Muonium localization in solid krypton

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Muonium spin relaxation in zero, longitudinal, and transverse magnetic fields has been studied in solid and liquid krypton in the temperature range of 2 to 120 K. In the solid at low temperatures, the spin dynamics exhibit features characteristic of a magnetically dilute crystal, permitting measurements of exceptionally low muonium diffusion rates. At the lowest temperatures, the static Kubo-Toyabe relaxation function is found to describe muon spin relaxation in the *atomic* muonium state, indicating strong interstitial localization in the Kr lattice; muonium is determined to be localized at the tetrahedral interstitial position. At high temperatures, muonium diffusion in solid Kr exhibits a nonclassical behavior.

Rare-gas solids (RGS) form a group of cryocrystals characterized by very weak van der Waals interatomic interactions. These substances are among the simplest solids and the most amenable to theoretical modeling. They have similar phonon spectra with very low Debye temperatures.[1] All RGS with the exception of helium crystallize in a face-centered-cubic structure. The extremely low potential barriers for neutral interstitial particles in these materials make them ideal lattices for studying quantum transport phenomena.[2–4] For these purposes, the pseudoisotope of hydrogen known as muonium ($\mathrm{Mu}=\mu^{+}+e^{-}$) is the ideal choice for a diffusing particle, both by virtue of its light mass—almost an order of magnitude smaller than that of hydrogen (protium) itself—and by virtue of the ease with which its mobility can be measured *via* the muon spin relaxation (μSR) technique.[5] Although the bandwidth characterizing this motion may be extremely small (it must be larger than about 10^{-6} K to be detectable by μSR), particle delocalization should inevitably take place in perfect crystals at low temperatures, as a consequence of translational symmetry. This has been confirmed experimentally for muonium diffusion in alkali halides and compound semiconductors.[6]

On the other hand, strong localization of interstitial muonium at low temperatures has been observed in cryocrystals of solid nitrogen.[7] This contrasting behavior could be associated with the rotational ordering of this molecular crystal at low temperatures. However, muonium localization at low temperatures has most recently been revealed in studies of solid xenon.[8] This result is intriguing, in view of the absence of any rotational or structural crystal transitions, and has yet to be understood. The question arises as to whether localization of particles in this mass range at low temperatures is a general feature of "soft" cryocrystal lattices.

An understanding of the possible mechanisms of particle localization at low temperatures is also very important for matrix isolation spectroscopy, as the manner in which RGS are easily doped with guest atoms and molecules by codeposition on a cold substrate forms the basis for this widely used technique.[9,10] The advantage of RGS as hosts is that the characteristic electronic and vibrational states of the guest are almost unperturbed by the host matrix. The absence of guest diffusion at low temperatures could rule out the possibility of pairwise interactions which could perturb the isolated guest state. Of course, μSR measurements can only detect motion on a time scale up to about 100 μs, so we can only speculate about *muonium* motion over many seconds; however, the heavier atoms or molecules used in such experiments are unlikely to diffuse as rapidly as muonium.

There are two most important effects governing interactions of the diffusing particle with the medium. The first of these is the *polaron effect* (PE), which may lead to complete

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localization of the tunneling particle in a crystal despite the presence of the translational symmetry, but which turns out to be much weaker for neutral interstitial atoms in insulators[3] than for charged particles in metals.[11] (In any case, although the ‘‘bare’’ particle may be unable to tunnel due to the PE, the polaron itself—a quasiparticle consisting of the original particle plus an accompanying lattice distortion—may have a small tunneling bandwidth of its own.) The second is the effect of *fluctuational preparation of the barrier*,[12] which can decrease the effective barrier for the optimal particle path. This effect causes an increase in the tunneling bandwidth and can become dominant in the case of phonon coupling in insulators,[8] whereas in the presence of coupling to conduction electrons this barrier reduction is insignificant.[4] The first measurements of muonium tunneling kinetics in solid xenon[8] showed that both effects could be important at temperatures comparable with the Debye temperature of the crystal, Θ. An unprecedented variation of the muonium hop rate over four decades in solid Xe clearly demonstrated the tendency of muonium atoms to become localized at low temperatures. However, interpretation of the longitudinal field data at low temperatures in solid Xe (i.e., the regime of muonium localization) is hampered by a rather complicated multiexponential form of the muonium spin relaxation function. A strong hyperfine interaction between the muonium electron and the xenon nuclei (almost half of which have nonzero magnetic moments) hinders transverse field[13] (TF) and zero-field (ZF) μSR measurements at low temperatures. In consequence, muonium localization and slow tunneling have not previously been studied experimentally in RGS.

