Quantum Diffusion of the Positive Muon in Superconducting Tantalum

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The tunneling probability for a positive muon in high-purity tantalum is significantly enhanced by the onset of superconductivity. This establishes the predominant influence of low energy infrared couplings to the conduction electrons in the quantum tunneling motion of a charged interstitial atom in metals. [S0031-9007(97)03507-2]

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The diffusion of light interstitial atoms in a crystalline solid is interesting because of the expected crossover between classical incoherent hopping motion at high temperatures to coherent quantum tunneling with long range coherence at low temperatures. Such a system is a model for studying the influence of dissipation on quantum mechanical phenomena [1,2] since the lattice excitations can be represented as a bath of harmonic oscillators which perturb the coherent tunneling. One of the most intriguing aspects of the diffusion in metals is the idea that the cloud of normal conduction electrons around a charged particle cannot follow the tunneling motion of the particle immediately, as a result of the orthogonality catastrophe originating from divergent infrared couplings to the electrons [3]. This nonadiabatic electron-drag effect leads to two competing consequences with decreasing temperature, i.e., (i) the reduction of tunneling matrix element which can be regarded as “friction” or “Ohmic damping” of the tunneling motion [1,2], and (ii) an increase in the density of final states (and an associated increase of the transition probability) due to a narrowing of the impurity band.

The results reported in this paper provide proof that the low energy couplings to the conduction electrons play a dominant role in controlling the *long-range* quantum diffusion of the muon at low temperatures, which is complementary to the case of *local tunneling* of H trapped near O atoms in $Nb(OH)_x$ [4]. The key aspect of the experiment is that the low energy couplings to the electrons are suppressed in the superconducting state due to a BCS energy gap at the Fermi surface. We find that the enhancement in the muon hopping rate in the superconducting state below T_c is close to that predicted by the theory of diffusion in a crystal after including some small effects due to residual impurities [5–7].

Recent experimental results on muon diffusion in Cu [8] and in Al [9] have demonstrated that below a temperature T^* (typically a small fraction of the Debye temperature) the muon diffusion rate *increases* with decreasing temperature according to an approximate power law ($T^{-\alpha}$), where α is about 0.6. This behavior has been understood as a consequence of the electron-drag effect which leads to the small power [$2K - 1 (= -\alpha)$ with an electron-muon coupling constant K in the range between 0 and $\frac{1}{2}$] predicted by theory [10,11]. In addition, studies of muonium (μ^+e^-) diffusion in insulators [12] show that α is considerably larger ($\gtrsim$3), also predicted by theory in the absence of conduction electrons [13,14].

However, the most direct test of the influence of infrared couplings to the conduction electrons is to compare the diffusion rate in the normal and superconducting states of a simple metal. The muon hopping rate ($\propto$ diffusion rate) in superconducting metal is predicted to follow

$$\nu(T) = 2\,\frac{J^2(T)}{\Omega(T)}\,, \tag{1}$$

$$J(T) \simeq J_0\left(\frac{\pi k_B T}{w_0}\right)^K, \tag{2}$$

$$\Omega(T) = \frac{4\pi K k_B T}{1 + \exp(\Delta_S/k_B T)}\,, \tag{3}$$

where J_0 is the muon tunneling matrix element renormalized by the phonon-polaron effect, w_0 is the energy to the first excited state of a muon in the potential well, and Δ_S is the superconducting energy gap [5]. The factor $(\pi k_B T/w_0)^K$ comes from the further renormalization of the tunneling matrix due to Ohmic damping while $\Omega(T)$ represents the level broadening due to muon-electron interaction. It is clear from Eqs. (1)–(3) that, in the normal state (where $\Delta_S = 0$), $\nu(T)$ is proportional to T^{2K-1}, while in the superconducting state it would show a steep increase with decreasing temperature just below T_c as $\nu(T) \propto \exp(\Delta_S/k_B T)$. [It is predicted that $J(T)$ remains constant at $J(T_c)$ in the superconducting state.]

The first attempts to observe such an effect were made in Al dilute alloys [15,16]. However, there were difficulties in the data due to the presence of impurities which were introduced in order to monitor the diffusion rate [17]. (In pure Al the diffusion rate is too fast to observe directly.) Their main effect is to introduce a static energy asymmetry ξ (i.e., a mismatch in the potential well depth for the muon between adjacent sites), leading to a muon hopping rate which depends on ξ, T, and Δ_S [5–7]. A further Monte-Carlo simulation has revealed that the observed effect of superconductivity seen only below $T \le 0.3T_c$, which is largely different from Eq. (1), may be attributed primarily to the *slowing down* of muon diffusion caused by large energy asymmetry [18]. Thus, while these results in Al led to the development of a theory for quantum diffusion in the presence of impurities, there is still need to test the effect of superconductivity in a system where the muon diffusion can be studied below T_c without gross effect due to impurities. Tantalum ($T_c = 4.48$ K) is a candidate for such a study since previous reports suggest that the muon hopping rate is much slower than that in Al and can be measured directly by conventional μSR techniques [19,20].

