

Quantum diffusion and localization of positive muons in superconducting aluminum

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The diffusion of positive muons was studied in the normal and superconducting states of aluminum. Large differences were observed, indicating that the diffusion mechanism depends sensitively on the interaction between the muon and the conduction electrons. The samples were high-purity aluminum doped with controlled amounts of lithium. The lithium impurities act as traps for the muons, leading to a loss of muon polarization. However, the time and temperature dependencies of the muon depolarization function $G(t)$ could not be satisfactorily explained using a simple diffusion limited trapping model. In particular, in the superconducting state the measured $G(t)$ requires a more complex model for a reasonable fit. These data can be described, in a first approximation, with two fractions, one corresponding to muons stopped in the defect potential created by the doping element and the other representing muons which diffuse freely without being trapped. In this model, which supports a microscopic theory by Kagan and Prokof'ev taking into account static as well as dynamic effects on the tunneling of positive particles in a conducting medium, a large fraction of the muons is practically immobile below 0.3 K in doped samples. The data can also be fitted by assuming that the spatial distribution of doping elements leads to a distribution in trapping times and that the fraction of diffusing muons is described by a so-called stretched exponential time dependence. The relative merits of these two types of interpretation are discussed.

I. INTRODUCTION

The study of diffusion of light particles in metallic matrices has been the subject of numerous studies involving a number of experimental techniques during the last three decades. Proton diffusion has been studied using, primarily, NMR and inelastic neutron scattering and these studies have been complemented with muon diffusion studies using the muon spin-rotation (μ SR) technique. In parallel with the experimental progress, a development of theoretical models has taken place. In most processes the proton motion is thermally activated and can be described in terms of an activation energy and an attempt frequency. Due to the lower mass of the muon (about 1/9 of the proton mass) its motion is more susceptible to quantum phenomena. The diffusion of muons in metals at low temperatures can therefore, under certain conditions, be dominated by quantum-mechanical tunneling. Furthermore, since the lattice can be represented as a bath of phonons and electrons with which the particle interacts, such diffusion can be regarded as an example of quantum-mechanical phenomena in

dissipative systems.^{1,2} For the interaction with the electron bath, the presence of a Fermi surface and the close connection with the orthogonality catastrophe³ is of particular interest. Earlier experimental results on muon diffusion in copper⁴ and in aluminum⁵ have demonstrated that below a certain temperature T^* (typically a small fraction of the Debye temperature), the muon diffusion rate increases with decreasing temperature according to a power law ($T^{-\alpha}$) where α is about 0.6.

This behavior is consistent with theories developed independently by Kondo⁶ and Yamada⁷ for a simple metal for which the power α is expected to lie between zero and one depending on the strength of the muon-electron coupling. In a metal the muon charge is screened through a conduction-electron redistribution. The diffusion process depends on to which extent the screening electrons can follow the motion of the muon and this is governed by the muon-electron interaction strength K . If the muon moves predominantly by tunneling, its motion is influenced by a "friction" with energy being dissipated to the electron bath.

Kondo⁶ and Yamada⁷ considered the quantum

diffusion of a “heavy” positively charged particle in a metal taking into account the interaction with the conduction electrons. Within the framework of the small polaron theory they found strong renormalization of the tunneling matrix element J (which already contains the renormalization with respect to local elastic distortions of the lattice), for the transition between neighboring equivalent sites

$$J_{\text{eff}} = J(\pi k_B T / \varepsilon_F)^K, \quad (1)$$

where ε_F is the Fermi energy. In later work by Kagan and Prokof'ev (see below) it was shown that ε_F should be replaced by the energy $\hbar\omega_0$ corresponding to the muon zero-point vibration energy (of the order of 0.1 eV). The values of K and ω_0 depend on the metal under investigation.

