Diffusion and localization of muonium in Na-doped KCl

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Quantum diffusion of muonium has been studied in crystals of KCl doped with small amounts of Na using the technique of muon spin relaxation. At low temperatures the crystal volume may be divided into two parts, one where the muonium diffuses rapidly as in pure KCl and the other where the muonium is effectively localized. These results are in accordance with recent theories on quantum diffusion in inhomogeneous media which predict a volume around an impurity inside which localization occurs and outside which the particle diffuses freely.

Quantum diffusion is an interesting phenomenon in crystalline solids where light interstitial particles like protons and muons migrate by tunneling under the potential barrier between adjacent sites. The tunneling matrix is determined by the overlap integral between the initial and final polaronic states and therefore quite sensitive to the energy level shift due to the presence of other crystalline defects (impurities, dislocations, etc.). Such disorder may control the diffusion of muonium (Mu, the muonic analog of hydrogen in which the proton is substituted by a positive muon) in nonmetallic crystals and muon diffusion in superconducting metals[1] because the predominant two-phonon damping factor for the tunneling decreases steeply with decreasing temperature and accordingly the level shift plays a limiting role at low temperatures.[2] However, there always remains some uncertainty about the effect of unintentional impurities/defects in nominally pure specimens because of the lack of quantitative information on the concentration and type of defect/impurity. A study of quantum diffusion in well-characterized samples with known impurities is thus desirable.

In this paper we report on the diffusion of muonium centers in KCl doped with Na substitutional impurities. KCl is one of the materials in which muonium diffusion is well understood in terms of the current theory for quantum diffusion in homogeneous media.[2] Our results indicate that the crystal volume is split into two parts upon Na doping, one in which the diffusion slows down with decreasing temperature and the other in which the character of rapid quantum diffusion is preserved. This is in qualitative agreement with recent theoretical predictions[3] and similar experiments in solid N_2.[4] In addition, we report a decrease of the missing muon spin polarization with increasing Na concentration and propose an explanation in terms of diffusion-controlled reaction of muonium centers with excitons produced by muon radiolysis.

Quantum diffusion in the absence of disorder results in a power-law temperature dependence of the particle hop rate ν over a certain temperature range, i.e.,

$$\nu \simeq \frac{2\lambda J^2}{\Omega(T)} \propto T^{-\alpha+2\beta}, \qquad (1)$$

where $J \propto T^\beta$ is a renormalized tunneling matrix or the muonium bandwidth ($\hbar = 1$), λ is a factor determined by the crystal structure, $\Omega(T) \propto T^\alpha$ is the damping rate due to coupling to the environment, and α and β are constants predominantly dependent on the nature of dissipation. Equation (1) is valid in the incoherent hopping regime: $T \ll \Theta_D$ (the Debye temperature) and $\Omega(T) \gg J$. Such a power law has been clearly observed both in metals with $1 - 2\beta \simeq 0.6$ ($\alpha = 1$),[5] and in ionic crystals with $\alpha \simeq 3-7$ ($\beta = 0$),[4–9] where the smaller power in the former case is due to the electronic damping of the muon tunneling matrix[10,11] while particle-phonon interaction is explicit in the latter.[12] If there is an energy difference $\xi \equiv \epsilon_i - \epsilon_f$ between the initial (ϵ_i) and final (ϵ_f) Mu states due to static disorder in the potential surface, then Eq. (1) must be modified:[13]

$$\nu \simeq \frac{2\lambda J^2 \Omega(T)}{\xi^2 + \Omega^2(T)} \frac{\xi/T}{1 - e^{-\xi/T}}. \qquad (2)$$

In insulators where $\beta = 0$, $\Omega(T)$ shows a much steeper temperature dependence than the Gibbs factor [i.e., $\xi/T(1 - e^{-\xi/T})$]. Consequently, Eq. (2) implies that the hopping frequency as a function of decreasing T turns over at the temperature where $\Omega(T) \simeq |\xi|$ and at lower T decreases as

$$\nu \simeq \frac{2\lambda J^2 \Omega(T)}{\xi^2} \propto T^\alpha (\Omega \ll |\xi|). \qquad (3)$$

