Muonium in Superfluid Helium

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We present the first muon-spin-relaxation studies in superfluid ^4He in the temperature range 0.5–1.5 K. At temperatures below 1 K we observed muonium spin precession. From the magnetic-field dependence of the amplitude of the muonium component we estimate a muonium formation rate of about $5 \times 10^7 \text{ s}^{-1}$ at 0.5 K. We find that external electric fields, preventing the convergence of muon and electron, reduce the amplitude of the muonium precession signal.

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The previous unsuccessful attempts to detect muonium atoms (μ^+e^-) in gaseous [1] and liquid [2] helium at 4.2 and 2 K were ascribed to the high ionization potential of the He atom ($E_i = 25 \text{ eV}$) compared with the binding energy of muonium ($E_b = 13.5 \text{ eV}$). In a magnetic field applied transversely to the initial μ^+ polarization the muon was found to precess with the Larmor frequency of a free muon and no sizable depolarization was observed [2]. In more recent studies a low and essentially temperature-independent depolarization rate for μ^+ in ^4He in the normal state was observed, whereas in the superfluid state the depolarization was found to increase rapidly with decreasing temperature below the λ point [3]. Additionally, below the λ point, a fast-relaxing component of the muon spin polarization was observed and it was suggested that the muon depolarization was caused by the interaction of the muon with an electron at a distance below 10^{-4} – 10^{-5} cm , produced during the muon thermalization, but no muonium signal was detected [4]. In this paper we report on the first direct muonium observation in superfluid helium. The amplitude of the muonium spin precession is found to be a strong function of applied electric and magnetic fields. The results can be understood in terms of delayed muonium formation arising from the combination of the μ^+ and an electron near the end of the slowing-down track.

Positive and negative ions have been widely used to probe the properties of superfluid helium and several investigations have been performed to study the state of these ions [5]. It is known that heavy positive ions in liquid helium attract the neighboring neutral atoms sufficiently to generate a high-density permanent cluster of He atoms around the ion (snowball) [6]. On the other hand, an electron in the liquid will become localized

within a bubble [7]. Because of the intermediate mass of the μ^+ (small in comparison to heavy ions, large in comparison to the electron), the state of the μ^+ -He complex in liquid He is *a priori* not well defined, but from a hydrodynamic point of view all charged-particle complexes are expected to have effective masses of several tens of He atoms and move according to the equations of viscous motion [6].

The mobility of charges in superfluid helium has been investigated extensively. Below the λ point it rises exponentially with decreasing temperature and at 0.6 K $b_+ = 1700 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and $b_- = 155 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for positive and negative charges, respectively [8,9]. The observation of long-lived muonium in superfluid He may therefore be ascribed to the huge mobilities in the superfluid state which allow the thermalized μ^+ and free electrons near the end of the μ^+ track to combine sufficiently fast to allow the evolution of phase-coherent muonium precession.

The experiments were performed at the surface muon beam channel πM3 of PSI utilizing the PSI μSR -facility spectrometer [10] in conjunction with a continuous-flow ^3He cryostat, designed and built at the Kurchatov Institute for Atomic Energy, which allowed us to use temperatures down to 0.5 K. The liquid ^4He , in which the μ^+ were stopped, was held in a 20-mm-diam, 9-mm-long copper cell, with an entrance window on one side made of 0.1-mm-thick Kapton foil. A 5–8- μm -thick copper foil in front of this window served as an electrode for the generation of an electric field in parallel to the μ^+ beam in the sample region. Transverse field muon-spin-relaxation (μSR) spectra were recorded using conventional time differential techniques. The muons were stopped quantitatively in the liquid He and no signal from μ^+ stopped

in the Cu cell could be seen.

Measurements in a field of 300 G and above 1 K reproduced previous results [3]; a two-component signal is observed which is well described by the following expression for the time evolution of the muon's polarization:

$$P(t) \propto [A_f \exp(-\lambda_f t) + A_s \exp(-\lambda_s t)] \cos(\omega_\mu t + \phi). \quad (1)$$

This expression consists of a fast-relaxing component with amplitude (equal to decay asymmetry) A_f and relaxation rate λ_f and a slowly relaxing component with A_s and λ_s . $\omega_\mu = \gamma_\mu H_{\text{ext}}$ is the μ^+ Larmor frequency in the external field H_{ext} . λ_f is about 2 to 3 orders of magnitude larger than λ_s ($\lambda_s < 0.3 \mu\text{s}^{-1}$). A significant fraction of the initial muon polarization is missing, as already observed in Ref. [4], which is strongly suggestive of slow muonium formation.

