} at 80 K to 0.22 μs^{-1} at 2 K, while in the [111] direction σ changes from 0.13 to 0.11 μs^{-1}.

The temperature dependence of the linewidth in aluminum 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 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 manganese.

The field dependence of the μSR linewidth in AlMn single crystal measurements is shown in Fig. 3. The data from the 1300 ppm sample and the corresponding theore-

tical curves were published previously /11/. It is clear that the muons occupy tetrahedral sites at temperatures around 15 K, but a lattice dilatation of ≈ 7 % is needed to fit the absolute values of σ with theory. The absolute value of σ can probably also be explained by an appropriate extension of the muonic wave function /12/. A new result is that the [111] linewidth is field dependent at 150 mK. Therefore, the muon site is not tetrahedral at these low temperatures /13/. An indication of this fact was found also in ref. /3/, where the pure tetrahedral behaviour 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 that only a fraction of the muons find these sites.

It is important to realize that every muon starts at a definite site in a localized state. At the end of the implantation process, a muon comes to rest at a certain lattice site, but there is a local lattice heating lasting roughly 0.1 - 1 ns /14/which for a short time interval prevents coherent diffusion processes. After this period the muon wave function develops in space and time. The resulting diffusion process will be discussed separately for ideal and non-ideal crystals.

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 /15 16,17/. At low temperatures coherent diffusion predominates, as in band propagation, although the mean free path need not be large compared to the lattice constant /17/. 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.

Hence the magnitudes of both 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 μSR-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 /16/. Since thermal activation is necessary for these processes, they cannot explain values of τ_c of about 10^{-8} s at these temperatures without assuming unrealistically large tunneling matrix elements. We note that recent experiments in very pure Nb /18/ 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 state of the muon when the temperature goes below 5 K. The linewidth is approximately temperature-independent between 0.05 and 1 K. Hence an explanation of the reduced linewidth in terms of motional narrowing seems unlikely. 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. Qualitatively, it is possible to fit our low-temperature data by assuming a distribution of the muon density with a small fraction residing on the eight neighboring octahedral sites. This fraction would be ∿10 % at 2 K and ∿30 % below 1 K.

The localization of the muons in the 2 - 80 K range in the purest available Cu and the apparent limited extension of the muonic wave function below 2 K indicate that some essential parameter(s) prevent extension over larger volumes and/or coherent propagation, in contrast to the situation in Al. At the moment, 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 μSR data, as has been done for Cu by Teichler /19/. Using new data for our Cu sample above 100 K (not shown), we find $J \approx 32$ meV and $E_a \approx 95$ meV. Using these values, the effective tunneling matrix element, reduced by small polaron effects is estimated as $J_{eff} = J \exp(-S)$, where in the Debye approximation, at $T = 0$, $S = 2.63\ E_a/k\theta_D$. We find $J_{eff} = 0.02$ 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 behaviour, 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. Unfortunately we cannot compare with the case of Al, since none of the required parameters is known. In Al, either J must be considerably larger and/or E_a smaller than for Cu in order to lead to $J_{eff} > W$.

For the AlMn alloys, one of our main observations is that the maximum of σ appears at the same temperature (≈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 AlMg sample of our own show that for each different substitutional impurity the maximum occurs at a characteristic temperature. This temperature increases for defects with increasing volume dilatation

in Al. These observations are consistent with a picture of trapping of muons on definite trapping 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. We do not know whether the motion is coherent or incoherent but remark that coherent motion is increasingly likely at lower temperatures. The low-T behaviour 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.

The increase in damping in the 42 ppm sample for the lowest temperatures could be due to the following process: J_{eff} might be somewhat smaller than the average energy shift W in this concentration range, which would lead to localization at very low temperatures. Thermal motion could allow the onset of coherent processes, since the static disorder can be momentarily compensated by thermal fluctuations. This would lead to an increase in the diffusion rate D with temperature /23/. The increase is followed by a drop in D with increasing temperature, when the thermal fluctuations become so large as to impede the coherent transfer processes. Thus we arrive at a rather consistent picture for the behaviour of the damping rate in the 42 ppm sample and in the other samples.

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References and Footnotes

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23) One of the authors (K.W.K.) thanks Prof. Yu. Kagan for pointing out this possibility to him

Figure captions

Fig. 1. Damping parameter σ for polycrystalline Al (circles) and Cu (squares) as function of temperature.

