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Muonium diffusion in solid CO₂

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The quantum diffusion of interstitial muonium atoms in solid CO₂ is studied in the temperature range from 5 to 200 K using the technique of muonium spin rotation and relaxation. Muonium exhibits coherent bandlike dynamics between 140 and 160 K. At low temperature Mu undergoes strong localization. © 2001 American Institute of Physics. [DOI: 10.1063/1.1414576]

When positive muons are implanted into insulators they often form muonium (μ^+e^- or Mu) atoms, analogous to hydrogen atoms but nearly an order of magnitude lighter.¹ The dynamics of such light atoms, being intrinsically quantum mechanical in nature, is of special interest because it provides critical tests of quantum diffusion theories.^{2–4} The basic issue in nonclassical transport is whether a wavelike or particlelike description is appropriate, i.e., whether the tunneling is coherent or incoherent. This depends on whether the interaction with the environment is such as to lead to spatial localization of the wave function or to bandlike (Bloch wave) motion. One of the possible channels for localization of a particle is through its interaction with lattice excitations (phonons, librations, magnons, etc.). In a dissipative environment^{5,6} the lattice excitations can be represented as a bath of harmonic oscillators; interaction with this environment causes a crossover from coherent quantum tunneling to incoherent hopping dynamics when the particle “dressed” with the lattice excitations can be effectively thought of as a polaron.

At low temperatures, the environmental excitations are frozen out. In this case, conventional understanding suggests that the only possible channel for particle localization is the introduction of crystal disorder, which thus may dramatically change the transport properties of a solid. A well-known example is the spatial localization of electron states near the Fermi level in a disordered metallic system, which leads to a transition into a dielectric state (the Anderson transition):⁷ coherent tunneling of a particle is possible only between levels with the same energy (e.g., between equivalent sites in a crystalline lattice); in the case of strong randomness, states with the same energy may be too spatially separated for tunneling to be effective.

Although the concept of localization by disorder has been introduced primarily in order to describe the electronic transport properties of solids, it may also be applied to the quantum dynamics of heavier particles, whether charged or neutral.^{4,8,9} Recent experimental results for positive muons (μ^+) in dilute metallic alloys,¹⁰ as well as for muonium atoms ($\text{Mu} = \mu^+ + e^-$) in “dirty” insulators,^{11,12} have clearly indicated that crystal disorder introduced by impurities dramatically changes the nature of quantum diffusion for particles ~ 200 times heavier than the electron. In these experiments the environmental coupling could be varied or switched on and off by changing the temperature—for interstitial muons in aluminum metal,¹⁰ the low-energy couplings to conduction electrons are suppressed below the superconducting transition, as a BCS energy gap opens at the Fermi surface, while for muonium in insulators^{11,12} the inelastic interactions with phonons are frozen out at low temperatures. Similar arguments apply for the well-known system of a dilute solution of heavy ³He atoms in a ⁴He crystal.^{13,14}

A common and crucial feature of all the above-mentioned experiments is that the particle tunneling bandwidth Δ is small compared with all other relevant energy parameters, especially U , the characteristic shift in the particle’s energy level (relative to its value in the perfect lattice) due to crystalline disorder. For elastic strains, U is expected to vary as $U_0(a/r)^{-3}$ with the distance r from a point defect, where U_0 is the maximum shift corresponding to a site one lattice constant a away from the defect center. (Of course, U actually has discrete values near the center; this continuous formulation is only meaningful when $r \gg a$.) For example, typical values of $U_0 \sim 10^3$ K exceed muon bandwidths in metals by 6–7 orders of magnitude; in insulators the difference is not so extreme, but still $U_0 \gg \Delta$.^{4,15} Under these cir-

cumstances, the influence of crystalline defects extends over distances $R \sim a[U_0/\Delta]^{1/3} \gg a$. The quantum diffusion of the interstitial particle is therefore dominated in these case by strong, long-range crystal disorder.