Indeed, by lowering the temperature below 1 K and using magnetic fields of less than 20 G, a long-lived muonium precession signal in the triplet spin state could be directly observed, whose gyromagnetic ratio is approximately 100 times that of a free muon and which approximately accounts for the missing polarization above 1 K. In order to describe the data Eq. (1) has to be complemented as follows:

FIG. 1. Time evolution of μ^+ polarization in superfluid He at 0.52 K in an external field of 8.8 G. The solid line corresponds to a three-component fit [Eq. (2)] to the data, the dotted lines showing the individual components. (a) Early part of the spectrum, which shows clearly the muonium precession signal and the fast relaxing "free" μ^+ signal. (b) Full spectrum which shows the fast and slowly relaxing "free" μ^+ signals. The signal in (a) close to time zero indicates the presence of some additional very fast-relaxing component out of which the muonium signal has probably evolved.

$$P(t) \propto [A_f \exp(-\lambda_f t) + A_s \exp(-\lambda_s t)] \times \cos(\omega_\mu t + \phi) + A_{\text{Mu}} \exp(-\lambda_{\text{Mu}} t) \cos(\omega_{\text{Mu}} t + \phi'). \quad (2)$$

$\omega_{\text{Mu}} \approx \frac{1}{2} m_\mu / m_e \gamma_\mu H_{\text{ext}}$ is the muonium precession frequency in the triplet state in the linear Zeeman regime, i.e., $H_{\text{ext}} \ll 1585$ G.

As an example Fig. 1 shows the μSR signal obtained at 0.52 K in a transverse field of ~ 0.8 G. One notices the precession pattern of muonium in the triplet state [Fig. 1(a)] and the fast and the slow polarization decay patterns of the diamagnetic (free) muon signal [Fig. 1(b)]. Not visible is the 50% fraction of muonium atoms in the singlet state which are associated with a frequency in the GHz range and which is not resolved by the spectrometer. λ_{Mu} is typically of the order of $0.1 \mu\text{s}^{-1}$ at 0.52 K.

The amplitude A_{Mu} of the muonium signal depends strongly on temperature, applied magnetic field, and also electrical field. A_{Mu} decreases with increasing temperature and is practically zero at 1.55 K. Figure 2 shows the field dependence of A_{Mu} at 0.52 K. It declines quickly with rising field and the muonium signal has disappeared for external fields $H_{\text{ext}} \gtrsim 20$ G. Application of an electrical field parallel to the μ^+ beam likewise suppresses A_{Mu} as can be seen from Fig. 3. All three observations together suggest that one observes slow muonium formation by combination of initially free thermalized μ^+ and electrons created during the slowing down of the μ^+ close to the end of the μ^+ track.

Assuming for simplicity that the combination can be

FIG. 2. Field dependence of the muonium amplitude A_{Mu} and the sum of the "free" μ^+ amplitudes A_s and A_f at 0.52 K. The solid line is a fit of A_{Mu} , defined in Eq. (4), to the data below 20 G. Note that $A_s + A_f$ appears to be field independent.

FIG. 3. Dependence of A_{Mu} and $A_s + A_f$ on an external electrical field at 0.52 K ($H_{\text{ext}} = 2.5$ G).