In this paper we describe an experimental study of muonium localization at low temperature in a rare-gas crystal, presenting data for solid krypton. This study is made possible by the low natural abundance of the sole isotope (^{83}Kr) to carry a magnetic moment: distinctive ZF and weak longitudinal field (LF) μSR spectra characterize the extremely slow muonium dynamics in the Kr lattice.

The effective-spin Hamiltonian of muonium in solid krypton is taken to be of the form[7]

$$\mathscr{H}=hA\mathbf{S}_e\cdot\mathbf{S}_\mu-g_e\mu_B\mathbf{S}_e\cdot\mathbf{H}-g_\mu\mu_\mu\mathbf{S}_\mu\cdot\mathbf{H}+h\sum_n\delta\mathbf{S}_e\cdot\mathbf{S}_n-\sum_n g_n\mu_n\mathbf{S}_n\cdot\mathbf{H}, \quad (1)$$

where A is the muonium hyperfine (HF) frequency, $\mathbf{H}$ is the external magnetic field and the $\mathbf{S}$, g, and μ terms are, respectively, the spins, g factors, and magnetic moments of the various particles. The summations are over all nearby nuclei. The nuclear hyperfine interaction (NHI) between the muonium electron and neighboring nuclear dipoles is characterized by the frequency δ. The NHI term is represented as isotropic in Eq. (1) and so may represent either an average magnetic dipole interaction, in a local-field approximation, or a contact interaction, in the event that the muonium electron's wavefunction overlaps with the surrounding nuclei. The muonium HF frequency in solid Kr has been reported to be $A = 4462.9(3.7)$ MHz,[14] which is virtually identical to the vacuum-state value $A_{\mathrm{vac}}=4463$ MHz. The NHI frequency δ for interstitial muonium in solid krypton has not previously been measured. In solid xenon this parameter was determined[8] to be about 1% of A. Considering that natural Kr has a natural abundance of isotopes with nonzero nuclear magnetic moments that is about four times less than that of Xe, one might expect an even smaller ratio of δ to A in krypton. If muonium and hydrogen occupy equivalent lattice sites, the NHI frequency for Kr nuclei adjacent to muonium in solid krypton is expected to be only slightly different (due to zero point motion) from the value for those adjacent to interstitial hydrogen (protium) itself, which has been measured by electron-spin resonance[15] to be about 15 MHz. It is this interaction which sets the time scale for muon spin relaxation.

Qualitatively, modulation of the nuclear hyperfine interactions results in relaxation of the muonium electron spin, which in turn leads to depolarization of the muon spin *via* the muonium hyperfine interaction.

In transverse field, the relaxation rate T_2^{-1} of the muonium precession signal has a simple form in two limits: if muonium ‘‘hops’’ from site to site at a rate τ_c^{-1} which is much larger than the NHI frequency δ (*fast hopping* limit), then the transverse relaxation rate is given by $T_2^{-1}\approx\delta^2\tau_c$, which is proportional to the effective width of the local-field distribution due to nuclear hyperfine interactions, motionally averaged (hence ‘‘narrowed’’) by fast muonium diffusion with $\tau_c^{-1}\gg\delta$. For very *slow* diffusion ($\tau_c^{-1}\lesssim\delta$) muon spin relaxation takes place on a time scale shorter than τ_c and $T_2^{-1}\approx\delta$. (For this reason, the parameter δ is sometimes referred to as the ‘‘static width’’ due to NHI.) Slow muonium dynamics cannot, therefore, be studied by transverse field measurements.