Electron scattering in a normal metal gives rise to a dynamical energy width $\Gamma = 2\pi K k_B T$ for the muon levels since the number of electron states at the Fermi surface available for scattering is proportional to T . With $\sqrt{z} \cdot J_{\text{eff}} < \Gamma$ (z is the coordination number; $z = 12$ for muons in an octahedral site in a fcc lattice) the quantum diffusion coefficient has the form

$$D(T) = \frac{za^2}{3} \frac{J_{\text{eff}}^2}{\Gamma} \frac{\Gamma^2(1+K)}{\Gamma(1+2K)}, \quad (2)$$

where a is the distance between octahedral sites and Γ is the gamma function. The temperature dependence $D(T)$ for a normal metal originates from a T^{2K} factor coming from the squared matrix element and a T^{-1} factor from the electrons available for scattering, i.e., totally T^{2K-1} . Therefore, the empirical parameter α corresponds to $2K-1$. Another effect of the muon-electron interaction is that, in normal metals, the coherence time is so strongly restricted by inelastic collisions with the conduction electrons that the coherence is lost after each tunneling transition.

For the superconducting state one should replace the width Γ with the expression^{8,9}

$$\Gamma_s = \{4\pi K k_B T\} \{1 + \exp(\Delta_s / k_B T)\}^{-1}, \quad (3)$$

where Δ_s is the superconducting energy gap. The electron density at the Fermi surface is then drastically reduced due to the formation of the energy gap Δ_s below T_c . The T^{-1} term in the Kondo-Yamada theory has to be multiplied by, in a first approximation, $\exp(-\Delta_s / k_B T)$. A strong influence on the diffusion rate is expected when the gap develops.

The first investigation testing this aspect of the theory was performed on Al,¹⁰ which is one of the systems most thoroughly studied by the μ SR technique and which is known to undergo a superconducting transition at 1.2 K. In pure Al, the muon spin depolarization is negligible due to the very rapid diffusion which motionally averages the ²⁷Al nuclear dipole fields. Since all information about mobility from μ SR measurements is derived from depolarization rates, it is necessary to work with a certain number of impurity atoms that trap muons to make studies of muon mobility in Al possible. For the first test a

sample with a Li concentration of 75 ppm was chosen and a marked difference in the diffusion rates of muons in aluminum in the normal and superconducting states was reported¹⁰ providing direct evidence of the important role of the conduction electrons. There was, however, insufficient data to make a quantitative comparison with theory. These measurements were later extended to complete the temperature scan as reported in Ref. 11. In the present paper we report on a more detailed study of the muon depolarization rate in lithium-doped aluminum in the normal and superconducting state.

II. EXPERIMENTS AND DATA REDUCTION

The present experiments were performed on the M15 beam line at TRIUMF which provides a beam of nearly 100% spin-polarized positive muons of momentum 28.6 MeV/c. Muons were stopped in a high-purity (6N) polycrystalline aluminum sample and also in the same material doped with nominally 75 at. ppm of Li, later analyzed to 76 (4) ppm, as well as two samples with lower Li concentration: nominally 10 and 20 ppm and with analyzed composition of 8.3 and 17.5 ppm, respectively. The 75 ppm sample was cut from the larger sample used in Ref. 10 and details of the sample preparation can be found in Ref. 12. The samples, which measured $14 \times 22 \times 5$ mm, were bolted to the cold finger of a top loading Oxford Instruments ³He-⁴He dilution refrigerator. The μ SR spectra in the normal state were taken with an external field of 13 mT (10 and 20 ppm) or 20 mT (75 ppm), applied transverse to the initial polarization direction. The critical field at $T = 0$ K is $H_c = 10$ mT so that a 13 mT field is sufficient to quench the superconductivity. Measurements in the superconducting state were made in zero applied field (the stray field at the sample position was less than 0.01 mT). Any residual stray field is automatically excluded in a type-I superconductor due to the Meissner-Ochsenfeld effect. Application of a weak transverse magnetic field of 5 mT (well below H_c) revealed a precession of low amplitude attributed to 15% of the muons stopping outside the sample. The data were fitted with this background contribution included but in the displayed spectra the background contribution has been subtracted out.