An essential characteristic of particle transport under conditions of long-range crystal disorder is its inhomogeneity at low temperatures,^{4,16,17} where the particle dynamics cannot be described in terms of a single correlation time τ_c . The particle hop rate is in this case predicted⁴ to follow

$$\tau_c^{-1}(R) \propto \frac{\tilde{\Delta}_0^2 \Omega(T)}{\xi^2(R) + \Omega^2(T)}, \quad (1)$$

where $\tilde{\Delta}_0$ is the renormalized tunneling amplitude, $\Omega(T)$ is the particle's energy level broadening due to coupling with lattice excitations, and $\xi(R) = a[dU/dr]_{r=R}$ is the difference between energy levels at two adjacent tunneling sites due to disorder. (We ignore the possibility of degenerate adjacent sites at the same R .) At low temperatures the phonon width is reduced and

$$\tau_c^{-1}(R) \propto \frac{\tilde{\Delta}_0^2 \Omega(T)}{\xi^2(R)}. \quad (2)$$

Equation (2) describes the dynamics of particles in the vicinity of defects, where the inhomogeneity of the crystal results in a spatial distribution of hop rates, $\tau_c^{-1}(R)$. Another fraction of particles, initially located so far from impurities or defects that $\tilde{\Delta}_0$ exceeds $\xi(R)$ [as well as $\Omega(T)$], undergoes bandlike propagation. In the limiting case of very low temperatures (or below a superconducting transition), inelastic scattering by phonons (or conduction electrons) is strongly suppressed. The two fractions cannot then exchange particles—they are manifest in the appearance of two distinct components in the muon polarization function $P(t)$;^{10–12} this may be taken as a signature of spatial inhomogeneity of the particle dynamics when the disorder is described by a long-range potential.

The foregoing discussion, like all previous theoretical treatments of quantum diffusion (see, e.g., Refs. 4, 8, and 9) implicitly assumed that the diffusing particle has access to only one site per unit cell.

In the harmonic approximation, the transport properties of a neutral particle in a simple crystalline insulator (e.g., a monoatomic or ionic crystal) depend only on the phonon modes of the lattice. For crystals composed of molecules, two additional contributions enter from the internal vibrational and rotational degrees of freedom of the molecules. Internal vibrations of molecules scarcely change the particle dynamics because of their extremely high frequencies. Molecular rotation, however, is a different matter. Two extremes are possible: the molecules may rotate almost freely in the crystal, or the rotational motion may be severely restricted and hence transformed into torsional excitations (librons). Since the typical rotational frequencies of molecules in crystals are still much higher than the particle bandwidth, in the first extreme the energy levels for a particle moving in different unit cells are degenerate, and therefore its dynamics remain unperturbed. In the second extreme the anisotropic interaction between molecules (which causes orientational ordering in the first place) changes the crystalline potential so that this degeneracy is lifted. As far as the particle dynam-

ics are concerned, this splitting of adjacent energy levels acts as an effective disorder, creating the bias ξ in Eq. (1). Here we report our study of muonium dynamics in solid CO₂, in which the anisotropic part of intermolecular interaction is so strong that the lattice keeps its orientational order in the entire solid phase.

Solid carbon dioxide was one of the first few solids (also quartz and ice) in which muonium atom formation was observed, about 30 years ago.¹⁸ However, there was no explanation for the mysterious muonium relaxation in CO₂. The muonium relaxation rate was measured in a transverse magnetic field of 7.2 Oe to be about $0.6 \times 10^6 \text{ s}^{-1}$. This relaxation rate, although not high, is, however, noticeable and cannot be associated with nonuniformity of the magnetic field, inasmuch as in the same experiment a few-times-lower relaxation rate has also been measured. Nor can the muonium relaxation in CO₂ be explained by its interaction with nuclear magnetic moments, as all of them are equal to zero in carbon dioxide.