The interpretation of longitudinal field (LF) measurements of the muonium spin relaxation rate (T_1^{-1}) is based on the notion that the nuclear hyperfine interactions may be treated as an effective magnetic field acting on the muonium electron. Muonium diffusion causes fluctuations of this effective field which induce transitions between the coupled spin states of the electron and muon. The resultant muon depolarization is revealed in the forward-backward asymmetry of the muon decay. Such measurements allow values of τ_c^{-1} and δ to be determined independently.[16] A general expression involves various transitions between the coupled spin states[17] but a reasonable approximation is obtained by assuming dominance of the lowest frequency transition (within the muonium triplet spin states), leading to the expression

$$T_1^{-1}=\frac{2\delta^2\tau_c}{1+\omega_{\mathrm{Mu}}^2\tau_c^2}, \quad (2)$$

where $\omega_{\mathrm{Mu}}=\gamma_{\mathrm{Mu}}H$ is the muonium intratriplet transition frequency in the magnetic field ($\gamma_{\mathrm{Mu}}/2\pi=1.4012$ MHz/G). This approach is restricted to the limit of relatively high longitudinal fields, however, since the effective magnetic field approximation is valid only if $\gamma_e H\gg\delta$, where γ_e is the electron gyromagnetic ratio. Moreover, in the limit of slow muonium hopping and high magnetic field, T_1^{-1} is generally too slow [see Eq. (2)] to be measured by the standard μSR technique. Thus high LF measurements, which are very sensitive to muonium dynamics in the fast fluctuation regime, are rather ineffective for the study of very slow dynamics.

Zero-field (ZF) and weak longitudinal field (wLF) measurements turn out to be much more sensitive to slow muonium dynamics, at least in the case where the spin response is determined by dipolar interactions with neighboring nuclei. This is due primarily to the fact that these techniques involve the *full* dipolar Hamiltonian rather than just the *secular* part (as is adequate for the transverse field technique; see, for example, Ref. 18). A classical treatment[19] gives an analytical expression for the time evolution of the muon polarization function in zero field, known as the Kubo-Toyabe function: for a Gaussian distribution of static local fields, with second moment $\propto \Delta^2$,

$$P(t) = \frac{1}{3} + \frac{2}{3}(1-\Delta^2 t^2)\exp\left(-\frac{1}{2}\Delta^2 t^2\right). \quad (3)$$

An important feature of Eq. (3) is the "1/3 tail" which, roughly stated, reflects the fact that 1/3 of the muon polarization is, on average, aligned parallel to the local field while 2/3 is transverse to the local field and thus precesses. Although this function[19] was adopted and experimentally confirmed for μ^+ spin relaxation,[20] it is equally applicable for the triplet state of muonium, which can be treated in low magnetic field as one particle with spin $S=1$. Very slow diffusion, in which the muonium atom jumps between sites with uncorrelated arrangements of neighboring nuclear spins, is manifest in an observable relaxation of this "1/3 tail."[19,20] A "strong-collision" treatment[21,20] gives a relaxation function in this regime which may be written (for times $t \gg \Delta^{-1}$) as

$$P(t) \approx \frac{1}{3}\exp\left(-\frac{2}{3}\frac{t}{\tau_c}\right). \quad (4)$$

This expression for $P(t)$, being independent of Δ, may be used for *direct* measurements of very slow muonium hop rates.

In weak longitudinal field (wLF), the "tail" of the relaxation function which is sensitive to slow dynamics is shifted to earlier times and has an amplitude several times higher than the "1/3 tail" of the zero-field function.[22] This makes the wLF technique even more sensitive than ZF for measurements of extremely slow muonium hop rates.

In this paper we present the first observation of Kubo-Toyabe relaxation for a paramagnetic atom. The use of both ZF and wLF techniques allows the determination of particle hop rates two orders of magnitude slower than the inverse μ^+ lifetime, in a regime previously inaccessible.

