Polarized Muon Spins in Pulsed Magnetic Fields: A New Method to Study Delayed Muonium Formation

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A transverse pulsed ($\lesssim 10$ mT) magnetic field method in μSR is presented and studied in detail in view of applications to experimental investigations of "delayed" muonium and muonium radical formation. The presence of longitudinal static fields for initial "spin-locking" purposes is analyzed and tested in the context of the adiabatic approximation. The crossover between the sudden and adiabatic regimes is measured for both muonium and free muons. The method also makes μSR experiments possible at frequencies above the natural bandwidth of pulsed beams.

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Fully spin-polarized positive muons are established probes of the magnetic and electronic properties of matter. The main experimental technique using implanted muons, known as muon spin rotation (μSR) [1,2], surveys the time evolution of the muon spins in a transverse magnetic field, once they thermalize in the sample. In this process muons adopt a variety of "chemical" configurations ranging from nearly free diamagnetic muons, (μ^+ with spin $s = 1/2$) to nearly free hydrogenlike muonium atoms, Mu $\equiv e^- + \mu^+$, whose electron-muon hyperfine levels are described by the Breit-Rabi diagram. At low fields, the transitions between muonium triplet states yield a single precession frequency characterized by the gyromagnetic ratio $\gamma_{\mathrm{Mu}}/2\pi = 13.945$ GHz/T, while μ^+ precesses at a much lower frequency due to its lower $\gamma_\mu/2\pi = 135.53$ MHz/T. Muonium, as a highly reactive paramagnetic species, often forms muonium adduct radicals, still paramagnetic but with reduced hyperfine coupling [3].

The formation of muonium may be "hot," with the prompt capture of electrons from the host material, or thermal, i.e., occurring when the muon has already thermalized. A debated and still largely unanswered question deals with the time scale over which these formation mechanisms take place. A frequent sign of thermal (or *delayed*) processes has been the existence of missing fractions of muonium/radical signals. Only recently some more direct evidence has been produced in condensed rare gasses and semiconductors by using electric fields [4,5]. Predictions for liquid hydrocarbons (typically n-hexane) are also available [6].

In conventional μSR experiments in static transverse magnetic fields a serious difficulty arises from the loss of phase coherence with time: the formation time, short for epithermal regimes (hot reactions), is no longer negligible at thermal regimes and a comparison with the precession period is needed to establish whether phase coherence is conserved or lost. If alternatively the magnetic fields were switched on suddenly at a controlled delay from the muon implantation time the phase coherence problem could be avoided.

In this Letter we demonstrate that such a pulsed field technique is experimentally possible when using pulsed sources of polarized muons which offer the possibility of applying a variety of pulsed excitations, such as the already established pulsed radio-frequency resonance [7]. In principle, both are equally applicable to the study of delayed muonium formation processes. Intuitively speaking, in a *delayed* formation process a *delayed* applied transverse magnetic field recovers the precession phase coherence that could otherwise be lost if the magnetic field were constantly present.

At the same time, the technique also overcomes the only drawback of a pulsed beam, namely, the limited frequency bandwidth due to the finite muon pulse width. Indeed, the delayed onset of the precession, defined by the onset of the pulsed magnetic field, will cancel the effect of the time spread of the muon pulse.

A slightly different experimental setup, of physical interest in the present work, consists in adding a constant field, parallel to the initial muon polarization. Depending on its intensity, this field can suitably lock the muon polarization while muons are incoming and before the transverse field is switched on. This configuration offers the unique opportunity to test the "sudden-versus-adiabatic" condition (in Ehrenfest's sense [8,9]) while observing, in real time, the evolution of the muon spins. The connection with the similar case in NMR [10,11] is straightforward. Put in simple terms, for suddenly changing fields muons will precess around the final field direction, whereas in the opposite adiabatic limit the muon spins will closely follow the field direction.

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We point out that μSR measures the muon spin precession by detecting the decay positron count rates given by

$$N(t) = N_0 \exp(-t/\tau_\mu)[1 + A_0 G(t) \cos(\omega t + \phi)],$$

where A_0 is the precession asymmetry amplitude (due to positron emission anisotropy), $G(t)$ a relaxation function, ω and ϕ, respectively, the angular frequency and the initial precession phase.

