Coherent Propagation and Strain-Induced Localization of Muons in Al

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Positive muons implanted in single-crystal Al doped with Mn are localized below 20 K in contrast to the case of pure Al, where the muon is completely delocalized down to 1 K. Applying the Anderson theory of localization, we show consistently that a coherent tunneling state, which exists in pure Al, is destroyed under the influence of lattice strains generated by the Mn impurities. Our data suggest strongly that the muons are localized at tetrahedral sites at 15 K.

The muon-spin-rotation method has shown that it can be used to determine diffusion properties of positive muons in various metals. The muons have, for example, been found to be localized at low temperatures in Cu,[1] Nb,[2] Ta,[3] and V.[4] A remarkable exception to the general behavior is Al, where the muons appear to be mobile at all temperatures down to 1 K.[2,5] In this paper we report measurements on AlMn alloys with 500 and 1300 ppm Mn. The muon-spin-rotation linewidth measurements show clearly a localization of the muon below 20 K. At 15 K the field dependence of linewidth for different crystal orientations indicates strongly that the muons occupy tetrahedral interstitial sites.

Following early reports on localization and diffusion of positive muons in Cu and Nb,[1,2] it was observed that the low-temperature behavior of the linewidth often does not follow the flat curve to be expected and indeed found for Cu. The appearance of "dips" in the linewidth curves in V,[3,4] Bi,[6] and Nb[7,8] has been interpreted either in terms of quantum diffusion or trapping of muons by impurities. The question of trap-induced localization is interesting especially for the case of Al, where no evidence of self-trapping has been found. Our present observation of a linewidth in Al doped with Mn shows that a localization of the muon in this material can be induced by impurity atoms.

The experiments have been performed at the 600-MeV synchrocyclotron at CERN. Background corrections were made using data from runs on dummy samples with known performance. The Al single crystals were grown from the melt us-

ing the Bridgman method. As starting materials 6N Al and 4N Mn were used. At room temperature and below, Mn is practically not soluble in Al. Diffuse neutron scattering experiments on $AlMn_{0.0013}$, however, show only a small decrease ($< 10\%$) of the Laue scattering due to precipitation of Mn atoms in going from 800 to 300 K.[9,10] Since the Laue scattering is proportional to the number of randomly distributed defects, this indicates that the statistical high-temperature distribution is frozen in at room temperature.

The temperature dependence of linewidth Λ observed in the muon-spin-rotation measurements on polycrystalline pure Al (6N) and on $AlMn_x$ ($x = 500$ ppm, 1300 ppm) single crystals is shown in Fig. 1. While Λ is zero within the experimental accuracy for pure Al, both $AlMn_x$ crystals exhibit
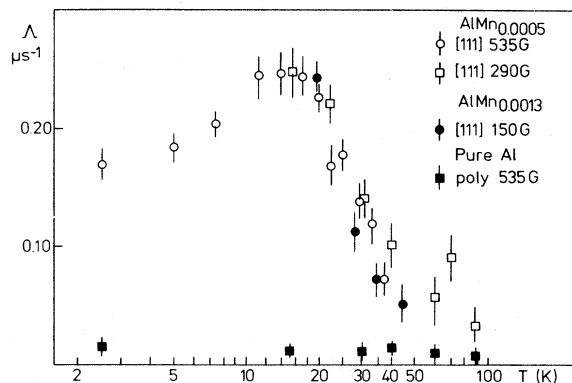

FIG. 1. The linewidth parameter Λ as a function of temperature for $AlMn_x$ and pure Al.

the same behavior showing localization up to about 20 K (Gaussian line shape). The linewidth parameter Λ is defined as the inverse time for the asymmetry to decay to $1/e$ of the initial value. In evaluating Λ and the correlation time τ_c from the experimental spectra, the usual Abragam formula

$$a(t) = a(0) \exp\{-2\sigma^2 \tau_c^2 (\exp(-t/\tau_c) - 1 + t/\tau_c)\}$$

(1)

was employed.[1] Here the low-temperature σ was taken to be the value at 15 K.