The experiments were performed on the EMu beamline of the ISIS Pulsed Muon Facility at the Rutherford Appleton Laboratory and on the M20 beamline at TRIUMF. At ISIS, ultrahigh-purity krypton (less than 10^{-5} impurity content) was condensed from the gas phase into a liquid and then frozen into a disc-shaped cell (20 mm in diameter and 5 mm deep) The same krypton, recovered after the ISIS experiments, was used subsequently at TRIUMF, where an absolutely transparent crystal (22×22×6 mm) was carefully grown by applying a vertical temperature gradient of about 5 K across the cell for about 6 h. At both laboratories, positive muons of 28 MeV/c momentum and 100% spin polarization were stopped in the solid Kr and μSR spectra recorded at various different temperatures and applied magnetic fields.

Formation of atomic muonium in solid and liquid Kr (melting point 115.95 K) was detected by observing the precession signal at the characteristic muonium frequency $\omega_{Mu} = \gamma_{Mu} H$ in a transverse magnetic field. Typical TF-μSR spectra in solid Kr are shown in Fig. 1.

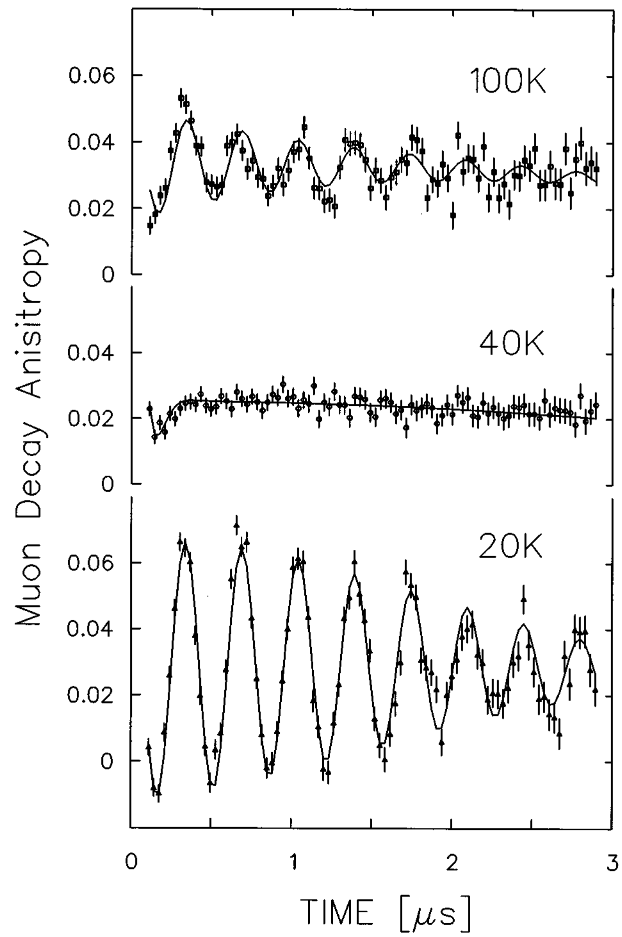

FIG. 1. Temperature dependence of μSR spectra in a transverse magnetic field $H=2$ G in solid krypton.

The diamagnetic fraction—that is, the proportion of muons which reach a state where they experience no contact hyperfine interaction from unpaired electron spin density—accounted for about 25% of the total polarization. In condensed Kr this fraction is believed to correspond to the molecular ion $KrMu^+$, since formation of the analogous species $NeMu^+$ has been suggested in gaseous neon[23] and direct experimental evidence for formation of the molecular ion N_2Mu^+ has recently been obtained[24] in solid nitrogen. However, some part of the diamagnetic fraction could also be due to muons stopped in the metallic sample cell. (This explanation is consistent with the very small diamagnetic fraction reported by Kiefl *et al.*[14])

Figure 2 presents the temperature dependence of the muonium transverse relaxation rate T_2^{-1} in solid and liquid krypton, extracted from the data by fitting single-exponential relaxation functions. Such fits were for the most part satisfactory, although it should be noted that at 100 K the relaxation was better described by a two-component function (see Fig. 1). The limit on timing resolution imposed by a pulsed muon source does imply that a fast initial component could be overlooked in the ISIS data. Two-component muonium relaxation has also been reported by Kiefl *et al.*[14] in solid Kr at 90 K (the only temperature investigated in that