The source of muonium relaxation in solid CO₂ can be understood if muonium is anisotropic. The reason for the muonium anisotropy is that in the environment of surrounding atoms of the medium, muonium's electron function is not necessarily the spherically symmetric $1S$ state. The effective spin Hamiltonian for static Mu in solid CO₂ in an external field \mathbf{H} , assuming an anisotropic muon-electron hyperfine interaction, has the form¹⁹

$$H = hA\mathbf{S}_e \cdot \mathbf{S}_\mu - g_e\mu_B\mathbf{S}_e \cdot \mathbf{H} - g_\mu\mu_\mu\mathbf{S}_\mu \cdot \mathbf{H}, \quad (3)$$

where A is the muonium hyperfine tensor, and \mathbf{S} , g , and μ are the spins, g factors, and magnetic moments of the respective particles. For example, in a variety of cubic semiconductors muonium centers are observed with a hyperfine interaction that is axially symmetric about any of the $[111]$ crystal axes. For a state of this type (which is the simplest case of anisotropic muonium), A is diagonal with elements A_{\parallel} and A_{\perp} . The essential feature of anisotropic muonium is that at zero magnetic field the muon spin will oscillate at a frequency which is equal to one-half of the anisotropic part of the hyperfine interaction ($A_{\parallel} - A_{\perp}$) divided by Planck's constant. In the general case (when A is a matrix of high order) several frequencies are observed.²⁰ The amplitudes of oscillations at these frequencies are functions of the angles between the principal axes of A (which are determined by the configuration of muonium sites in the crystalline cell) and the initial muon polarization. In the case of muonium diffusion this will result in apparent muonium relaxation.²¹ The hopping sites are equivalent, but have principal axes making different angles with the muon polarization direction; thus when a Mu atom hops to a new site its spin begins to oscillate with different amplitudes for all the frequency components, resulting in an effective relaxation mechanism on the time scale of a mean oscillation quarter-period. To date the theory of the time evolution of the anomalous muonium polarization function in the hopping regime is undeveloped. In quartz,²¹ anomalous muonium diffusion was described using a Kobo-Toyabe zero-field stochastic relaxation function with hopping,²² using a constant empirical "static width" of 4 times the highest oscillation frequency. In the current paper we present a way of estimating anomalous muonium hop

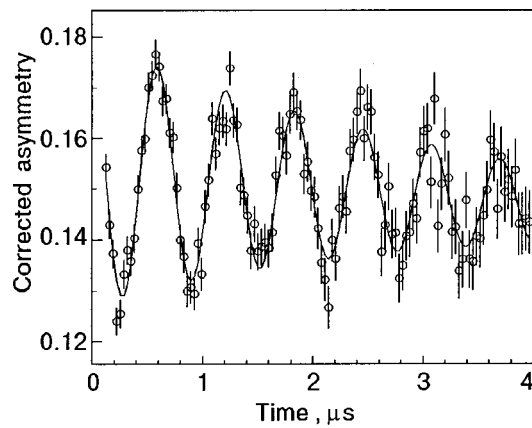


FIG. 1. Experimental μ SR spectrum in pure CO_2 in zero magnetic field at 40 K.

rates in a host with zero nuclear moments regardless of the exact notions about the muonium hyperfine tensor and depolarization function.

The experiments were performed on the EMu beamline of the ISIS Pulsed Muon Facility at the Rutherford Appleton Laboratory. Ultrahigh-purity CO_2 (less than 10^{-5} impurity content) was condensed from the gas phase into a liquid and then frozen into a disc-shaped cell (24 mm in diameter and 5 mm deep). Solid samples were carefully grown from the liquid phase at typical speeds of about 5 mm/h by applying a vertical temperature gradient of about 5 K across the cell. Positive muons of 28 MeV/c momentum and 100% spin polarization were stopped in the samples, and μ^+ spin rotation (SR) time spectra were recorded at various different temperatures and applied magnetic fields. Positrons from the muon decay are emitted preferentially along the direction of the muon polarization, so that the time-differential μ^+ SR technique, details of which can be found elsewhere,¹ produces direct measurements of the time dependence of the muon decay asymmetry $A(t)$, which is proportional to the muon polarization function $P(t)$.