Such behavior has recently been reported in solid nitrogen where $\alpha \simeq 7$.[4] Note that when the energy difference ($|\xi|$) is small compared with the tunneling matrix element the tunneling probability levels off at the value $2\lambda J$ provided $J \gg \Omega(T)$, i.e., the band motion limit.[14]

We have recently shown that the longitudinal spin relaxation of the muon polarization is a sensitive measure of the Mu diffusion rate.[6] The connection between the diffusion rate and the muon spin relaxation rate originates with the nuclear hyperfine (nhf) interaction which produces an effective magnetic field on the unpaired muonium electron which depends on the orientation of the nuclear spins. As the muo-

nium diffuses the direction of this effective field fluctuates, thereby inducing transitions between the muonium hyperfine levels. From the resulting muon relaxation rate and its T dependence (after appropriate modeling of the spin dynamics using the Redfield equations of motion[15,16]) one can extract an average electron-nuclear coupling constant δ_{ex} and the correlation time τ_c (corresponding to ν^{-1}) for the field fluctuations. The general expression for the longitudinal muon polarization $P_z(t)$ is complicated, depending upon δ_{ex}, τ_c, and the external longitudinal field B; however, for the purpose of illustrating the essential features of our data it is sufficient to consider the idealized situation where τ_c^{-1} is small with respect to all transition frequencies except the smallest intratriplet transition ω_{12}. Ignoring all higher-frequency transitions leads to single-exponential relaxation of the muon spin polarization:

$$P_z(t) \approx P_z^0(x) \exp[-\Lambda_z(x)t], \qquad (4)$$

where

$$P_z^0(x) = 1 - x/\sqrt{1+x^2},$$

$$\Lambda_z(x) = \frac{\delta_{ex}^2 \tau_c}{1 + \omega_{12}^2 \tau_c^2},$$

$$\omega_{12} = \tfrac{1}{2}\omega_0\left(1 + \frac{\Gamma_-}{\Gamma_+}x - \sqrt{1+x^2}\right),$$

where ω_0 is the muonium hyperfine parameter, $\Gamma_{\pm} = \tfrac{1}{2}(\gamma_e \pm \gamma_\mu)$ with γ_e and γ_μ being the respective electron and muon gyromagnetic ratios, and $x = 2\Gamma_+ B/\omega_0$. Since both $P_z^0(x)$ and $\Lambda_z(x)$ depend on the applied field B (or x), the other physical parameters like δ_{ex} and τ_c at a temperature are uniquely determined by comparing a set of time spectra obtained under different fields with the model function $P_z(t) \equiv P_z(t; \delta_{ex}, \tau_c, x)$. The parameter δ_{ex} can be related to the nearest-neighbor halide nhf parameters $\omega_\parallel$ and $\omega_\perp$ with the result

$$\delta_{ex}^2 = (\omega_\parallel^2 + 2\omega_\perp^2)\frac{nI(I+1)}{3}, \qquad (5)$$

where the second term is the mean squared value of the component of total nuclear spin along the applied field for n nearest-neighbor nuclei with spin I.[7]

The experiment was performed on the M13 and M15 beamlines at TRIUMF, which provide beams of 100% spin-polarized positive muons of momentum 28 MeV/c. Single-crystal specimens of KCl with Na concentrations of $2.7(3) \times 10^{-3}$ and $2.7(3) \times 10^{-2}$ were obtained from the University of Utah. At each temperature conventional muon spin relaxation (μSR) spectra were taken with an external field applied along the initial muon polarization direction and a [100] crystalline direction. This longitudinal field (LF)-μSR technique, details of which can be found elsewhere,[16] measures directly the time evolution of the μ-e decay asymmetry $A(t) = A_0 f P_z(t)$ in the time scale of the average muon lifetime (2.2 μs), where A_0 is the maximum experimental decay asymmetry and f is the fractional yield of muonium. Some typical examples of the time spectra are shown in Fig. 1.
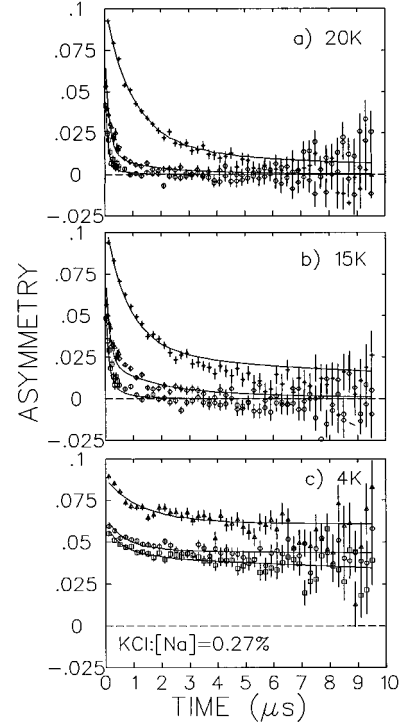