The experimental configuration adopted is shown in Fig. 1, where the applied magnetic field $\boldsymbol{B}$ is decomposed into a constant longitudinal magnetic field parallel to the initial muon spin $\boldsymbol{s_0}$ (along $\hat{\boldsymbol{x}}$) and a transverse component, switched on at a controlled rate, i.e., $\boldsymbol{B} = \boldsymbol{B}_\parallel + \boldsymbol{B}_\perp(t) = B_\parallel \hat{\boldsymbol{x}} + B_\perp(t)\hat{\boldsymbol{z}}$.

While $\boldsymbol{B}_\perp(t)$ increases, the total magnetic field $\boldsymbol{B}$ rotates in the xz plane with angular velocity controlled by $\dot{B}_\perp(t)$. The asymmetry signal $AG(t)\cos(\omega t + \phi) = (N_1 - N_2)/(N_1 + N_2)$, obtained from two counters with opposite phases, is measured along the $\hat{\boldsymbol{y}}$ direction which singles out the precession of the muon spin from its rotation.

The μSR experiments were carried out on the MuSR instrument of the ISIS Pulsed Muon Facility at the Rutherford Appleton Laboratory, U.K., whose typical 80 ns FWHM muon pulses imply a 6 MHz frequency bandwidth. The MuSR spectrometer consists of a set of 32 independent positron detectors, symmetrically positioned with respect to the sample and each covering the same solid angle.

The sample consisted of a 40 mm diameter disk of fused quartz, thick enough to stop the 26.5 MeV/c muon beam. Muons implanted in quartz form muonium atoms [12,13] and a small fraction remains as free, or diamagnetic, muons. The very different gyromagnetic ratios of these two species not only clearly distinguish the two signals but also offer the possibility of testing the adiabatic condition in two extreme limits.

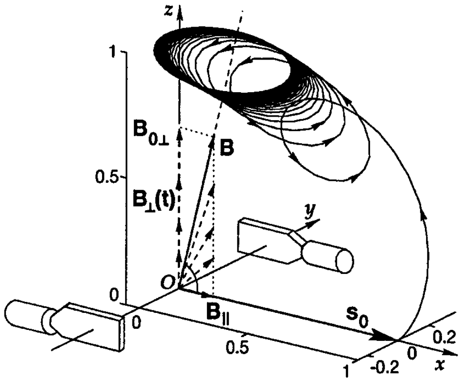

FIG. 1. Experimental setup for studying the adiabatic crossover of Mu. A 3D view of the Mu precession, simulated for a realistic case (relaxation omitted for clarity), is also shown. $\boldsymbol{B}_{0\perp}$ is the final value of $\boldsymbol{B}_\perp(t)$.

The chosen experimental geometry (cf. Fig. 1) enables us to measure the instantaneous precession cone angle $\phi(t)$ from the signal asymmetry $A(t)$ (respectively, equal to A_{Mu} and A_μ for Mu and μ^+), being $A = A_0 \sin\phi$. Since for an *ideal* sudden pulse, the corresponding maximum signal asymmetry would be $A_M = A_0 \sin\phi_{\mathrm{sp}} = A_0/\sqrt{1 + (B_\parallel/B_\perp)^2}$, for a *real* pulse, by defining the suddenness factor,

$$\mathcal{A} = \frac{\sin\phi}{\sin\phi_{\mathrm{sp}}}, \tag{1}$$

we can measure how sudden it is.

Longitudinal magnetic fields are generated by a pair of Helmholtz coils, whereas for a transverse pulsed magnetic field we adopted a solution described in a previous work [14]. Essentially, the pulsed field is produced by a current pulse fed into a thin metallic slab of thickness v and width w making a one-turn flat loop around the sample, giving thus a spatially uniform magnetic field equal to $B_\perp = \mu_0 I/(2vw)$ in the sample volume and no field outside it.