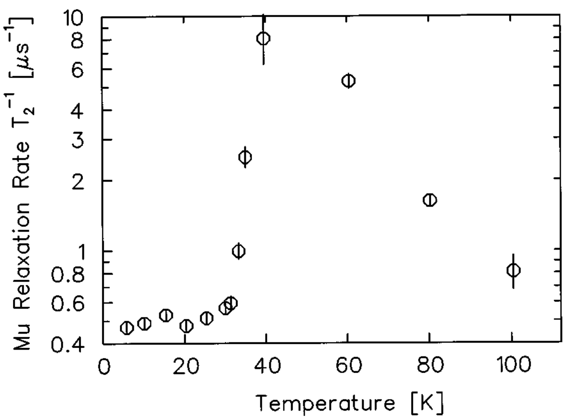

FIG. 2. Temperature dependence of the muonium relaxation rate T_2^{-1} in solid Kr in a transverse magnetic field $H=2$ G.

work) and attributed by those authors to inhomogeneous muonium diffusion. An alternative explanation could be proposed involving interactions of Mu atoms with paramagnetic species liberated in the incoming muon's ionization track.[25] An additional fast-relaxing component has also been observed at high temperatures in the muonium relaxation function for a number of other substances, such as solid xenon,[8] solid argon,[26] ice,[27] etc.

The main mechanism of muonium relaxation in solid Kr undoubtedly involves dipolar or hyperfine interactions between the Mu spin and ^{83}Kr nuclei (natural abundance 11.55%). The increase in T_2^{-1} upon lowering the temperature from 100 to 40 K may be explained by a slowing down of the muonium diffusion in solid Kr. It would be tempting to explain the characteristic maximum and abrupt drop in T_2^{-1} at about 40 K in terms of motional averaging of these interactions due to fast muonium diffusion, which would imply a rather abrupt onset of quantum tunneling at low temperatures. This drop, however, is accompanied by a drop in the amplitude of the TF precession signal from 0.0671(5) to 0.0395(5), corresponding to a reduction of the apparent muonium fraction to 58.9% of its value at higher temperatures. This behavior cannot be explained by the onset of a fast diffusion regime. It can, on the other hand, be explained in terms of the isotopic composition of krypton and the consequent fact that not all crystallographically equivalent sites have equivalent local nuclear magnetism. Those muonium atoms which localize at sites having nearest-neighbor ^{83}Kr nuclei experience strong nuclear hyperfine interactions and are rapidly and completely depolarized, whereas those which localize at sites with spinless nearest neighbors (experiencing much smaller, and predominantly dipolar, local fields from remote ^{83}Kr nuclei) exhibit a relatively slowly damped precession signal. An abrupt decrease of T_2^{-1} with decreasing temperature, accompanied by a simultaneous reduction in the precession amplitude, can therefore be explained by *localization* of the *whole* muonium fraction in solid Kr at low temperatures. Above about 35 K, the muonium atoms become delocalized and the entire Mu ensemble experiences close-range interactions with ^{83}Kr nuclei in the course of diffusion through the lattice, leading to the stepwise increase in $T_2^{-1}(T)$. If this explanation is correct, the nature of the muonium localization site in the Kr lattice can be determined by comparison of the muonium fraction which remains observable at low temperature (58.9%) with the probability of finding sites of different symmetry having spinless nearest neighbors. Assuming a uniform random distribution of ^{83}Kr, the probability that a tetrahedral site should have all four near neighbors spinless is 61.2%, while the probability that an octahedral site should have all six nearest neighbors spinless is 47.9%. It seems most likely, therefore, that muonium occupies the tetrahedral interstitial position in the Kr lattice. A substitutional position can reasonably be discounted, in view of the very low probability that all 12 nearest neighbors should be spinless. (This contrasts with the situation for codeposited hydrogen atoms, which do apparently take up the substitutional position[15]—the NHI frequencies in the vicinity of muonium and hydrogen atoms in solid krypton could therefore be considerably different, as our LF measurements reveal; see below.)