FIG. 1. LF-μSR time spectra in KCl ([Na]=0.27%) at 0.02, 0.05, and 0.15 T. Solid curves are fitted results by a model for muonium diffusion with two components (see text).

Note that the field-dependent relaxation predicted by Eq. (4) is clearly observed.

The spectra also imply that there is little or no contribution from the diffusion-limited trapping (DLT) process associated with the doped impurities. The trapping rate under a simple DLT model is given by

$$\nu_t = 4\pi r_t c_t D_\mu \qquad (6)$$

$$= \frac{2\pi}{3}\rho\nu, \qquad (7)$$

where r_t is the trapping radius (in units of the lattice constant a), c_t is the concentration of trapping centers (ρ is the equivalent atomic concentration), and D_μ ($= a^2\nu/6$ for the interstitial Mu in KCl) is the diffusion coefficient of muonium.[17] The spin depolarization in this model is given by $\exp[-(\nu_t + \Lambda_z)t]$. [Note that muons are depolarized during diffusion whereas no depolarization occurs in the stationary (trapped) state; this is opposite to the case discussed for the muon trapping in metals.[18]] It has been inferred from the previous measurements in nondoped Cl that $\nu \gtrsim 10^8$ s^{-1},[6,7] and accordingly the trapping rate ν_t to the sodium impurity estimated from Eq. (6) is greater than about 10^6 s^{-1} for both 0.27% and 2.7% samples. On the other hand, the observed spin relaxation rate in the time spectra indicates $(\nu_t + \Lambda_z) \lesssim 10^6$ s^{-1} over the entire temperature range. Since ν_t is independent of magnetic field, the strong field dependence of the observed relaxation rate further evidences that $\nu_t \ll \Lambda_z(x) \sim 10^6$ s^{-1}. Thus, we are lead to conclude that the DLT model is inappropriate for the interpretation of the current data.

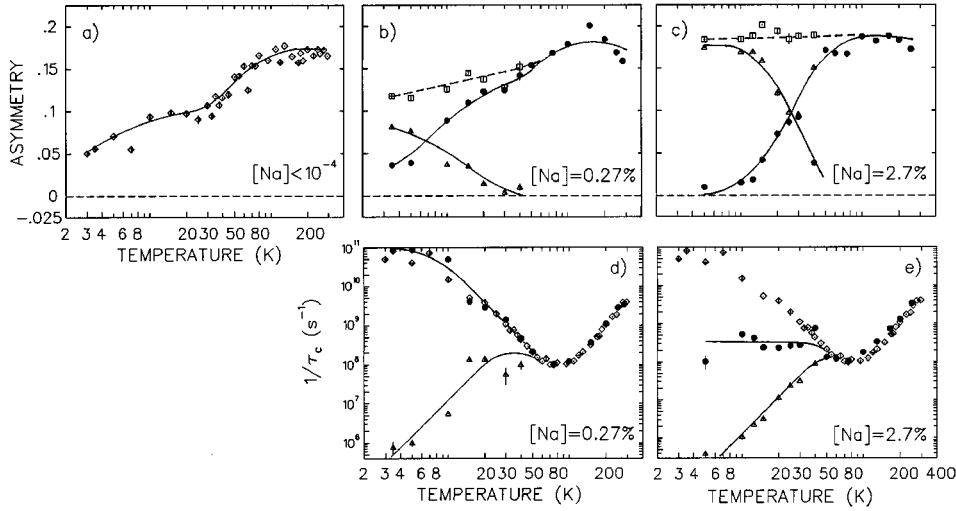FIG. 2. Muonium hopping rate ($\equiv$ inverse correlation time) and partial asymmetry in KCl ([Na] = 2.7×10^{-3} and 2.7×10^{-2}) compared with data in pure KCl ([Na] $< 10^{-4}$, shown by diamonds). Triangles are A_s and τ_s^{-1}, filled circles are A_f and τ_f^{-1}, and squares are $A_s + A_f$ in each sample. Lines are merely intended to guide eyes.