III. DATA ANALYSIS BASED ON TRAPPING MODELS

The time differential μ SR spectrum measures $A(t) = AG_{\beta\beta}(t)$, where A is an experimental asymmetry parameter determined by the properties of muon decay and detector geometry, and $G_{\beta\beta}(t)$ is a relaxation function. In order to compare the temperature dependence of the μ SR spectra in zero ($\beta = z$) and transverse fields ($\beta = x$) the data were, initially, fitted within the restricted time range (0–5 μ s) with a common relaxation function $G_{\beta\beta}(t) = \exp[-(\Lambda t)^\gamma]$, where γ is a free parameter with a value close to 2 in the zero-field data at the lowest temperatures. The depolarization rate parameter Λ may be interpreted, in a frequency representation, as a linewidth and is related to the trapping rate and the diffusion constant. In an undistorted lattice, like in very pure Al, the

linewidth is proportional to $\sigma^2\tau_c$ if $\omega\tau_c \ll 1$, where τ_c is the correlation time (of the order of the muon life time) and σ is the static dipolar broadening. For a complete evaluation of the time spectra, however, a more elaborate model has to be used (see below).

In the normal state, Λ shows a slow monotonic increase with decreasing temperature as reported previously.¹² This is shown in Fig. 1 for the 75 ppm sample. The temperature dependence of Λ in the superconducting state is clearly different. In particular, a steep increase in Λ is observed below 0.5 K and the damping rate saturates at about $0.35 \mu\text{s}^{-1}$ for temperatures below 0.2 K. (This value should be divided by $\sqrt{2}$ in order to make a comparison with the transverse field data.)

The trapping model used for the first analysis of the muon spin depolarization rates is the same as the one used for muon diffusion in impurity doped aluminum in the normal conducting state⁵ and it should be noted that, because of the trapping, an increased diffusion rate leads to a larger depolarization, which is contrary to conventional motional narrowing observations. In this model, the muons stop at random sites in the lattice and then start to diffuse with a diffusion coefficient D_μ . After a certain time t , a fraction of the muons is within a radius r_t (the trapping radius), of a lithium impurity where it is trapped and depolarizes. The remaining fraction, $P(t)$, still diffuses. According to Waite,¹³

$$P(t) = \exp\{-v_t t [1 - 2r_t(\pi D_\mu t)^{-0.5}]\}, \quad (4)$$

where $v_t = 4\pi c r_t D_\mu$ is the trapping as $t \rightarrow \infty$, and c is the impurity concentration. The muon spin-relaxation function is then given by¹⁴

$$G_{\beta\beta}(t) = P(t) - \int_0^t (dP/d\tau) g_{\beta\beta}(t-\tau) d\tau, \quad (5)$$

where $\beta=x$ if an external magnetic field is applied perpendicular to the initial polarization direction (transverse field geometry), in which case $g_{xx}(t)$ is a Gaussian function. This is because there is a Gaussian distribution of

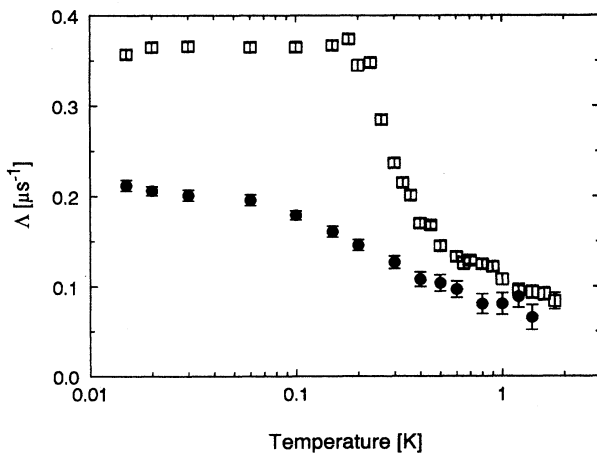


FIG. 1. The linewidth (depolarization rate) Λ for the 75 ppm sample as a function of temperature in the normal state (filled symbols) and in the superconducting state (open symbols).

randomly oriented nuclear dipolar fields at the trap site. With observation along the initial polarization direction, $\beta=z$ and zero applied field, the relaxation function takes a different form and a full recovery to one third of the initial value is expected at long times.¹⁴ Figure 2 shows examples of both zero- and transverse-field μSR time spectra at 0.2 and 0.5 K and the fits obtained using the trapping model.