A high purity polycrystalline Ta specimen was prepared for the present experiment following the steps of successive annealing: (1) at 2700 °C in ultrahigh vacuum (30 min), (2) at 1900 °C with 2×10^{-6} hPa O_2 gas (30 min), (3) at 2750 °C under a vacuum of 3×10^{-9} hPa, and (4) the formation of a protective oxide layer during cooling down. The residual resistivity ratio after these processing steps was 6900 with Nb (<10 ppm) as the main residual impurity. The μSR measurement was performed on the M20 beam line at TRIUMF which provides a beam of nearly 100% spin polarized positive muons of momentum 28.6 MeV/c. Muons were implanted into the specimen (measuring 20 mm × 22 mm with 0.5 mm thickness) after passing through a 9 mm diameter collimator. The specimen was placed in the cold He gas-flow cryostat, and the temperature was varied between 1.6 and 200 K. Positrons from muon decay are emitted preferentially along the direction of muon polarization [21] at the time of decay so that the time differential μSR spectrum yields a direct measurement of $AG_{\rho\rho}(t)$, where A is an experimental asymmetry parameter determined by the properties of muon decay and $G_{\rho\rho}(t)$ is the spin relaxation function describing the decay of the muon polarization. The μSR spectra in zero external field (ZF), $G_{zz}(t)$, were measured by cooling the sample in a small residual magnetic field of less than ±0.01 mT. Transverse field (TF) μSR spectra $G_{xx}(t)$ in the normal state were taken under an external field $H_0 = 0.1$ T which was larger than the critical field $H_c(0)$ (= 83 mT) and perpendicular to the initial muon spin polarization. Note that any residual stray field is automatically excluded in a type-I superconductor due to the Meissner-Ochsenfeld effect.

Muon spin depolarization in nonmagnetic metals is induced by the random local fields $\vec{H}_l$ from nuclear magnetic moments (typically $\sqrt{\langle|\vec{H}_l|^2\rangle} \sim 10^{-4}$ T. If the muon is diffusing, the time dependent polarization is given by an analytic function in a transverse magnetic field [22]:

$$G_{xx}(t) = \exp\left[-\frac{2\sigma^2}{\nu^2}(e^{-\nu t} - 1 + \nu t)\right], \qquad (4)$$

where σ is the static linewidth (in a frequency representation of the spectrum) and ν is the muon hopping rate in the *normal state* of the specimen. Note that Eq. (4) is a simple Gaussian function $\exp(-\sigma^2 t^2)$ at the static limit ($\nu \to 0$), and reduces it to an exponential $\exp[-2(\sigma^2/\nu)t]$ for large ν. The linewidth in the limit of fast diffusion is reduced due to motional narrowing (an effect well known in NMR). In ZF the muon depolarization function for static muons is given by a Kubo-Toyabe function

$$g_{zz}(t) = \frac{1}{3} + \frac{2}{3}(1 - \Delta^2 t^2)\exp\left(-\frac{1}{2}\Delta^2 t^2\right), \qquad (5)$$

where Δ is the static linewidth under zero field. There is no analytic expression for finite ν but the function can be generated numerically by an integral equation [23]

$$G_{zz}(t) = g_{zz}(t)e^{-\nu t} + \nu \int_0^t g_{zz}(\tau)e^{-\nu\tau}G_{zz}(t - \tau)d\tau, \qquad (6)$$

where ν corresponds to the hopping rate in the *superconducting state* of the specimen below T_c. The linewidth is estimated to be $\gamma_\mu\sqrt{\langle|\vec{H}_l|^2\rangle} \sim 10^{-1}\ \mu s^{-1}$ ($\gamma_\mu = 2\pi \times 135.5$ MHz/T) with a relationship $\Delta/\sigma \simeq \sqrt{2}$ [23]. The actual linewidth σ in Ta is indeed close to 0.1 μs^{-1} [19], and the recovery of $G_{zz}(t)$ to $\frac{1}{3}$ for $t > 1.5/\Delta \simeq 10\ \mu$s [see Eq. (5)] is not observed in the current time range of measurement $[0 \sim 10\ \mu s]$. In this case, the initial part of Eq. (5) shows a Gaussian-like decay which is approximated by $\exp(-\Delta^2 t^2)$, showing a ν dependence similar to Eq. (4) upon muon diffusion [23].