Synchronization of the magnetic field pulse with the muon pulse is obtained by triggering the current switching circuit with a suitably delayed reference signal from the ISIS accelerator.

The pulse profile, monitored by measuring the voltage drop across a series reference resistor in the current loop, has a double exponential shape, a best fit of which yields

$$B_\perp(t) = \frac{B_{0\perp}}{2}[c_1(1 - e^{-t/\tau_1}) + c_2(1 - e^{-t/\tau_2})], \tag{2}$$

where $B_{0\perp} = 1.88(1)$ mT, $c_1 = 0.86(5)$, $c_2 = 1.13(7)$, $\tau_1 = 42(2)$ ns, and $\tau_2 = 580(70)$ ns.

By using the classical law of time evolution for a spin $\boldsymbol{s}$ in a magnetic field $\boldsymbol{B}$: $d\boldsymbol{s}/dt = \boldsymbol{s} \times (\gamma \boldsymbol{B})$, as in the standard NMR treatment [10,11], we find that the evolution toward a stationary value of the angle $\phi(t)$ between the spin and the magnetic field is essentially controlled by the ratio $R(t) = \Omega_B/\gamma B$, with $\boldsymbol{\Omega_B}$ the angular velocity of the vector $\boldsymbol{B}$. Explicit substitution for Ω_B, gives

$$R(t) = \frac{\dot{B}_\perp(t) B_\parallel}{\gamma B^3}.$$

The exponential rise of $B_\perp(t)$ makes the maximum initial value $R_0 = R(0)$ control the spin behavior. In practice,

$$R_0 = \frac{\dot{B}_\perp(0)}{\gamma B_\parallel^2} \gg 1 \tag{3}$$

implies a large value for the asymptotic aperture angle ϕ_0, indicating a predominantly sudden regime behavior.

An easy experimental way of exploring the crossover from the sudden to the adiabatic regime is by varying the longitudinal field $B_\parallel$ with the largest $\dot{B}_\perp(t)$ achievable kept fixed.

Thus, the condition (3), easily satisfied for small $B_\parallel$ values, is violated for large values of $B_\parallel$ implying adiabatic behavior and a negligible aperture angle for the precession cone.

The presence of γ in the denominator makes the sudden regimes easier to achieve for μ^+ rather than for Mu, since $\gamma_{\mathrm{Mu}} \simeq 103\gamma_\mu$ and, consequently, the precession angle for μ^+ is expected to be quite close to the ideal angle ϕ_{sp}, which occurs for a completely nonadiabatic spin precession.

The first set of experiments was performed in zero longitudinal field and consisted of varying the *time delay* after which the transverse field was switched on with respect to the arrival of the muon pulse.

The case $B_\parallel = 0$, as seen by Eq. (3), is the only special case where the precession cone angle is maximum (being $\pi/2$ for both Mu and μ^+, thus implying a maximum asymmetry signal) and is independent of the rate $\dot{B}_\perp(t)$.

The results in two typical cases are shown in Fig. 2.

A pulsed field which reaches its steady value *before* the muon pulse arrives, as in Fig. 2(a), is equivalent to a constantly applied field and, since the muonium frequency in 1.88 mT (26.2 MHz) exceeds the passband limit, the Mu signal is lost due to dephasing. Unlike Mu, μ^+ precesses at a frequency well within the bandwidth and its signal is still observable. If, on the other hand, the pulse is started after the arrival of muons, no dephasing will occur and full asymmetry is measured also for muonium, regardless of the amplitude $B_\perp$. In particular, in Fig. 2(b) $B_\perp$ is delayed by $\sim$100 ns, enough to allow the entire muon pulse ($\sim$80 ns) to be implanted before any precession takes place.

The measurements of the adiabatic crossover for Mu and μ^+ were performed for longitudinal field values in the range $0.1 \lesssim B_\parallel \lesssim 2$ mT. Raw data, similar to those of Fig. 2(b), confirm that Mu and μ^+ signals lose their asymmetry amplitude as $B_\parallel$ is increased, and the adiabatic regime is reached, due to the consequent reduction of the precession cone aperture ϕ_0. Changes in A_{Mu} are far more dramatic than those in A_μ due to 2 orders of magnitude in their gyromagnetic ratios γ.