The diffusion limited trapping model allows a good fit of the depolarization data for the normal conducting state as demonstrated earlier for the 75 ppm sample.¹¹ The present study shows that the model also works well at lower concentrations in the normal state. For the superconducting state, however, the evaluation of the data, based on this model, leads to relatively poor fits (in particular in the range 0.2–0.5 K, as demonstrated in Fig. 2). Also one notices immediately from Fig. 1 that the steep increase in Λ in the superconducting state occurs around 0.3 K instead of just below $T_c = 1.2$ K, which would be expected from the Kondo-Yamada theory according to Eqs. (2) and (3).

Even if the model can explain the 75 ppm data reasonably well in at least part of the temperature range the discrepancies are very pronounced for the lower Li concentrations. The μSR spectra measured at 0.10 K in the superconducting state for the different Li concentrations are presented in Fig. 3 in which the shape $A(t)$ suggests one fraction a_t of muons with the characteristic shape for immobile muons, superimposed on a flat background with amplitude a_{free} corresponding to fast moving muons. Figure 3 also shows how the data can be fitted with such a two-component model, and how the trapped fraction a_t depends on the doping concentration c . A physical basis exists for such a decomposition into a trapped and a freely diffusing fraction (the Kagan-Prokof'ev theory,^{15–17} see Sec. IV A below). This leads to the trapping rates displayed in Fig. 4. In an earlier publication by Kadono¹⁸ it was shown that the same experimental $A(t)$ data can also be fitted with a one-component trapping model in which there is a random distribution of trapping energies. The two approaches to interpret the data will be compared in Sec. IV.

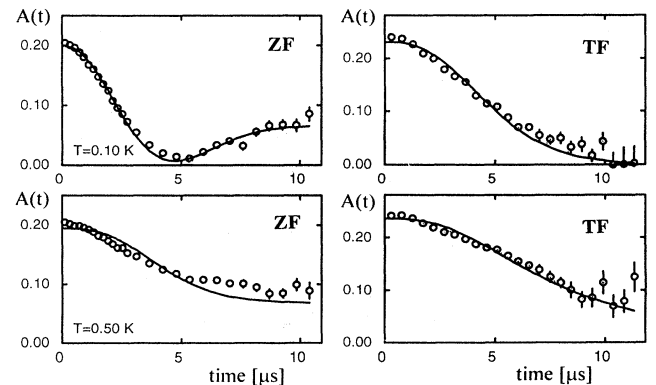


FIG. 2. Zero-field and transverse field spectra at 0.1 and 0.5 K for 75 ppm Li in Al, fitted with a simple trapping function.

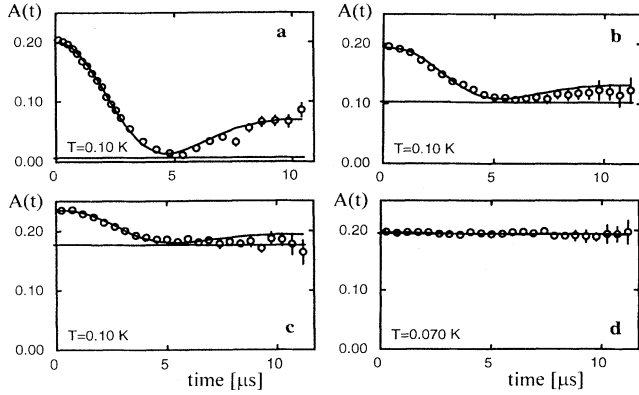


FIG. 3. Decomposition of the measured zero-field time spectra at 0.10 K into a depolarizing and nondepolarizing fraction for Li concentrations of (a) 75 ppm, (b) 20 ppm, and (c) 10 ppm. (d) The same information for high-purity aluminum ($> 6N$) at 0.07 K. The nondepolarizing fractions are < 5 , ~ 50 , ~ 75 , and ~ 100 %, respectively.