In order to compare the temperature dependence of the μSR spectra in ZF and TF in a manner which is model independent, we use a common muon spin relaxation function $G_{\rho\rho}(t) = \exp[-(\Lambda_\rho t)^\gamma]$ with which the fit was quite satisfactory for both ZF ($\rho = z$) and TF ($\rho = x$) data. The obtained parameters (Λ_ρ and γ) are shown in Fig. 1. Since the parameter Λ_ρ may be interpreted as an effective linewidth, the reduction of Λ_ρ both above 50 K and below 20 K strongly evidences the presence of muon diffusion in these temperature regions. The associated reduction of γ in Fig. 1(b) is also in line with the change of the line shape from Gaussian to an exponential decay function as seen in Eq. (4) with increasing ν. Moreover, a steep decrease of Λ_z is observed just below T_c in accordance with the onset of superconductivity, whereas no such tendency is found in Λ_x around T_c. Thus, the reduction of Λ_z below T_c is strong evidence that the muon diffusion rate is enhanced in the superconducting state.

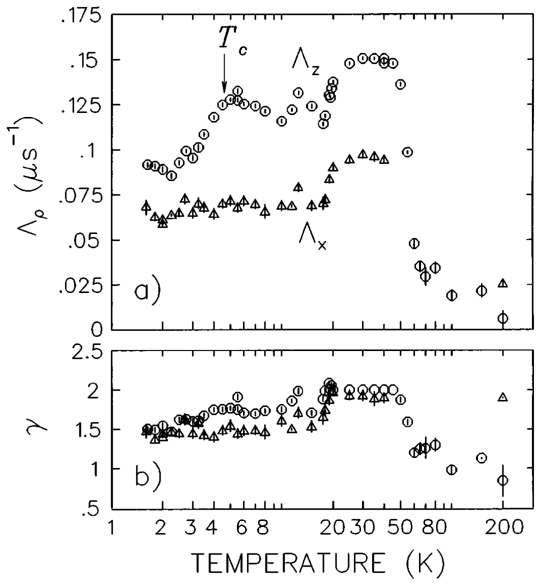

FIG. 1. (a) μSR linewidth parameter Λ_ρ and (b) the power γ obtained by fitting data with a stretched exponential function $\exp[-(\Lambda t)^\gamma]$ in the normal and superconducting states of Ta. Λ_ρ is a measure of the time averaged nuclear dipolar fields seen by the muon.

The muon hopping rate can be deduced by analyzing TF data with Eq. (4) and ZF data with the "dynamical" Kubo-Toyabe (KT) function [23], respectively. In particular, a stringent test for the presence of motional effects is provided by comparing the μSR time spectra in a low-longitudinal field with the corresponding dynamical KT function. Figure 2 shows an example at 5 K, where the spectra at 0, 0.5, and 1 mT are fitted by the dynamical KT function for such fields with common parameter values for Δ and ν. No such consistency is possible without including this motional effect. This shows that the reduction of the effective linewidth below 20 K is due to muon diffusion. The static linewidths in TF and ZF were deduced from the data ~20–50 K to be $\sigma = 0.0978(5)\ \mu s^{-1}$ and $\Delta = 0.1433(5)\ \mu s^{-1}$, respectively. The most reliable value of the hopping rate is deduced by fitting all the spectra with σ and Δ fixed to these values. The final results below 30 K are show in Fig. 3. The hopping rate deduced from TF data shows excellent agreement with that from ZF data above T_c, providing confirmation of the current model of motional narrowing. Note the clear enhancement of the hopping rate below T_c which agrees with the theoretical prediction.

However, there are two features in the data shown in Fig. 3 which are not readily understood by Eq. (1), namely, (i) the peaks in the hopping rate ν about 10 and 19 K, and (ii) leveling off of ν in the superconducting state (ZF) below ~3 K. Taking into account that the temperature dependence of ν in this temperature region is considerably different from the previous result in less pure Ta (99.997%, untreated) [20], we interpret these features in terms of residual impurities. In bcc metals, muons are anisotropic defects both at tetrahedral and octahedral

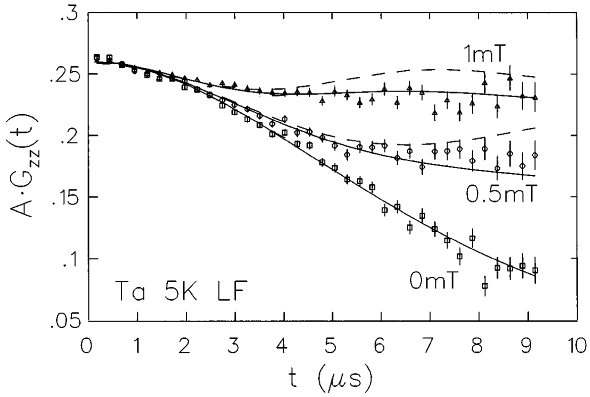

FIG. 2. Time-resolved muon spin polarization at 5 K under a longitudinal field of 0, 0.5, and 1 mT. Solid curves are the best fit results with a dynamical Kubo-Toyabe function. Dashed curves are calculated with a set of parameters to reproduce the spectra in zero field with no hopping motion, which fails to reproduce those at finite fields.