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 dilatation of 7 %. The dashed lines are reduced by a 50 % scale factor.

σ (μs⁻¹)
0.20
0.10
Cu poly 520 G
Al poly 120 G
Al poly 520 G
0.02
0.05
0.1
0.2
0.5
1
2
5
10
20
50
100
T(K)

Fig. 1

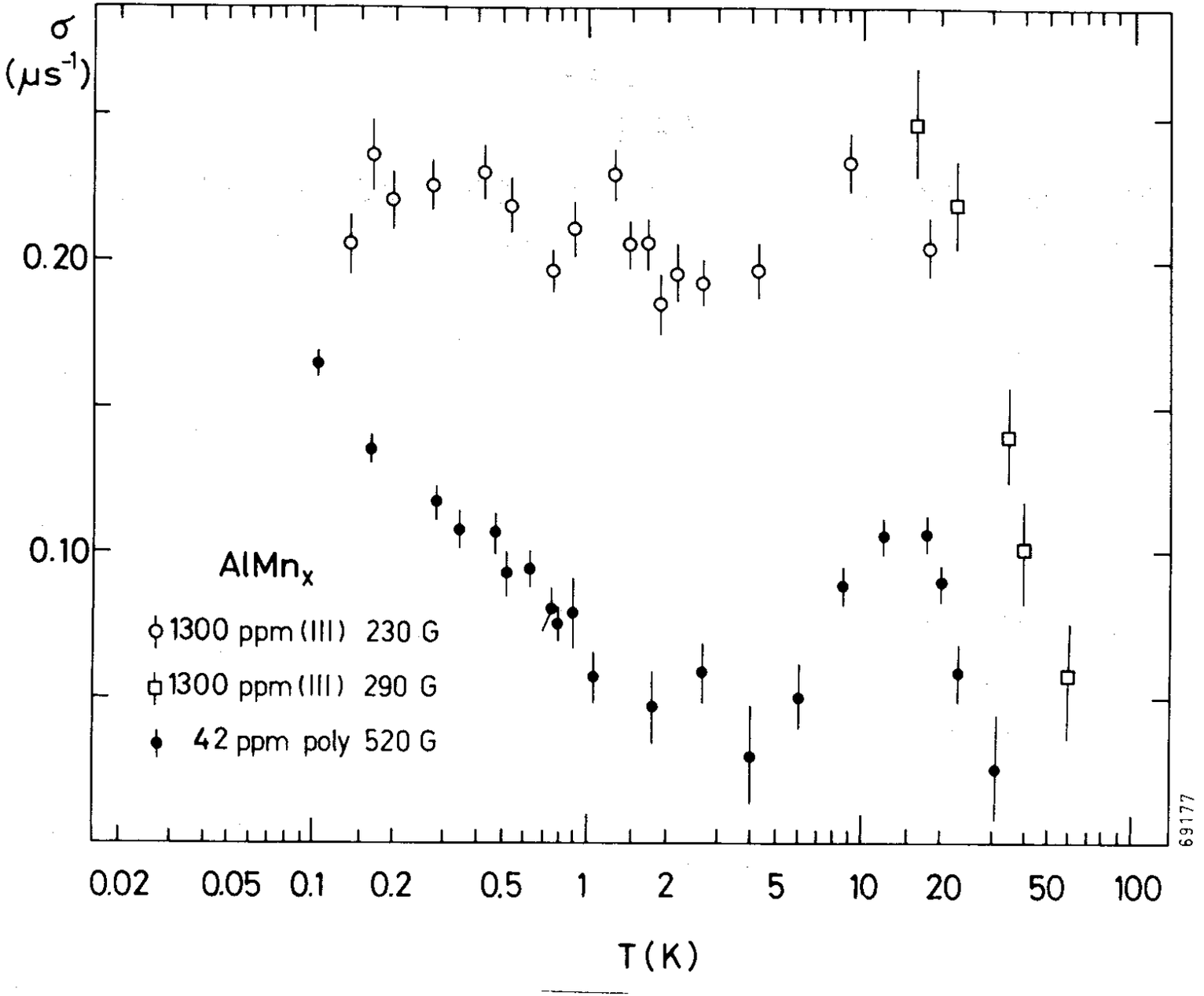
σ
(μs⁻¹)
0.20
0.10
AlMnx
1300 ppm (III) 230 G
1300 ppm (III) 290 G
42 ppm poly 520 G
0.02
0.05
0.1
0.2
0.5
1
2
5
10
20
50
100
T(K)
69177