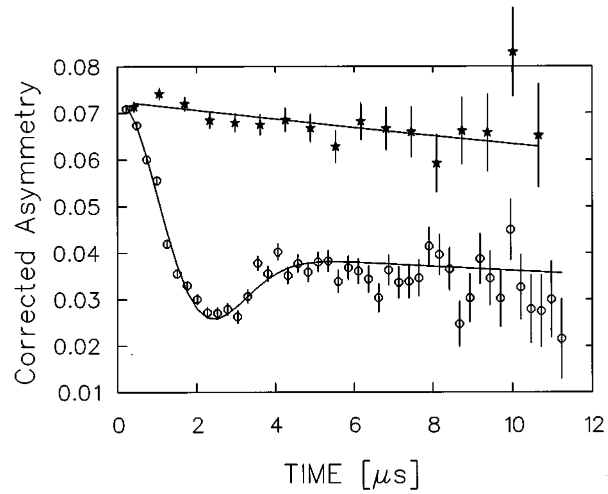

FIG. 3. Time dependence of the muonium polarization in solid Kr at $T=20.3$ K in zero magnetic field (circles) and in a weak longitudinal magnetic field $H=5$ G (stars). Note the Kubo-Toyabe form of the muon polarization function in zero magnetic field.

In order to confirm the hypothesis of muonium localization at low temperatures, we undertook zero-field measurements at $T=20.3$ K. Figure 3 presents the time evolution of the muon polarization (circles) and shows that it exhibits a relaxation function of the static Kubo-Toyabe type [see Eq. (3)]. The same relaxation function was observed in solid Kr at temperatures below 20 K down to the lowest measured temperatures. This can only be attributed to essentially static interstitial muonium. The line drawn through the experimental points represents a best fit to a "dynamicized" version[28,22] of Eq. (3), taking into account both sites with nearest neighbor ^{83}Kr nuclear spins and those without. The former contribute a Kubo-Toyabe function with its characteristic minimum shifted to very early times (invisible in the dead time of the μSR spectrometer); the "1/3 tail" of this component constitutes a baseline to the Kubo-Toyabe function actually observed, which is characteristic of the local-field distribution from more distant dipolar nuclei at the latter type of sites whose immediate neighbors are spinless. The rather long correlation time τ_c is assumed to be the same for both types of sites, since they are electrostatically identical. For comparison, Fig. 3 also shows the muonium polarization function in solid Kr for a longitudinal field of 5 G (stars).

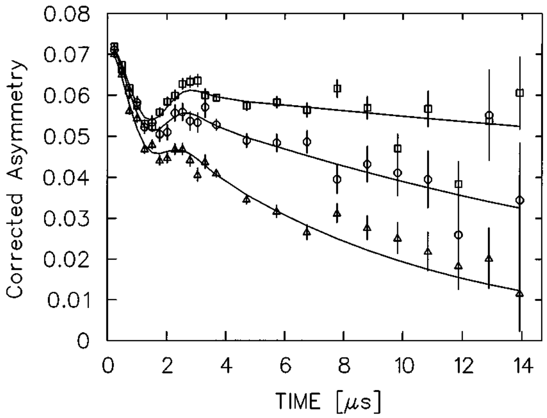

FIG. 4. Muonium polarization time spectra in solid Kr in a very weak longitudinal magnetic field $H = 0.22$ G at different temperatures: 25 K (squares), 28 K (circles) and 30 K (triangles).

Very slow muonium diffusion rates were measured both in zero field and in a *very* weak longitudinal field (0.2 G). Typical wLF time spectra are shown in Fig. 4. Both the hop rate τ_c^{-1} and the NHI frequency δ_{nnn} (with next-nearest neighbors) were determined by simultaneously fitting ZF and wLF data to corresponding dynamical Gaussian Kubo-Toyabe relaxation functions[28,22] with $\Delta \equiv \delta_{\mathrm{nnn}}$ [recall Eq. (3)].