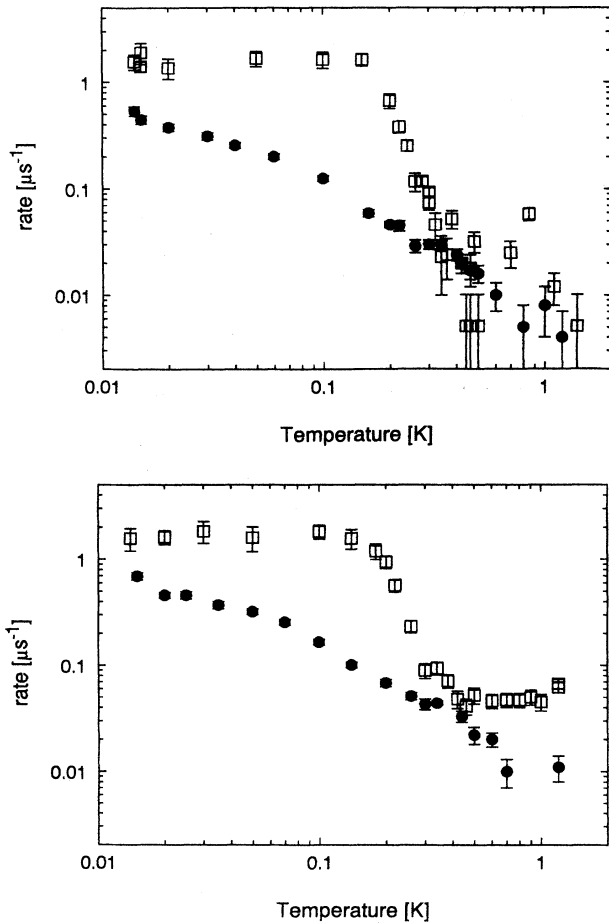


FIG. 4. The trapping rate as a function of temperature for Al-doped with Li concentrations of 10 (upper) and 20 (lower) ppm. For the superconducting state the rate displayed corresponds to the trapping fraction. (Same notation as in Fig. 1). The absolute values of the trapping rates depend sensitively on the choice of relative fractions.

IV. QUANTUM DIFFUSION IN A METAL AND THE EFFECTS OF A DISTRIBUTION OF ENERGY LEVELS

As already noted above, the temperature dependence of Λ in Fig. 1 shows a large discrepancy when compared to the Kondo-Yamada theory for a perfect crystal; the steep increase in Λ in Fig. 1 does not occur at T_c but at a much reduced temperature. Even more abrupt changes are observed for the low-doped samples around 0.3 K for the depolarization of the fraction a_l , when a two-component separation is done in the fit. Such sharp transitions are indeed expected in the Kagan-Prokof'ev theory.

A. The Kagan-Prokof'ev theory

Kagan and Prokof'ev¹⁵ developed a detailed theory of the tunneling diffusion in a metal taking into account also the effect of a distribution of energy levels due to crystal defects. They showed first, as mentioned in the Introduction, that the nonadiabatic interaction between the muon and the electrons is restricted to the energy interval $\hbar\omega_0 \ll \varepsilon_F$ near the Fermi surface.

Below the superconducting transition the tunneling matrix element J_{eff} should then be replaced by¹⁶

$$J_{\text{eff}} = J(\Delta_s/2\hbar\omega_0)^K. \quad (6)$$

Secondly, the quantities Γ and Γ_s in Eqs. (2) and (3) are modified due the presence of the static distribution over the energy levels which is measured by the parameter ξ . For the superconducting state when $(\xi, \Gamma_s)_{\text{max}} > \Delta_c$, Δ_c being the coherent bandwidth in the pure crystal, Γ_s has to be replaced by $(\Gamma_s^2 + \xi^2)/\Gamma_s$.

The local diffusion coefficient is then expected to have the temperature dependence

$$D(T) \approx \frac{za^2}{3} \frac{J_{\text{eff}}^2 \Gamma_s(T)}{\xi^2 + \Gamma_s(T)^2} \quad (7)$$

for $\xi < k_B T$.

