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Negative muon spin rotation and relaxation on superconducting MgB_2

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Abstract. The internal nuclear magnetic field in a superconducting MgB_2 powder sample was studied with a μ^-SR technique. Although the past μ^+SR study on MgB_2 reported the appearance of a dynamic behavior even below T_c due to μ^+ diffusion, μ^-SR shows a static behavior in the whole temperature range measured, as expected. The ZF-μ^-SR spectra do not suggest any appearance of additional magnetic field below T_c within the experimental accuracy. Considering the small asymmetry of the μ^-SR signal, it is a challenge to detect the appearance of an internal magnetic field below T_c caused by the time reversal symmetry breaking.

1. Introduction

Although positive muon spin rotation and relaxation (μ^+SR) is widely used as a tool for studying a microscopic internal magnetic field in condensed matters over 40 years [1, 2], the counterpart technique, i.e., μ^-SR is less common for such purpose mainly due to a low counting rate for reaching reliable statistics. Since a majority of μ^- decay into electron within 2.2 μs or below depending on the element captured by, it is very difficult to observe a nuclear magnetic field with μ^-SR, which typically ranges below 10 Oe (corresponding to 0.135 MHz). Such μ^-SR measurements naturally require the data in the time domain up to $15 - 20$ μs, suggesting that a pulsed muon beam is more suitable than a continuous muon beam.

The problem for μ^-SR is the loss of asymmetry, which arises from the cascade of the μ^- from the outermost shell orbit to the inner orbits of a muonic atom. At least 5/6 of the μ^- spin polarization is lost during this process, whereas the μ^+ stops almost 100% spin polarized at the interstitial site in the lattice. This means that high statistics by about $(1 - 5/6)^{-2} = 36$ times is needed for for μ^-SR than for μ^+SR to obtain comparably reliable data.

Nevertheless, due to developments in the beam power [3] and counting system [4], a nuclear magnetic field was successfully observed in MgH_2 with μ^-SR [5]. In order to confirm the result and expand the μ^-SR work, we have attempted to measure the μ^-SR spectra for superconducting MgB_2 [6]. This is because Mg almost lacks nuclear magnetic moments (since the natural abundance of ^{25}Mg with $I = 5/2$ is 10% but those of ^{24}Mg and ^{26}Mg with $I = 0$ are 79 and 11%, respectively), and as a result, the majority of μ^- captured by Mg feel a nuclear magnetic field formed by surrounding B. Note that the natural abundance of ^{10}B with $I = 3$ is 19.9% and that of ^{11}B with $I = 3/2$ is 80.1%. Thus, the μ^- captured by B should form hyperfine coupling states, i.e., $F^{\pm} = I \pm 1/2$ states, leading to a slowly relaxing behavior [7].

The other scientific motivation for the μ^-SR experiment on MgB_2 is to determine more precise μSR parameters at low temperatures across the superconducting transition temperature ($T_c \sim 39$ K): i.e., the presence or absence of the additional fluctuation in the internal nuclear magnetic field. The past μ^+SR work on MgB_2 [8] showed the presence of dynamic behavior even above 12.5 K due to μ^+ diffusion. However, for the μ^-SR case, such effect should be negligibly small up to the vicinity of the decomposition temperature of MgB_2, because μ^- is captured by Mg. Therefore, we have attempted to extract the change in the internal magnetic field across T_c with μ^-SR.

2. Experimental

A powder sample of MgB_2 was purchased from Aldrich. The μ^-SR time spectra were measured on the decay muon beamline ARGUS at ISIS of Rutherford Appleton Laboratory in the United Kingdom. An approximately 34 g powder sample was placed in a copper container with $4 \times 5 \times 2$ cm^3 volume, made of 0.5 mm thick Cu plate. Two additional Cu plates, each 0.5 mm thick, were attached to the container as a degrader. The copper container was then set onto the bottom of the stick for the He-flow cryostat. The momentum of the μ^- beam was adjusted to 62 MeV/c to maximize the number of μ^- stopped in the sample. The experimental techniques are described in more detail elsewhere [1, 2]. The obtained μ^-SR spectra were analyzed using musrfit [9].