The dependence of the low-temperature linewidth σ on applied field strength was studied for different crystal orientations at 5 and 15 K. The data for the 500- and 1300-ppm samples appear identical and are plotted in Figs. 2(a) and 2(b). The dependence of σ on B_0 is due to the electric field gradient (EFG) created by the muon, which competes with the applied magnetic field in influencing the nuclear dipoles. Theoretical linewidths for this situation have been calculated.[11] The behavior at 15 K is strong evidence that the
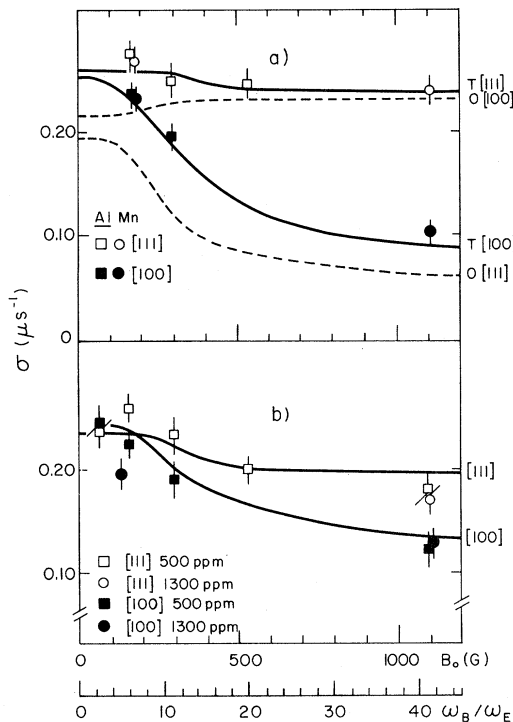

FIG. 2. Magnetic field dependence of linewidth σ.
(a) Data at $T = 15$ K. Included are the theoretical curves for tetrahedral positions (full lines) and octahedral positions (broken lines) (Ref. 11) scaled by a factor of 0.8. (b) Data at $T = 5$ K. The theoretical curves were obtained from a sum of 30% octahedral and 70% tetrahedral occupation.

muon occupies tetrahedral positions [Fig. 2(a)] in agreement with channeling results on H and D in Al.[12] For octahedral sites the orientation dependence would be reversed. The observed linewidths at 5 K suggest a dominating tetrahedral site, with a small admixture of octahedral sites [Fig. 2(b)].

In discussing the magnitude and field dependence of linewidth, we can first exclude the possibility of an electronic moment at the Mn atoms which would affect the linewidth. If AlMn can be regarded as a Kondo system, then its Kondo temperature would be 700 K,[13] giving spin fluctuations of the order of 10^{-14} s. Even if a muon sits at the nearest interstitial site (1.7 Å) to a Mn atom, the depolarization rate would be less than 0.001 μs^{-1}. As in the case of copper[1] and niobium,[2] the observed low-temperature σ values are lower than the theoretical values, which for the $(1,1,1)$ direction in Al is 0.32 μs^{-1} compared with 0.24 μs^{-1} observed at 15 K. In addition to the tendency for octahedral site occupancy as the temperature is reduced to 5 K, we observe a drop in linewidth to 0.17 μs^{-1}. While the difference between theory and experiment at 15 K may be attributed to a local lattice expansion of $\sim 10\%$, the smaller value at 5 K indicates a somewhat extended wave function.

From the field dependence of σ at 15 K an estimate for the strength of the EFG created by the muon can be deduced. The result, 0.18 Å^{-3} at nearest neighbors, i.e., 0.12 Å^{-3} at a distance of half a lattice constant, agrees well with the theoretical value of 0.13 Å^{-3} predicted by Jena et al.[14] The observed EFG at the Al nuclei is an order of magnitude smaller than that created by the Mn at nearest Al neighbors.[15] The orientational dependence is in agreement with a radial EFG created by the muon. Both results indicate a localized muon surrounded by Al host atoms.