As the temperature decreases below T_c one easily meets the condition $\Gamma_s \ll \xi$, in regions close to impurities, which leads to the limit

$$D(T) \approx \frac{za^2}{3} \frac{J_{\text{eff}}^2 \Gamma_s(T)}{\xi^2}. \quad (8)$$

In these regions, the local diffusion coefficient decreases exponentially with increasing T [see Eq. (3)]. It should be pointed out that the temperature dependence of D given by Eq. (7) is quite different from the one in Eqs. (2) and (3), in particular in the superconducting state where Γ_s becomes small. Further reduction of the temperature leads to the fulfilment of the condition $\Gamma_s \ll \xi$ over large regions around the impurities and a strong restriction of the muon mobility in most of the sample volume.

B. Analysis of the experimental data in terms of the Kagan-Prokof'ev theory

The existence of an energy-level distribution in a doped sample is thus of crucial importance for the muon depo-

larization in the superconducting state. Two important consequences are

(1) One would expect to observe two fractions of muons that differ in their depolarization behavior. The first fraction corresponds to muons thermalizing inside a region of radius r_Δ around each defect in which the energy shift is larger than the coherent band width of the muon Δ_c . With $T/T_c \ll 1$ the muons in this first region are localized [Eq. (2) is valid for $r > r_\Delta$ only]. For the second fraction, corresponding to muons thermalizing in the rest of the crystal, one expects a band motion to develop since, with the superconducting gap present, there would be no inelastic muon-electron collisions that could lead to loss of coherence. Once a band motion has developed, the transition probability into the region of localization must involve inelastic processes due to thermal electron excitations and this probability is, for these temperatures, small. Thus, the muons initially stopped in the first region are expected to be static and will depolarize, while the polarization of the other muons is essentially preserved when $T \rightarrow 0$ since a bandlike motion does not lead to depolarization. As has been noted already, the $G_{zz}(t)$ data in Fig. 3 for the low-doped samples are compatible with a "static muon" fraction superimposed on a constant background.

(2) At the trapping radius r_t , where the dopant induced interaction energy ξ is of order $k_B T$ and $\xi < \Gamma_s$, we expect, according to Eqs. (2) and (3), an increase in the diffusion rate as the temperature is lowered from T_c . This leads to increased trapping and, consequently, an increase in the depolarization rate according to Eq. (5). At slightly lower temperatures the diffusion coefficient at the trapping radius takes the form of Eq. (8) and the trapping rate decreases (exponentially) with decreasing T . There is an indication of a weak maximum in the depolarization rate at $T_{\max} = 0.8 T_c$ in the low-doped material which can be explained by this interplay between inhomogeneous and thermal broadening of the energy levels for the initial and final states.

(3) At still lower temperatures, the radius around the defect within which muons show static depolarization increases rapidly when lowering the temperature. This is due to the exponential dependence of $\Gamma_s(T)$ (which should be compared to the actual ξ at this radius). A localization radius r_{loc} can be defined as the radius at which the jump rate τ_c^{-1} is equal to the full depolarization rate σ (i.e., $\sigma \tau_c = 1$). Muons stopping inside this radius are practically immobile. With realistic assumptions of the r dependence of ξ and the band width Δ_c it is possible to calculate r_{loc} and therefore the expected temperature for complete localization in the defect region. As will be shown below an almost complete localization (and static behavior of the muons inside r_Δ) is expected below 0.2 K for all Li concentrations. Between this temperature range where "direct localization" occurs and T_{\max} there will be a minimum in the depolarization rate $\Lambda(T)$ as the localization radius r_{loc} coincides with the trapping radius r_t .

An estimate of the extent to which inhomogeneities due to imperfections will affect the muons can be made

by considering the mean distance l_{ii} between impurities at a specific impurity concentration c (in ppm). With lattice constant a one obtains

$$l_{ii} \approx 40ac^{-1/3}. \quad (9)$$

In a previous paper by some of the present authors⁵ the diffusion of positive muons in normal-conducting Al, doped with several different impurities, was analyzed in great detail. Trapping radii and sites, the effective small-polaron matrix elements J_{eff} for the tunneling of the muons in the unperturbed Al regions, and elastic strain tensors around the impurities were also calculated. The trapping radius for Li, a weak trap, was derived to be $1.1(1)a$ (a is the lattice parameter) for the low- T trapping process. The trapping radii data were normalized to that of an octahedral site at a Ag trap (radius $0.5a$), since Ag showed the weakest trapping of the dopants investigated. A typical energy variation due to elastic strain over the first few lattice distances around a Li atom is 0.5 meV (see Fig. 8 of Ref. 5).