The formation of anisotropic muonium in solid CO_2 was detected by observing the oscillation signal in zero magnetic field. A typical experimental spectrum in zero magnetic field at low temperature is represented in Fig. 1. Then a set of μ^+ SR spectra was measured in a weak transverse magnetic field $H=2\text{G}$. Transverse magnetic field spectra show a nice anisotropic muonium precession signal. In all the temperature interval the envelope of this precession is approximated by a simple exponential function $P(t) \sim \exp(-\lambda t)$. Usually the technique of muonium transverse relaxation is used to extract hopping rates of isotropic muonium in a host with nonzero nuclear moments. In this case the effective spin Hamiltonian of muonium in a crystal in an external magnetic field consists of electron, muon, and nuclear Zeeman interactions, the muonium hyperfine (HF), and the nuclear hyperfine (NHF) interactions (see, e.g., Ref. 15). Qualitatively, modulation of the NHF interactions results in relaxation of the muonium electron spin, which in turn leads to the observed muon depolarization via the muonium HF interaction. As the muonium HF constant A is usually several orders of magnitude higher than the NHF interaction (which for simplicity we characterize by a single parameter δ), it is the

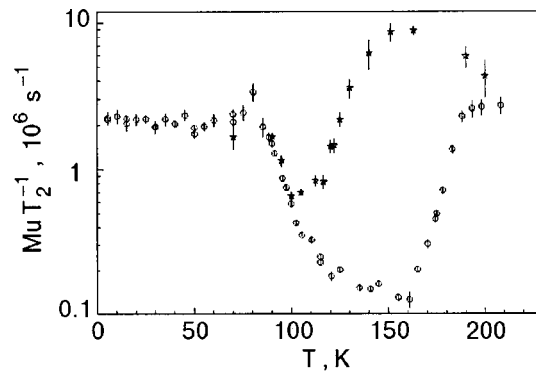


FIG. 2. Temperature dependence of muonium relaxation rate in solid CO_2 in weak transverse field $H=2\text{G}$ (circles: pure CO_2 ; stars: $\text{CO}_2 + 0.1\%$ N_2O).

latter which sets the time scale for isotropic muonium spin relaxation. In the case of anisotropic muonium, the hyperfine interaction changes from site to site or the HF interaction is modulated “by itself,” and the value of the muonium hyperfine interaction anisotropy about $2(A_{\parallel} - A_{\perp})/h$ will set the time scale for the anisotropic muonium spin relaxation. To make a connection with the standard theory of muonium diffusion this modulation can be presented by “effective” δ or $\tilde{\delta}$. In the standard theory the envelope of the muonium precession signal can be approximated by a simple exponential,

$$P(t) \sim \exp(-t/T_2), \quad (4)$$

where T_2 is the transverse relaxation time of the muonium spin. The muonium relaxation rate T_2^{-1} has a simple form in two limits: if muonium “hops” from site to site at a rate $\tau_c^{-1} \gg \tilde{\delta}$ (fast hopping limit), then the transverse relaxation rate is given by $T_2^{-1} \approx \tilde{\delta}^2 \tau_c$. For very slow diffusion ($\tau_c^{-1} \lesssim \tilde{\delta}$) the muonium spin relaxation takes place on a time scale shorter than τ_c , and $T_2^{-1} \approx \tilde{\delta}$. [In this case $P(t)$ is better approximated by a Gaussian relaxation function].

Figure 2 presents the temperature dependences of the muonium transverse relaxation rate T_2^{-1} in solid CO_2 and CO_2 plus 0.1% of N_2O impurities, extracted from the spectra by fitting single-exponential relaxation functions (4).