To discuss the dynamics of the muon in AlMn$_x$ we recall that the muon is completely delocalized in pure Al down to 1 K, corresponding to muon correlation times below 10^{-8} sec at this temperature. It is very unlikely that this behavior could be explained by a classical jump process or by a tunneling hopping mechanism as developed by Flynn and Stoneham.[16] In the low-temperature regime this theory predicts a T^7 dependence of $1/\tau_c$. In the framework of a Debye model we have

$$\tau^{-1} = \frac{450\pi J^2 E_a^2}{\hbar (k_B \Theta_D)^3} \left(\frac{2T}{\Theta_D}\right)^7 \exp\left(\frac{-5E_a}{k_B \Theta_D}\right),$$

(2)

where Θ_D is the Debye temperature, E_a is relat-

ed to the self-energy of the muon in the lattice, and J is the tunneling matrix element between adjacent sites. By insertion of $\Theta_D{}^{Al} = 428$ K, Eq. (2) yields unphysically large values for J [$J > 0.4$ eV for any value of E_a in Eq. (2)]. As an alternative mechanism, we consider therefore a coherent tunneling propagation.[17]

The observation of a localized muon in AlMn$_x$ up to 20 K must be related to the influence of the Mn impurities. Apart from the fact that there is no incoherent hopping process known to provide fast enough transport to the impurities, the activation energy $E_a = 94 \pm 12$ K derived from $\ln(1/\tau_c)$ vs $1/T$ above 20 K is too small to be interpreted as the binding energy at the traps. For thermodynamical reasons, a binding energy of the order of 90 K would prevent large occupation numbers of the traps in the region between 10 and 20 K and is, therefore, inconsistent with the observed localization up to 20 K.

If coherent quantum propagation is assumed in pure Al, the localization of the muon in AlMn$_x$ would be induced by the lattice strains introduced by the Mn impurities. The elastic interaction between the strain, generated by the Mn atoms, and the muon gives rise to static shifts ΔE of the energy levels at interstitial sites. Anderson[18] has treated the localization problem for a lattice with random energy shifts ΔE with a distribution function of width Γ. He calculated a critical value Γ_c above which localization occurs. It is given essentially by the tunneling matrix element J between adjacent sites. If J is smaller than Γ_c, a coherent quantum state cannot further develop and localization occurs. Anderson's arguments agree qualitatively with those of Kagan and Klinger.[17] As a prerequisite for coherent transport they demand crystal regions where the mean energy difference ΔE between adjacent sites is smaller than the tunneling matrix element J between those sites.

As a rough estimate of the elastic muon-manganese interaction we employ the first term of a series expansion for the interaction of isotropic defects within an anisotropic cubic crystal given by Leibfried[19]:

$$E_{int} = -\frac{1}{r^3}\frac{15}{8\pi} d\left(\frac{\bar{c}_{11} + 2\bar{c}_{12}}{3\bar{c}_{11}}\right)^2$$
$$\times \Delta V^\mu \Delta V^{Mn}\left|\frac{3}{5} - \sum_j \rho_j{}^4\right|, \qquad (3)$$

where $d = c_{11} - c_{12} - c_{44}$ is the anisotropy of the lattice, $\bar{c}_{12} = c_{12} + \frac{1}{5}d$ and $\bar{c}_{11} = \bar{c}_{12} + 2c_{44} + \frac{2}{5}d$ are the averaged elastic constants, ΔV^μ and ΔV^{Mn} (see

Ref. 10) are the volume expansion induced by the defects, and ρ_j are the direction cosines with respect to the cubic axes. If we neglect the angular dependence which enlarges E_{int} slightly and insert the elastic data of Al together with volume changes calculated from the lattice expansion —for the muon we use the hydrogen value of 2.8 Å^3 found in a variety of fcc metals[20]—we obtain $E_{int} = -0.39/r^3$ eV. The energy change between adjacent sites in a distance Δr is given by

$$(dE_{int}/dr)\Delta r = 3 \times 0.39\Delta r/r^4 \text{ eV}. \qquad (4)$$

Taking into account the impurity concentration and assuming for Δr the distance between tetrahedral sites, we find as an order of magnitude estimate for the region between the defects $\Delta E \simeq 70$ μeV. This value decreases to about 10^{-2} μeV for $6N$ Al. In order to decide whether the conception of strain-induced localization is correct, we estimate the value of J from the dynamical behavior of the muon above 20 K, where the decrease of linewidth indicates the onset of diffusion.