Differences in potential energies for neighboring muon sites are also caused by the oscillations in electron density around each impurity. This is, actually, a larger effect proportional to the Friedel-Kohn oscillation amplitude V_{KF} . At the muon-impurity distance $l_{ii}/2$, where ξ should have a minimum, the difference between the single impurity Friedel-Kohn potential at adjacent muon positions can be determined if the corresponding amplitude V_{KF} is known. Electron structure calculations by Mahajan and Prakash¹⁹ arrive at $V_{\text{KF}} \approx 30$ meV which gives, using the r^{-3} dependence of the distortion potential,

$$|\Delta E(l_{ii}/2)| \approx 1.3c \mu\text{eV}. \quad (10)$$

An estimate of $\Delta_c \approx 4 \mu\text{eV}$ for the muon coherent bandwidth in superconducting aluminum can be obtained from the results of Ref. 5. Comparing the asymmetry according to Eq. (10) with the bandwidth $z\Delta_c \approx 50 \mu\text{eV}$ one finds that the asymmetry term exceeds the bandwidth at $c \approx 40$ ppm which leads to Anderson localization, in reasonable agreement with the experimental observation. Also at lower Li concentrations there is a large region around a defect where $\xi > \Gamma(T_c)$ and, in particular, where $\xi > \Gamma_s(T)$ in the superconducting state so that the muon is effectively trapped.

This is also the concentration range where the undamped fraction in the experimental data starts to disappear. The actual depolarization functions $G_{zz}(t)$ expected from the Kagan-Prokofev theory can be obtained through Monte Carlo type simulations.²⁰ These simulations, although too time consuming to be used in fitting procedures, seem to be able to reproduce the experimental depolarization curves using only two parameters [Δ_c and $\xi(r=a)$] which in the present case take on the values $\Delta_c \approx 4 \mu\text{eV}$, $\xi(a) \approx 40$ meV. The basic features of all the depolarization curves $G_{zz}(t, T, c)$ are thus reproduced using only two parameters in the Kagan-Prokofev theory.

C. Alternative interpretations of the data

A feature that distinguishes the above interpretation of the depolarization data from earlier ones is that below 0.3 K a large fraction of the muons are immobile from the moment they are stopped after implantation, i.e., the trapping model as expressed by Eq. (4) is not applicable to this fraction. Above 0.3 K some of the muons can be released and go through a normal trapping process.

It should be remarked that all experimental data presented can actually also be fitted by a trapping model based on Eq. (5) if the fraction of diffusing muons $P(t)$ is not taken from Eq. (4), but instead takes on the stretched exponential form $P(t) = \exp[-(\nu_0 t)^\beta]$. Such functions have earlier been applied to random systems, like H diffusion in amorphous silicon.²¹ The physical background is an assumption about a distribution of energy levels between which the particles (or spins) can relax. The fitting parameter β (one usually finds values $0 < \beta < 1$, where $\beta = 1$ corresponds to the simple trapping model) will be temperature dependent and further analysis of $\beta(T)$ can give information on the width of the distribution of the site energies: the theory predicts, for instance, for random systems that β should increase linearly with temperature $\beta = T/T_0$ where $k_B T_0$ is a measure of the width of the distribution in question.