interstitial sites which change their orientation upon each jump. Assuming that the energy asymmetry ξ induced by impurities takes different values among different muon-defect orientations, we approximate the muon hopping rate as

$$\nu(T) = \sum_{i=1}^{n} \nu_i(T) = \sum_{i=1}^{n} 2J_i^2 \frac{\Gamma_i(T)}{\xi_i^2 + \Gamma_i^2(T)}, \qquad (7)$$

where J_i is the effective tunneling matrix and $\Gamma_i(T)$ is the damping factor for the jump to the site i with an energy asymmetry ξ_i. The steep temperature dependence of ν near the peak indicates the remaining contribution of two-phonon scattering to the level broadening, which can be taken into account by assuming

$$\Gamma_i(T) \simeq \Omega(T) + c_i\left(\frac{T}{\Theta_D}\right)^{m_i}, \qquad (8)$$

where c_i is the coupling constant, Θ_D is the Debye temperature, and m_i is the power which may depend

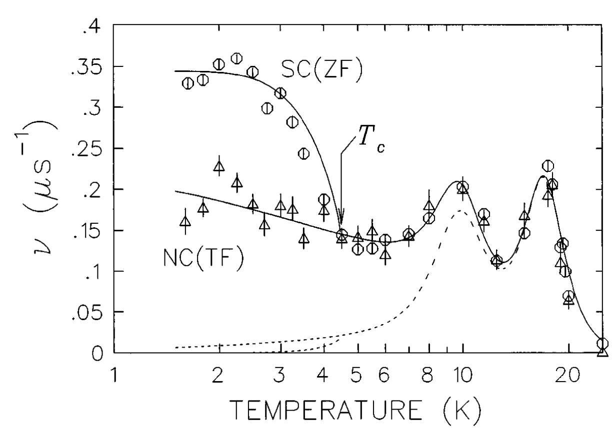

FIG. 3. Muon hopping rate in pure Ta deduced from ZF- and TF-μSR spectra below 30 K. Solid curves are calculated with a model described in the text, in which the contribution predominant above 5 K is shown by dashed curves.

TABLE I. Parameter values in Eqs. (6)–(8) to get the solid curves in Fig. 3, where $K = 0.06$ and $\Theta_D = 240$ K were assumed. Trianglar distribution $p(\xi_3) = (\xi_0 - |\xi_3|)/\xi_3^2$ was assumed for ξ_3 (see text).

	J_i (K)	c_i	m_i	ξ_i (K)	ξ_0 (K)
ν_1	0.027	6×10^9	9	72	⋯
ν_2	0.019	1×10^9	7	48	⋯
$\overline{\nu}_3$	0.0046	1×10^8	7	⋯	2.4

on i. The leveling off (or broad peak) of ν in the superconducting state is attributed to the inhomogeneity of ξ which must be important for small ξ [24]. Since the muon hops only a few times within the experimental time range of observation, the average hopping rate can be written as

$$\overline{\nu}_i(T) = \int p(\xi_i)\nu_i(T)d\xi_i\,, \tag{9}$$

where $p(\xi_i)$ gives the probability distribution for ξ_i. The solid curves in Fig. 3 are examples calculated by Eq. (7), assuming three components ($n = 3$) with the parameter values given in Table I and a simple triangular distribution $p(\xi_3) = (\xi_0 - |\xi_3|)/\xi_3^2$ $(-\xi_0 \leq \xi_3 \leq \xi_0)$ for the component ν_3 which dominates below $\sim$5 K. The value for K was taken from that of H in Nb [4]. Note that the sum of two components predominant above 5 K ($\nu_1 + \nu_2$, represented by the dashed curves in Fig. 3) have little contribution at lower temperatures. Thus, the observed effect of superconductivity is mainly on the third component, where the impurity effects play a minor role. The muon tunneling matrix elements are deduced to be 10 mK in order of magnitude (see Table I), consistent with the use of Eq. (3) which is valid for $J \ll \Delta_S$.

In conclusion our result proves that the muon hopping rate is enhanced below the superconducting transition temperature in high-purity Ta. This clearly establishes that the low energy infrared couplings to the conduction electrons control the long range quantum diffusion of a charged particle in a metal.

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