The analysis of time spectra below 50 K reveals that there exist at least two distinct components with different correlation times; however, only a single component is necessary to reproduce spectra at higher temperatures. For simplicity we assume just two components such that

$$A(t) = A_0[f_s P_z(t; \tau_s) + f_f P_z(t; \tau_f)] = A_s P_{sz}(t) + A_f P_{fz}(t),$$

where τ_s and τ_f denote the correlation times corresponding to slow and fast diffusion, A_i ($i = s, f$) being their (partial) asymmetry proportional to the fractional yields f_i with $A_0 \simeq 0.2$. Common values were adopted for the muonium hyperfine parameter ω_0 [$= 2\pi \times 4280$ MHz (Ref. 19)] and the nhf parameters δ_{ex} [$= 2\pi \times 60(2)$ MHz (Ref. 7)] throughout the following analysis: the nhf parameter is predominantly determined by the nearest-neighbor (NN) halogen nuclei and therefore the substitution of potassium by sodium has negligible effect on the value of δ_{ex}. The solid curves in Fig. 1 are the fitting results with this model. The temperature-dependent inverse correlation time and corresponding asymmetry are shown in Fig. 2 where the data[6,7] of pure KCl (nominally 99.99%, obtained from Harshaw Chemical Co.) are also included for comparison. It is evident from Figs. 2(d) and 2(e) that the hop rate in both samples is not affected by the presence of sodium impurities above 50 K, except for a slight enhancement in the sample with 2.7% sodium. This implies that the disorder in the muonium potential energy induced by the sodium impurity is far smaller than the thermal activation energy ($\sim 10^3$ K) required for phonon-assisted tunneling in the high-T region.

In the sample with 0.27% sodium the fast-diffusing component observed below 50 K is virtually unaffected by the impurity: The temperature dependence of the hopping rate [τ_f^{-1} in Fig. 2(d)] and the corresponding asymmetry [A_f in Fig. 2(b)] are very close to those observed in pure KCl [cf. Fig. 2(a)]. Moreover, τ_f levels off below 5 K and stays constant down to 15 mK. This is also the case for pure KCl,[20] and, therefore, leads us to conclude that impurities at a concentration of less than 10^{-3} have negligible effect on the sizable fraction of muonium undergoing quantum diffusion. A second slowly diffusing component emerges below 50 K and increases with decreasing temperature as inferred by A_s in Fig. 2(b). The hopping rate (τ_s^{-1}) associated with

this component seems to level off at 50 K and exhibits a relatively steep decrease below 15 K [see Fig. 2(d)], indicating that a certain fraction of the Mu becomes immobile at low temperature.