At temperatures above about 30 K, the high LF technique was applied to measure the faster hop rates. The temperature dependence of T_1^{-1} in different values of applied field is shown in Fig. 5. Between about 30 and 55 K, the LF relaxation data can safely be interpreted in terms of muonium diffusion, as the variation of T_1^{-1} with magnetic field is consistent with a diffusion model.[16] This model describes the time evolution of the muon polarization in a diffusing muonium atom under the influence of nuclear hyperfine interactions [see Eq. (1)]. The basic idea of this model is that the nuclear hyperfine interaction is treated as an effective magnetic field acting on the muonium electron. The modulation of this field as muonium diffuses through the lattice produces transitions between different coupled muon-electron spin

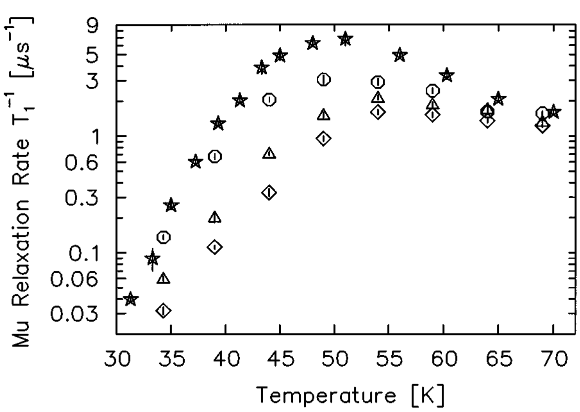

FIG. 5. Temperature dependence of the muonium relaxation rate T_1^{-1} in solid Kr in several longitudinal magnetic fields: 40 G (stars), 60 G (circles), 120 G (triangles), and 180 G (diamonds). Note the T_1 minima (T_1^{-1} maxima) around 50 K.

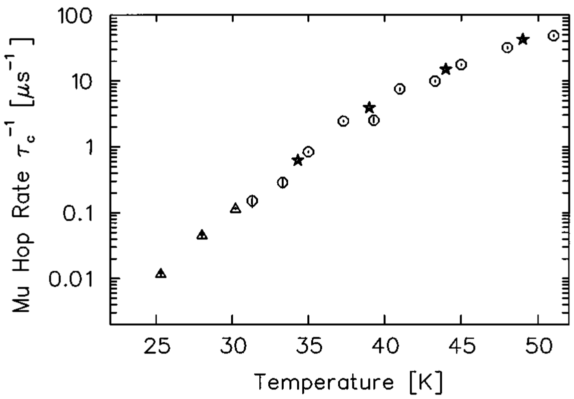

FIG. 6. Temperature dependence of the muonium hop rate derived from simultaneous fits to ZF and wLF data (triangles, ISIS data) and high LF data (circles, ISIS data; stars, TRIUMF data).

states, resulting in muon spin depolarization. The model[16] takes into account all four allowed transitions between muon-electron states in high LF. Since the muon polarization function turns out to be a sensitive function of applied field, one can easily draw conclusions about the validity of this model from the experimental results. Above about 55 K, it appears that the relaxation is no longer governed by local nuclear hyperfine interactions; a similar change of regime has also been observed in solid natural xenon and solid ^{136}Xe.[8]

Clear T_1^{-1} maxima (or, in NMR parlance,[18] "T_1 minima") are seen around 55 K, representing the condition for fastest relaxation: $\tau_c^{-1} = \omega_{ij}$, where ω_{ij} are the muonium hyperfine transition frequencies. For lower LF the T_1 minima are shifted to lower temperatures, indicating a gradual decrease of muonium mobility with decreasing temperature, which is consistent with the increasing localization seen in the ZF and wLF data. The motional correlation time τ_c and NHI frequency δ_{nn} (with nearest neighbors) were determined by simultaneous fitting of the LF-μSR spectra taken at several fields to a general expression.[16] The positions of the T_1 minima for several fields provided an absolute calibration for both parameters.[18]