described by a first-order kinetic equation (this approximation implies a time and space constant electron density which is certainly not realized in practice), the rate of muonium formation can be expressed as

$$dn_{\text{Mu}}(t) = -dn_{\mu}(t) = \lambda n_{\mu}(t) dt, \quad (3)$$

i.e., $n_{\mu}(t) = n_0 e^{-\lambda t}$, where λ is the formation rate constant. The evolution of the μ^+ polarization, e.g., along the initial polarization, is then given by [11]

$$P(t) = \frac{1}{2} P_0 \frac{\lambda}{(\lambda^2 + \omega_{\text{Mu}}^2)^{1/2}} \{ -e^{-\lambda t} \cos \varphi + \cos(\omega_{\text{Mu}} t - \varphi) \} + P_0 e^{-\lambda t} \cos \omega_{\mu} t, \quad (4)$$

where $t\varphi = \omega_{\text{Mu}}/\lambda$ and P_0 denotes initial polarization. The first term refers to the muonium and the second term to the free muon signal. The factor $\frac{1}{2}$ accounts for the invisible spin-singlet muonium state. (Note that the polarized μ^+ combine with unpolarized electrons leading to an equal population of spin parallel and spin antiparallel configurations. Only the former one is an eigenstate of the hyperfine coupling Hamiltonian.) The field-dependent, generally reduced, amplitude A_{Mu} of the muonium signal is a result of *dephasing* as the muonium state is formed at *different times* and the phase coherence among the precessing muonium atoms is lost. The faster the muonium atoms precess, i.e., the larger H_{ext} , the faster the effect of dephasing. Note that $\omega_{\text{Mu}} \gg \omega_{\mu}$ and that therefore the free muon precession has no effect on the dephasing. Equation (4) has almost the same form used to describe the data in Fig. 1. What is missing is the weakly damped free muon precession signal appearing in

Eqs. (1) and (2). The presence of this component implies that a certain fraction of μ^+ are not subject to muonium formation at least not within the time window accessible to μSR .

Fitting the data in Fig. 2 by the formula for the amplitude of the muon signal in Eq. (4) one finds $\lambda = 4.5 \times 10^7 \text{ s}^{-1}$ at 0.52 K. The same value within $\pm 10\%$ is found from an analysis of the relative phase shift of the muonium signal with respect to the free μ^+ signal [φ in Eq. (4)]. The fit misses the faster drop of A_{Mu} above 15 G, indicating that Eq. (3) is perhaps an oversimplification. Nevertheless, λ will be a good measure of the formation rate. The formation rate should also be deducible from the disappearance of the free muon signal, i.e., from the second component in Eq. (4) or the fast decay signal in the data. Direct fits to the data at 0.52 K yield a field-independent average value of $\lambda_f \approx 1 \times 10^7 \text{ s}^{-1}$ which is significantly smaller than the value found from the field dependence of A_{Mu} . In fact, if the effective λ is as small as 10^7 s^{-1} the muonium signal should already be reduced by more than 75% at 5 G, in contrast to what is observed. On the other hand, the disappearance of the diamagnetic μ^+ signal with a rate of $4.5 \times 10^7 \text{ s}^{-1}$ or decay time of 22 ns may have been masked by the initial dead time of the spectrometer. A hint for it may be seen in Fig. 1 (see caption). These considerations suggest that there are in fact two channels of muonium formation with significantly different rates. Altogether then, there would be at least three different channels of μ^+ behavior in superfluid He. It may be speculated that the different channels reflect different molecular ionic states of μ^+ (e.g., $\mu^+ \text{He}_n$ with different n) in liquid He. Alternatively, the different channels may be explained in terms of the behavior of the track electrons in time and space. For example, if the electron density would drop to zero at a certain time after formation, the surviving free μ^+ would remain free and could account for the weakly damped diamagnetic μ^+ signal. In this respect it is interesting to note that $A_s + A_f$ in Fig. 3 increases almost by the same proportion as $2A_{\text{Mu}}$ (triplet + singlet $\mu^+ e^-$) decreases with increasing voltage, quite in contrast to what is seen in Fig. 2. The decrease of A_{Mu} with voltage is therefore certainly not the result of a decrease of the formation rate λ alone [see Eq. (4)] but at least in part of a genuine suppression of muonium formation and a consequently increased diamagnetic signal.

The effect of an external electrical field on the muonium formation process must be sought for in the competition of the Coulomb attraction between μ^+ and e^- and their interaction with the external field. However, a detailed understanding will rest on a better knowledge of the state(s) of the μ^+ molecular ionic complex, of the state(s) of the electrons created in the track, and of the involved kinetic behavior in superfluid He. The present results suggest that a study of the formation and motion of molecular ions in superfluid He by μSR spectroscopy is feasible and will constitute a challenge for future work.

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