The effect of 2.7% sodium is more substantial. As evident in Fig. 2(e) no fraction of the muonium shows the characteristic increase of hopping rate with decreasing temperature below 50 K seen in undoped KCl. The hopping rate of the fast-diffusing component tends to level off below 30–40 K, although there is some uncertainty due to its diminishing fractional yield. Also the fractional yield (or A_f) shows a steeper decrease with decreasing temperature compared with that in the 0.27% sample [see Fig. 2(c)]. The slowly diffusing component emerges in place of the fast component with decreasing temperature. The hopping rate clearly shows a power law given as $\tau_s^{-1} \propto T^\alpha$ with $\alpha = 3.2$. This behavior of τ_s^{-1} and leveling off of τ_f^{-1} indicates that the increase of the fractional yield for the slowly diffusing component is due to expansion of the crystal volume V_s where muonium motion is being reduced, rather than trapping of fast-diffusing muonium near impurities. This may not be true for the 0.27% sample, since the increase of A_s is in parallel with the increase of the hopping rate τ_f^{-1}. It is therefore possible that the Mu is being trapped at a rate determined by its diffusion rate [the Gibbs factor in Eq. (2) also ensures such an irreversible process]. However, the fractional yield of the slowly diffusing component below 50 K is qualitatively proportional to the sodium concentration [see Figs. 2(a)–2(c)] and thus it can be regarded as the fractional volume itself [see Eq. (14)]. The leveling off of τ_f^{-1} below 30 K may be due to the residual disorder outside V_s or the effect of single-phonon scattering, although there remains some uncertainty due to smaller fractional yields at lower temperatures. The immobile Mu in the 2.7% sample at the lowest temperature is further demonstrated by the magnetic-field dependence of the muon polarization under longitudinal field. Figure 3 shows the quenching of the nhf field below 0.01 T and is in good agreement with the theoretical prediction for stationary Mu.[21] Because of rapid spin relaxation due to the fast muonium diffusion such field quenching of the nhf field is not observable in pure KCl.[20]
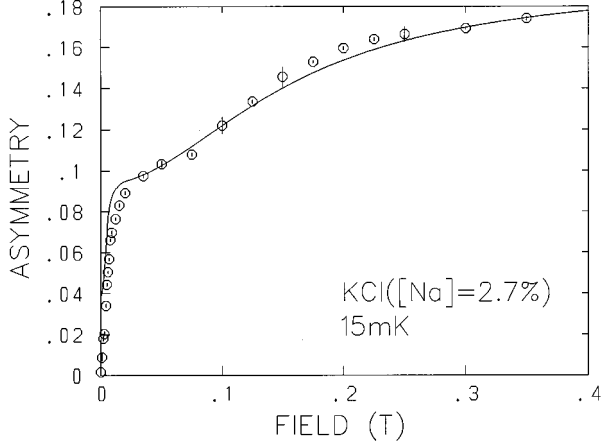
FIG. 3. Muon spin polarization at $t=0$. $P_z(t=0)$ versus longitudinal magnetic field at 15 mK in KCl with 2.7% sodium impurity. Solid line is a theoretical prediction by Beck *et al.* (Ref. 21).

The observed $T^{3.2}$ dependence of the hopping rate below 50 K in the 2.7% sodium sample (which is suggested also in the 0.27% sample below ~ 20 K), together with the previous result in pure KCl [$T \propto T^{-3.3}$] (Ref. 6), is in line with the theoretical prediction expressed in Eqs. (1)–(3). Assuming the same $\Omega(T) \propto (T/\Theta_D)^\alpha$ and J in both samples, we have

$$\tau_c^{-1} \simeq \frac{2\lambda J^2}{\gamma}\left(\frac{T}{\Theta_D}\right)^{-\alpha} = \tau_0^{-1}\left(\frac{T}{\Theta_D}\right)^{-\alpha} \qquad (8)$$

for the muonium in pure KCl and

$$\tau_c^{-1} \simeq \gamma\frac{2\lambda J^2}{\xi^2}\left(\frac{T}{\Theta_D}\right)^{\alpha} = \tau_1^{-1}\left(\frac{T}{\Theta_D}\right)^{\alpha} \qquad (9)$$

for the slowly diffusing component in the sodium-doped sample, where the values of τ_0^{-1} and τ_1^{-1} are obtained experimentally from the data below 50 K in Fig. 2(e) as $1.52(2) \times 10^6$ and $2.1(1) \times 10^{10}$ s^{-1}, respectively. Taking $J=0.13$ K (with $\lambda=4$ for simple cubic symmetry) deduced from pure KCl,[14] we obtain

$$|\xi| \sim 16 \pm 1 \quad \text{K} \qquad (10)$$

for the KCl samples with 2.7% sodium. It must be stressed, however, that the value of ξ varies from site to site in most cases where the energy difference (or disorder) is due to the strain fields from impurities spatially distributed at random, and thereby the above value should be regarded as a mean value (see below).