In pure CO_2 at high temperatures above 160 K, T_2^{-1} decreases with decreasing temperature, or the hop rate increases with decreasing temperature. This is an unambiguous manifestation of quantum diffusion which has also been recognized for muonium in KCl (Ref. 23) and solid nitrogen (Ref. 24). Such strong temperature dependences of the muonium relaxation at high temperatures in solid CO_2 cannot be due solely to the $\Omega(T)$ dependence in Eq. (1) in the regime of dynamical destruction of the band ($\Omega \gg \xi$), but must rather be caused by exponential renormalization of the tunneling amplitude, due to strong one-phonon scattering.

The muonium relaxation levels off at lower temperatures below about 160 K. This plateau must represent the onset of muonium band motion,^{4,15} which occurs if the coherence is preserved, $\Omega \ll \tilde{\Delta}_0$ (but still $T > \tilde{\Delta}_0$), and the disorder is weak, $\xi \ll \tilde{\Delta}_0$.

The band motion does not extend to the lowest temperatures, however: T_2^{-1} begins to increase with decreasing temperature below about 140 K. These data indicate that inter-

stitial muonium atoms undergo gradual localization in CO₂ at low temperatures where $\Omega \ll \xi$ and that the particle dynamics follow Eq. (2).

Experiments with doped CO₂ were performed to estimate the muonium hopping rate from the trapping regime of muonium diffusion. Transverse relaxation data in solid CO₂ with 0.1% N₂O impurities are represented in Fig. 2 by stars. When the Mu atom hops rapidly in CO₂ (causing low values of T_2^{-1} due to dynamical “narrowing”), it finds a N₂O impurity in the CO₂ crystal and reacts chemically (probably to form the MuNNO* radical),²⁵ which explains the fact that the maximum T_2^{-1} value for Mu in CO₂+N₂O significantly exceeds that for slowly hopping Mu in pure CO₂ at low temperatures, where it is determined by the anisotropy of Mu hyperfine tensor. This chemical reaction results in an exponential relaxation of the Mu polarization, the rate of which is determined by the time required for Mu to approach the N₂O impurity within a distance a of about one lattice constant, after which the reaction occurs immediately. Then the rate of trapping is²⁶

$$\lambda = 4\pi a D c, \quad (5)$$

where $D = a^2/4\tau_c$ is the muonium diffusion coefficient, and c is the concentration of impurities. From the maximum relaxation rate $T_{2\max}^{-1} \sim 10^7 \text{ s}^{-1}$ we can estimate $\tau_{c\max} \sim \pi c/T_{2\max}^{-1} \sim 3 \times 10^9 \text{ s}^{-1}$. For the bandlike regime the “hop rate” is given by $\tau_c^{-1} = 2\sqrt{2}\tilde{\Delta}_0$ (Ref. 27). By substituting $\tau_{c\max}$ we can estimate the muonium bandwidth in CO₂ to be about 10^{-2} K . This value may be compared with muonium bandwidths in KCl and solid nitrogen (0.16 and 10^{-2} K , respectively).¹⁵

At high temperatures the clear maximum in T_2^{-1} for Mu in CO₂+N₂O marks the crossover from fast to slow Mu diffusion near N₂O impurities. In this temperature range the strong coupling to phonons allows Mu to overcome the defect potential and move to react with N₂O. However, the energy shift ξ which the particle has to overcome is much larger close to the defect than far from it, making the Mu hop rate strongly dependent on the distance from the defect.

At low temperatures the suppression of inelastic interactions with the lattice changes Mu diffusion drastically—Mu atoms are stuck (or “frozen”) at some distance far from N₂O impurities, causing a strong reduction of the reaction rate (Mu relaxation rate). At very low temperature Mu atoms simply cannot approach N₂O impurities due to suppression of the inelastic interactions, and T_2^{-1} becomes the same in pure and doped crystals (Fig. 2).

In conclusion, we have presented evidence for the bandlike motion of light interstitial muonium atoms in CO₂ crystals.

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