Fits to the polarization decays for the 20 ppm as well as those for 75 ppm Li sample are shown in Fig. 5, leaving both parameters β and ν_0 free. For 75 ppm doping, where the largest energy spread is anticipated, the extracted β actually turns out to be a linear function of temperature for $T > 0.3$ K corresponding to an energy spread $k_B T_0 = 0.1$ meV. The same procedure for the 20-ppm-doped sample results in an average spread of 0.04 meV, both functions having a saturation value of $\beta = 0.4$ below

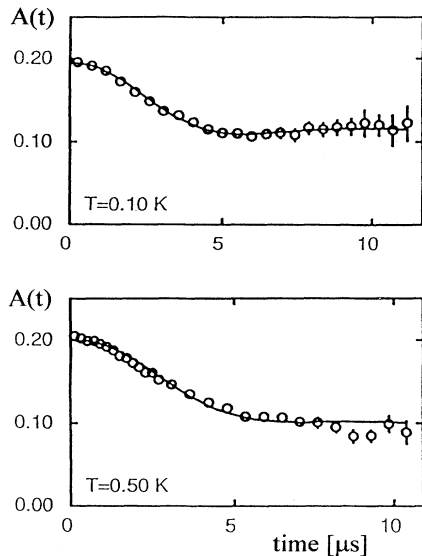


FIG. 5. Time spectrum of the 20 ppm sample at 0.10 K and the 75 ppm sample at 0.50 K fitted with a single fraction using the modified trapping model. The parameter β is 0.3, respectively, 0.5.

about 0.3 K (see Fig. 6 in Ref. 18). In this model, the trapping rate ν has a distribution $f(\nu)$, e.g., for $\beta = \frac{1}{2}$ given by $f(\nu) = \frac{1}{2} \nu_0 (\nu/\pi \nu_0)^{1/2} \exp(-\nu_0/4\nu)$, having greater spectral density at smaller ν for smaller β .²² Thus, the saturation would be due to the limiting contribution from a nontrapping muon ($\nu \sim 0$) which is in a tunneling state (i.e., corresponding to the region $\xi < \Delta_c$). For temperatures above 0.4 K for the 10 and 20 ppm samples, a simultaneous fit of ν_0 and β in the stretched exponential model was not meaningful since the two parameters are closely correlated.

It should be noted that particle diffusion and trapping in the oscillating Kohn-Friedel potential created by randomly distributed impurities (especially at large distances from defects) indeed resembles the situation with a wide distribution of trapping energies and trapping rates. This might explain why the formal description of the $G_{zz}(t)$ shapes based on the temperature-dependent parameters ν_0 and β above, and the $G_{zz}(t)$'s obtained in the Monte-Carlo simulations based on the Kagan-Prokofev model, both are in reasonable quantitative agreement with the data.

V. DISCUSSION

The experimental results reported here have revealed several new aspects of the effect of superconductivity on a diffusing particle. Furthermore, it has been clearly recognized that it is important to distinguish between quantum tunneling in a perfect crystal (as discussed in the Kondo-Yamada theory) and in a crystal with static disorder limiting the tunneling. A delicate interplay between static energy distribution effects and temperature-dependent effects in the particle-bath interaction has been demonstrated. For a fully quantitative comparison it is necessary to model a three-dimensional distribution with randomly placed impurities. The probability for the occurrence of the condition $\sigma \tau_c \leq 1$ can then be determined over the whole sample as a function of temperature. This is a rather straightforward procedure but requires considerable computing time.

The variation of the muon depolarization with doping concentration has also shed new light on the conditions for delocalization of a particle in disordered structures in general. For those experiments that were made in the superconducting state there is a strong indication that there is a fraction of muons a_{free} developing true delocalization, i.e., a coherent, extended wave function.

For the normal-conducting state it was pointed out by Kagan and Prokofev¹⁵ and by Kondo,⁶ and later discussed in detail by Hedegård,²³ that the time between inelastic collisions with conduction electrons (destroying coherence) τ_{coh} , is always shorter than the "residence time" $\approx \hbar/J_{\text{eff}}$ between two consecutive tunneling events, except possibly at very low temperatures. The process is then always of hopping type. For the superconductor it is not necessarily so since the coherence time gets longer when the probability for inelastic scattering against electrons is decreased due to the gap. In the low-temperature region studied here, where the phonon creation probability is negligible over the time range considered, most

scattering processes for the muon are likely to be elastic and thus preserving coherency. Therefore, extended muon states, limited only by the disorder in the local environment, could develop. Such states can exist in regions far from impurity centers, which can be considered as an analog of the "weak localization" of electrons in certain disordered materials with low conductivity.²⁴

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