Fig. 2

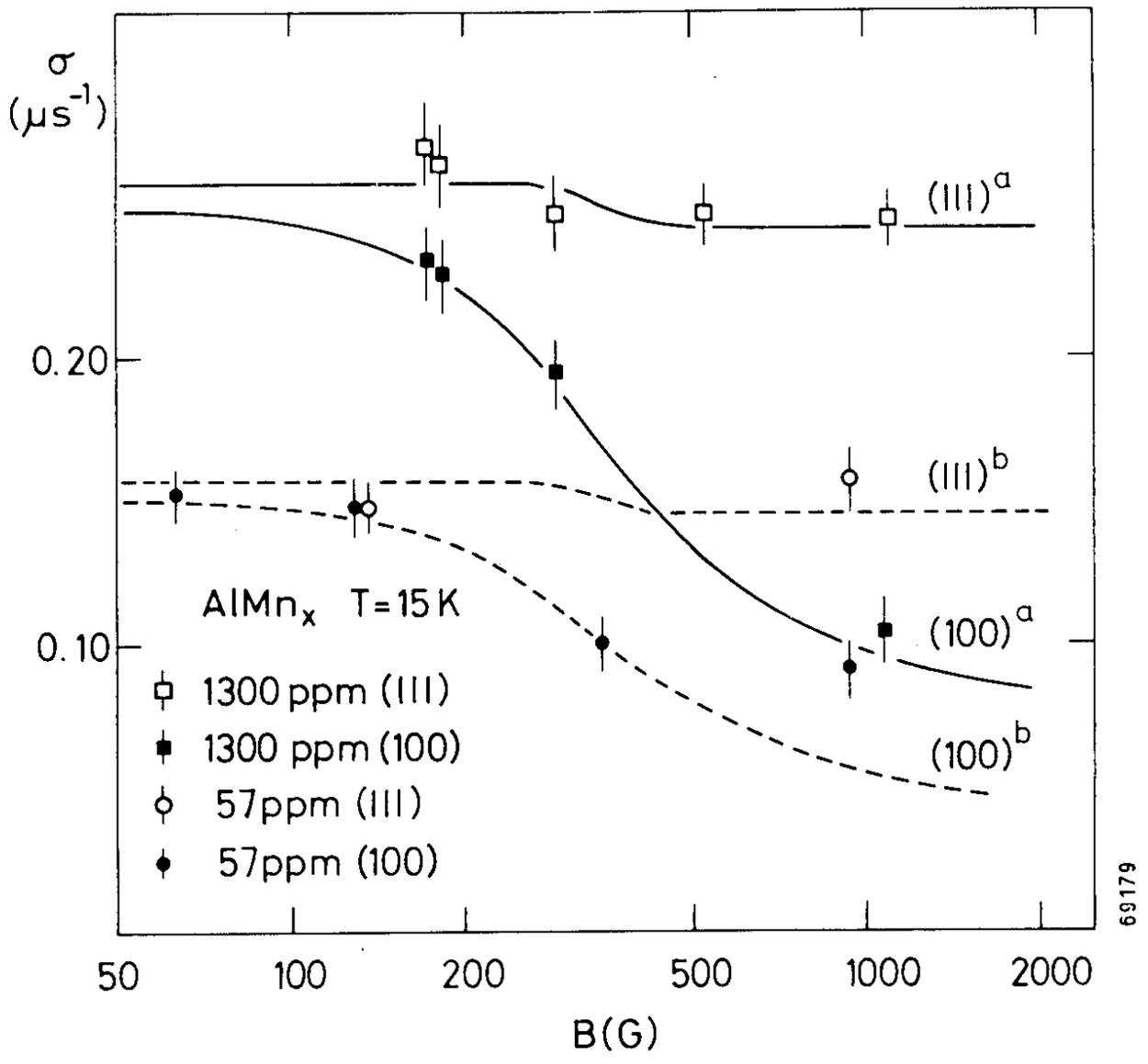
σ
(μs⁻¹)
0.20
0.10
AlMnx T=15K
1300 ppm (111)
1300 ppm (100)
57ppm (111)
57ppm (100)
(111)a
(111)b
(100)a
(100)b
50
100
200
500
1000
2000
B(G)
69179

Fig. 3