Muonium NHI frequencies with nearest and next-nearest neighbors were determined to be $\delta_{\mathrm{nn}} = 55(5)$ MHz (from LF data) and $\delta_{\mathrm{nnn}} = 0.67(6)$ MHz (from ZF and wLF data), respectively. This difference of almost two orders of magnitude between nearest and next-nearest neighbor interactions with ^{83}Kr nuclear moments suggests that these interactions are, respectively, contact and dipolar in character. The large value for the nearest neighbors is at the upper limit of values encountered for purely dipolar interactions; we therefore suspect that the muonium wave function actually overlaps onto neighboring krypton nuclei to give a contact term. Apart from this somewhat surprising result, treatment of the zero-field data with the Kubo-Toyabe relaxation function is almost certainly justified, since $\Delta \equiv \delta_{\mathrm{nnn}}$ surely has a dipolar origin.

The temperature dependence of the muonium hop rate in solid Kr derived from simultaneous fits to ZF and wLF data (triangles) and high LF data (circles, measured at ISIS, and stars, measured at TRIUMF) are shown in Fig. 6. The hop rate values obtained show very good agreement between high LF data and ZF and wLF data around 30 K. The low-temperature results represent the slowest interstitial hop rates ever measured by μSR, either for μ^+ or for neutral muonium. Below about 25 K, the muonium τ_c is some two orders of magnitude longer than the muon lifetime. Above 25 K, the muonium hop rate shows an increase of almost 4 orders of magnitude with rising temperature. The nature of the muonium diffusion in the temperature range 25 K$<T<$55 K is discussed below.

It is known that the transition from the classical diffusion regime to the quantum regime takes place at a temperature $T^* \sim \nu_0$, where ν_0 is the vibrational frequency of the particle in its potential well.[29] For muonium atoms in crystals, T^* can be estimated to be on the order of several hundred Kelvin. It is also known that one-phonon quantum diffusion[8] takes over from the low-temperature two-phonon quantum diffusion[7] above a few tenths of the Debye temperature.[4] As the Debye temperature for solid Kr is $\Theta = 72$ K,[1] muonium diffusion in the above-mentioned temperature range in solid Kr is expected to be governed by the one-phonon quantum mechanism. The alternative possibility of classical overbarrier hopping, with the Arrhenius dependence

$$\tau_c^{-1} = \tau_0^{-1} \exp(-E/T) \tag{5}$$

requires the pre-exponential factor τ_0^{-1} to be comparable with ν_0. Fitting the experimental $\tau_c^{-1}(T)$ dependence for 25 K$<T<$55 K to Eq. (5) gave a pre-exponential factor $\tau_0^{-1} = 1.6(3) \times 10^{11}$ s^{-1}, which is at least two orders of magnitude less than ν_0. Classical diffusion may therefore be eliminated as a possible mechanism of muonium diffusion in solid Kr at temperatures below 55 K. Analysis of the diffusion rate in this region showed that both the ''polaron effect'' and ''fluctuational preparation of the barrier'' should be taken into account in this material, in the framework of the one-phonon quantum diffusion mechanism. These effects have previously been shown to be important for muonium diffusion in KCl,[30] solid xenon,[8] and solid nitrogen.[31] Unfortunately, the muonium relaxation mechanism governed by nuclear hyperfine interactions is masked in solid Kr by the onset of additional relaxation mechanisms at temperatures comparable to and above the Debye temperature of the crystal. This circumstance hampers a quantitative separation of the polaron effect from the effect of fluctuational preparation of the barrier, the latter of which should dominate in the temperature regime near and above Θ. Precise determination of the parameters for one-phonon muonium diffusion in solid krypton therefore requires more detailed measurements in high longitudinal fields.

In conclusion, our data provide direct evidence for the localization of atomic muonium in solid krypton at low temperatures (below 25 K). The site adopted is identified as the tetrahedral interstice. At higher temperatures (25–55 K), dynamic relaxation of the muon spin proceeds by modulation of nuclear hyperfine interactions, as the muonium begins to diffuse through the lattice; the diffusion mechanism is supposed to be one-phonon assisted quantum hopping. In the temperature range above about 55 K, another process of muon spin relaxation comes into play which is as yet unidentified.

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