In order to make a more intuitive understanding of the effect of energy disorder we resort to a specific model in which the disorder is induced by a strain field from the sodium impurity:

$$U(r) = U_0\left(\frac{a}{r}\right)^3, \qquad (11)$$

where r is the distance from the impurity atom and a is the lattice constant. Then the energy difference between neighboring sites is given as
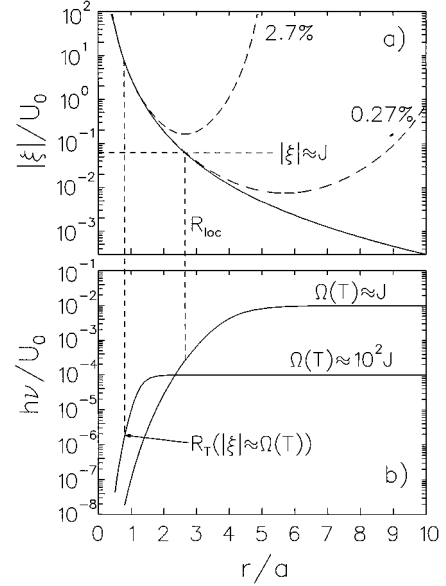

FIG. 4. (a) The solid curve is the energy difference between adjacent muonium sites [Eq. (12), in units of U_0] as a function of distance r (measured by the lattice constant a) from a single impurity atom at the origin. The dashed curves are those when the impurities are placed regularly with a mean distance $2R_0$. When $J/U_0 < 10^{-1}$, $|\xi|$ in the 2.7% sample may be larger than J at any r (i.e., $R_0 < R_{\text{loc}}$), so the fraction of slowly diffusing Mu may approach 100% at low temperatures [which is actually observed in Fig. 2(c)]. (b) Hopping rate ν ($U_0 = h = 1$) vs r at two temperatures such that $\Omega(T_1) \simeq 10^2 J$ and $\Omega(T_2) \simeq J$, respectively ($J/U_0 \sim 10^2$ is assumed). Note that R_T increases with decreasing temperature to reach R_{loc} [i.e., $\Omega(T) \propto T^{3.2}$].

$$\xi(r) \simeq \frac{a\partial U(r)}{\partial r} = -3U_0\left(\frac{a}{r}\right)^4. \qquad (12)$$

A boundary radius R_J may be defined by the equation $U(R_J) \simeq J$, over which muonium is not allowed to penetrate elastically.[3] However, the tunneling matrix, and accordingly R_J, are not temperature dependent in insulators ($\beta=0$) and this does not explain the observed temperature-dependent fractional yields of slow- and fast-diffusing components. The situation is the same for the radium R_{loc} defined by the strict localization condition $|\xi(R_{\text{loc}})| \simeq J$.[3]

Note that the temperature dependence of the hopping rate is almost opposite between the fast- and slow-diffusing components as described either by Eq. (1) ($|\xi| \ll \Omega$) or by Eq. (3) ($|\xi| \gg \Omega$), respectively. Clearly, the crossover between these two extremes is determined by a condition $\xi(R_T) \simeq \Omega(T)$. This is further justified by the steep r dependence of ξ^2 ($\propto r^{-8}$) in the denominator of Eq. (2). Thus we have

$$R_T = \left(\frac{3U_0}{\Omega(T)}\right)^{1/4} a \propto T^{-\alpha/4} \qquad (13)$$

and a fractional yield

$$x(|\xi| > \Omega) \simeq \frac{4\pi}{3}c\left(\frac{R_T}{a}\right)^3 \propto T^{-3\alpha/4} \qquad (14)$$

for the slowly diffusing component, where c is the number density of the impurity atom. As illustrated in Fig. 4, R_T

increases with decreasing $\Omega(T)$ until it reaches the maximum value R_{loc}. Muonium diffusion in the rest of the crystal volume (where $|\xi|<\Omega$) is faster than in the above region and its fractional yield (except the missing fraction) is given as $1-x$. Thus the temperature-dependent yields in Figs. 2(b) and 2(c) are qualitatively explained by the above model with $f_s/(f_s+f_f)\sim x$ and $f_f/(f_s+f_f)\sim 1-x$. Moreover, if $R_0\equiv ac^{-1/3}/2$ (half the mean separation) is larger than R_{loc} there always remains a fraction $1-(R_{\text{loc}}/R_0)^3$ where the muonium hopping rate is virtually unaffected by impurities (although the diffusion constant might be affected by the mean free path between elastic collisions).