In the $T > 20$-K range we assume phonon-assisted tunneling processes[16] as the main mechanism. The theory of Flynn and Stoneham predicts a change between a T^7 law and an exponential temperature dependence in the temperature region under consideration. Since our data can by no means be fitted by a T^7 behavior, we apply tentatively the exponential law which is a reasonable description of the observed temperature dependence of $\tau_c{}^{-1}$. We have

$$\frac{1}{\tau} = \left(\frac{\pi}{4\hbar^2 E_a k_B T}\right)^{1/2} J^2 \exp(-E_a/k_B T). \qquad (5)$$

Taking for E_a the value of 94 K given by the slope of $\ln(1/\tau_c)$ vs T^{-1}, we find that Eq. (5) yields $J \simeq 5$ μeV. The resulting tunneling matrix element is large enough to allow coherent transport in pure Al, whereas in AlMn$_x$ the lattice disturbance as estimated before is sufficient to suppress a coherent state.

We conclude now that we have here the first clear experimental evidence for the existence of coherent tunneling diffusion in pure Al and its destruction by impurities. While in pure Al coherent propagation of the muon occurs, the coherent state is destroyed in the samples containing 500–1300 ppm Mn and the muon is localized in the Al lattice up to 20 K (without being trapped at Mn atoms). Our conclusion is based on the following:

(1) The application of conventional tunneling hopping theories,[16] in order to explain the large jump rate at 1 K in pure Al, results in unrealistically large values of J.

(2) The very small activation energy derived from the τ_c data above 20 K rules out the trapping at impurities below 20 K.

(3) The relation between the strain-induced energy shift ΔE and the tunneling matrix element J of the muon fulfills consistently the requirements of the Anderson theory.[18] For the case of pure Al we find $J \gg \Delta E$.

The above interpretation is also supported by the fact that the EFG at the Al is characteristic for an interstitial muon acting on the Al rather than for a neighboring Mn atom. The absolute value of the EFG derived from the same data is close to the theoretical prediction given for pure Al metal.[14] The strain-induced self-trapping mechanism in Al is supposed to occur even at much lower concentrations of Mn than used here. In order to observe the transition between both regimes, experiments with lower Mn content are under way.

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Origin of Thermal Conductivity Anisotropy in Liquid Crystalline Phases

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Thermal conductivity measurements have been performed in nematic and smectic liquid crystalline phases by the forced Rayleigh light-scattering technique. Results show clearly that the thermal transport anisotropy is governed by the molecular shape anisotropy and by the molecular orientation but is independent of the smectic layer ordering.

Most of the past work on thermal transport has been devoted to thermal conductivity measurements in nematic phases. Despite some initial controversies, it is now accepted that the conductivity is anisotropic, being maximum (minimum) parallel (perpendicular) to the direction of the local optical axis.[1] Recently we have been able to extend the measurement down into the smectic-A phases. The preliminary results have indicated that thermal transport properties are not influenced by the regular layered structure.[2] This behavior is rather unexpected since it has been demonstrated that the smectic layers act as strong barriers to molecular motion in other transport properties as mass diffusion and ionic conductivity.[3]

In this Letter, we clearly ascertain that heat transport is primarily governed by the individual molecular properties and the orientational ordering but not by the long-range position ordering. Thermal conductivity data are presented on nematic compounds with different molecular lengths, in an attempt to vary the molecular geometrical shape anisotropy. The angular dependence of the thermal diffusivity is also investigated on both sides of a smectic-C to nematic phase transition.