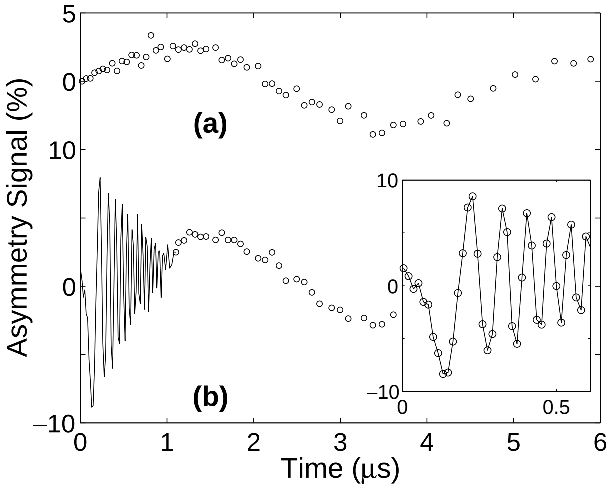

FIG. 2. Asymmetry signal for different delays of the pulsed transverse field. Magnetic field is switched on before (a), and after (b) the muon pulse arrival. The initial variable frequency signal due to the gradual increase of the transverse field [cf. Eq. (2)] is shown in the inset.

The asymmetry signal is shown in Fig. 3, where the raw data are well fitted by the Mu and μ^+ spin precessions in the presence of the time-dependent field of Eq. (2). The fit yields the initial value of the asymmetry signal.

After correction of the measured amplitudes by the factor $1/\sin\phi_{\mathrm{sp}} = \sqrt{1 + (B_\parallel/B_\perp)^2}$ and normalization to the $B_\parallel = 0$ value, one obtains the suddenness factor defined in Eq. (1) and the results are shown in Fig. 4.

For muonium, one can see that the sudden-to-adiabatic crossover occurs in the range $B_\parallel = 0.3$–0.6 mT while, as expected, for μ^+ the same transition takes place at much higher fields—by extrapolation—close to 10 mT. Note that the drop of the measured asymmetry is not a passband effect (there is no intrinsic frequency resolution limitation) but just the effect of the finite field switching velocity $\dot{B}_\perp$.

The numerical solution of the muon spin evolution yields the solid curves shown in Fig. 4 for the following values of the experimental parameters: $B_{0\perp} = 1.89(2)$ mT, $\tau_1 = 60(20)$ ns, $\tau_2 = 400(100)$ ns, $c_1 = 0.9(5)$, and $c_2 = 1.1(6)$. These values are in good agreement with the corresponding results of the direct measurement of the pulse shape as given below Eq. (2).

The same numerical procedure also provides an immediate visualization of the muon spin precession in muonium as shown in Fig. 1, for the particular case $B_\parallel = 0.4$ and $B_{0\perp} = 1.88$ mT. The μ^+ spin, initially dragged by the fast changing $\boldsymbol{B}$, enters an adiabatic regime and eventually stabilizes on the asymptotic precession cone as $\boldsymbol{B}$ approaches its final value.

Although the experimental data follow the theoretical curves rather well, some minor systematic discrepancies are caused by the muonium relaxation in quartz (Lorentzian with $\tau_L = 0.435\ \mu\mathrm{s}^{-1}$) and the beating

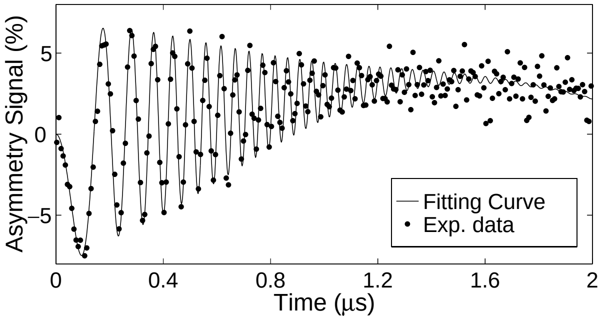

FIG. 3. Asymmetry signal for $B_\parallel = 0.2$ and $B_{0\perp} = 1.88$ mT; •: experimental data; —: fitting curve. The initial amplitude of the signal is the parameter of interest.