The model also provides a mechanism to hinder the trapping of muonium by impurities at temperatures as low as $\Omega(T)<\xi$. The probability of hopping to sites closer to the impurity (e.g., from $r=na$ to $[n-1]a$) is far smaller than that in the opposite direction (e.g., to $r=[n+1]a$; from Eqs. (3) and (11) we have

$$\frac{\nu(na\rightarrow[n-1]a)}{\nu(na\rightarrow[n+1]a)}=\frac{|\xi(na\rightarrow[n+1]a)|^2}{|\xi(na\rightarrow[n-1]a)|^2}\simeq\left(\frac{n-1}{n+1}\right)^6$$

$$(n>1). \quad (15)$$

Moreover, ξ may be much smaller for the tangential direction and thereby a much larger hopping probability may be expected. Thus the tunneling process in disordered systems has a tendency to be contained in the region of small ξ.[22]

The mean value of ξ experienced by muonium in the region of slow diffusion may be defined as

$$\bar{\xi}=\frac{1}{\sqrt{\langle\xi^{-2}\rangle}}=\left[\int_{a/\sqrt{3}}^{R_T}\xi(r)^{-2}r^2dr\ \bigg/\int_{a/\sqrt{3}}^{R_T}r^2dr\right]^{1/2}$$

$$\sim\frac{\sqrt{33}}{9}U_0\left(\frac{a}{R_T}\right)^4\propto T^\alpha, \quad (16)$$

which is also temperature dependent. However, slowly diffusing Mu is predominant in the case of higher impurity concentration, and therefore, R_T may be replaced by the mean halfway distance between impurities (see the curve of $|\xi|$ for $c=2.7\%$ in Fig. 4) to obtain

$$\bar{\xi}\sim\frac{\sqrt{33}}{9}U_0\left(\frac{a}{R_0}\right)^4=\frac{16\sqrt{33}}{9}U_0c^{4/3}. \quad (17)$$

This seems to be the case for the 2.7% sample where the experimentally obtained value $|\xi|$ may be regarded as U_0 in

order of magnitude. Thus this model provides a qualitatively consistent understanding of all the features observed in the current data in Fig. 2.

Finally, we discuss the observed recovery of the missing fraction due to the sodium impurity. As in the case of pure KCl, there is a missing fraction in the asymmetry [see Fig. 2(a)], i.e., the observed asymmetry is smaller than the maximum experimental value A_0 (i.e., $f<1$). Note from Figs. 2(a)–2(c) that the net missing fraction ($=1-f_s-f_f$) decreases with sodium concentration such that it virtually disappears in the 2.7% sample. An important feature of the missing muon polarization in alkali halides is that it strongly depends on the temperature [e.g., see Fig. 2(a)]. The above fact indicates that the fast depolarization process occurs *after* the thermalization of implanted muons. Moreover, the missing fraction is commonly found at lower temperatures where paramagnetic self-trapped excitons (STE's) induced by muon radiolysis are present. This correlation has been evidenced by the luminescence from muon-induced triplet STE's, upon which a model is proposed to attribute the missing fraction to the fast depolarization induced by some unknown dynamical process within the Mu-STE complex.[23–25]

It is evident from Figs. 2(a)–2(c) that the recovery of the missing fraction is correlated with the increase of the slowly diffusing component. This, together with the above-mentioned model, suggests that the depolarization is caused by the diffusion-controlled Mu-STE "reaction," which is suppressed by the decrease of Mu mobility due to the sodium impurity.

In summary, we have shown that a KCl crystal doped with Na appears to split into two distinct regions with regard to muonium diffusion. A specific model in which the boundary between these two regions is determined by a condition $|\xi|\simeq\Omega(T)$ provides a satisfactory account of the current data. In particular, in the sample with 0.27% sodium the quantum diffusion in the region $|\xi|<\Omega$ is virtually unaffected by the impurity down to 15 mK. This is further confirmation that previous studies of quantum diffusion in undoped KCl are characteristic of pure KCl.

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