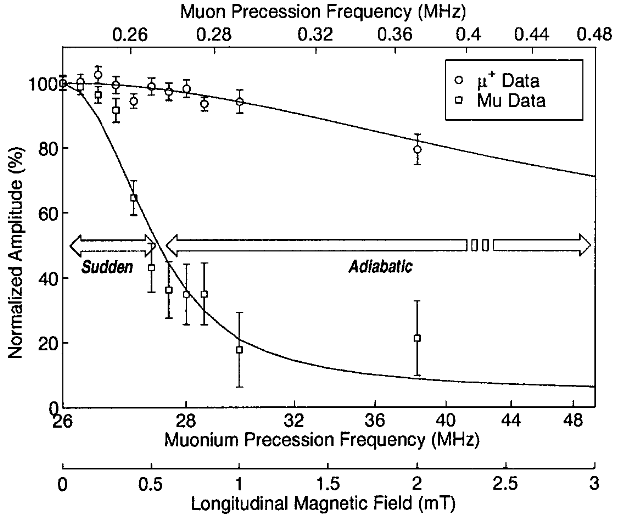

FIG. 4. Experimental results for the adiabatic crossover of Mu (□) and μ^+ (○) normalized and corrected for $1/\sin\phi_{\mathrm{sp}}$ (see text) as a function of the longitudinal magnetic field. Lines represent the results of numerical calculations. The frequency scales for Mu and μ^+ refer to the corresponding precession in the steady-state total field B.

between the two muonium triplet frequencies, which begin to split in the fields used in the present experiment.

For the envisaged applications of this pulsed procedure, the sudden regime is the most interesting since it allows switching to a precession mode with a large precession cone at the desired time. The presence of a static longitudinal field to preserve a large initial muon spin polarization can be very important typically in two cases: (a) when local random static fields present in the host material, such as nuclear dipole fields, would relax the spin polarization in the time lapse between the muon thermalization and the switching on of the transverse magnetic field; (b) when muonium radicals subject to transferred hyperfine interaction with host nuclei are formed, since the latter must be decoupled from the muon spin by a sufficiently large field to preserve the muon spin polarization.

For muonium, and probably for a variety of radical states, the sudden regime holds up to a few gauss, given the intensity and the rise time of the transverse field used in this experiment. However, the crossover field can be shifted to larger values of $B_\parallel$ by increasing $B_\perp$ and by reducing its rise time. From the present experiment, we know that $B_\perp$ as high as 10 mT are obtained without modifying the switching device. Further improvements of the device itself are envisaged which will allow even shorter rise times to be reached.

In summary, we have shown that in a pulsed muon beam characterized by periodic packets of muons having a time spread δt, the intrinsic frequency bandwidth can be extended by pulsing the external magnetic field *after* the muons have thermalized in the host material. For the ISIS time spread of ~80 ns the frequency bandwidth is ~6 MHz. In the present experiment the time resolution of the electronics (8 ns) limited the observable frequencies to ~40 MHz, which by no means is an intrinsic limit. Also we studied directly in the "laboratory frame of reference" the crossover between the sudden and adiabatic regimes by measuring the amplitude of the Larmor precession in the instantaneous magnetic field. The experiment performed on a quartz sample offers two independent spin probes: free muons and muons in muonium atoms, differing by a factor ~100 in their relative gyromagnetic ratios. We have shown how this factor effectively sets the scale for the transition between sudden and adiabatic regimes.

The technique which we have demonstrated, jointly to the existing pulsed rf resonance, opens new perspectives for a straightforward detection of the delayed paramagnetic species. Future developments of the present technique and of muon facilities with narrow muon pulses will allow new and more sophisticated experiments to be carried out, which will further extend the powerful NMR methods to the μSR field.

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