3. Results and Discussion

Figure 1 shows the TF-μ^-SR spectra with $H_{\rm TF} = 50$ and 114 Oe for MgB_2 recorded at 300 K. Because there are clearly two different frequency components in the spectra, the TF-μ^-SR spectrum was fitted with a combination of two exponentially relaxing cosine oscillations:

$$\begin{aligned} A_0 P_{\rm TF}(t) &= A_{\rm TF1}\cos(2\pi f_{\rm TF1} t + \phi_{\rm TF1})\exp(-\lambda_{\rm TF1} t) \\ &+ A_{\rm TF2}\cos(2\pi f_{\rm TF2} t + \phi_{\rm TF2})\exp(-\lambda_{\rm TF2} t), \end{aligned} \tag{1}$$

where A_0 denotes the initial asymmetry at $t = 0$, $P_{\rm TF}(t)$ denotes the μ^- spin depolarization function, $A_{\rm TF1}$ and $A_{\rm TF2}$ denote the asymmetries, $f_{\rm TF1}$ and $f_{\rm TF2}$ denote the μ^- spin precession frequencies, $\phi_{\rm TF1}$ and $\phi_{\rm TF2}$ denote the initial phases of the precession, and $\lambda_{\rm TF1}$ and $\lambda_{\rm TF2}$ denotes the exponential relaxation rates for the two signals.

Since we used a double pulsed μ^- beam for doubling the counting rate, the $A_{\rm TF}$ varies with $H_{\rm TF}$ due to the overlap of the oscillatory signal with different initial phases. As a result, the presence of the two frequency components in the TF-μ^-SR spectrum in $H_{\rm TF} = 114$ Oe is more obvious than that in $H_{\rm TF} = 50$ Oe. The magnitude of $f_{\rm TF1}$ is well explained by the equation: $f = \gamma_\mu/2\pi \times H_{\rm TF}$, where γ_μ is the muon gyromagnetic ratio and $\gamma_\mu/2\pi = 13.554$ kHz/Oe. This indicates that the $A_{\rm TF1}$ signal comes from the μ^- captured by ^{24}Mg and ^{26}Mg. On the contrary, the magnitude of $f_{\rm TF2}$ is found to be about 17.5% of $f_{\rm TF1}$ based on the linear relationship between $f_{\rm TF1}$ and $f_{\rm TF2}$ [Fig. 1(b)]. This suggests that the $A_{\rm TF2}$ signal comes from the μ^- captured by ^{11}B in the F^+ state: i.e., the hyperfine coupling state where the muon spin is parallel to the nuclear spin of ^{11}B. This is because the frequency ratio between the F^+ state of ^{11}B and bare μ^- is theoretically predicted as 0.174405 [10, 11]. The signals corresponding to the F^- state of muonic ^{11}B and the $F^{\pm}$ states of ^{10}B are not observed perhaps due to their small asymmetries.

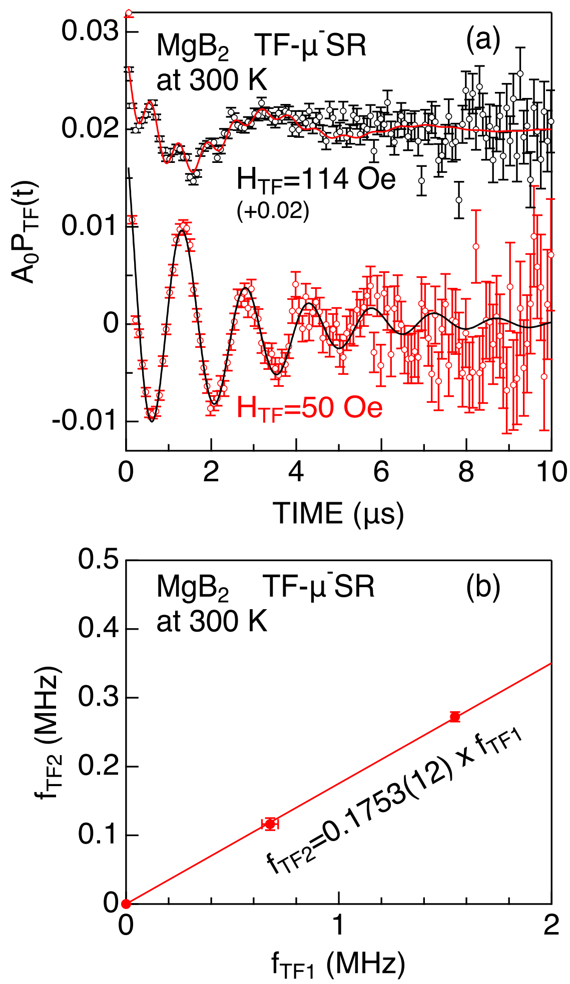

Figure 1. (a) The TF-μ^-SR spectra for MgB_2 recorded in $H_{TF} = 114$ Oe and 50 Oe at 300 K. (b) The relationship between two oscillation frequencies in the TF-μ^+SR spectrum. In (a), solid lines represent the best fit using Eq. (1). In (b), a linear line was obtained by a least square fit.

Figure 2(a) shows the temperature variation of the ZF-μ^-SR spectrum for MgB_2. The spectrum at 2 K looks essentially the same to those at 30 and 70 K, as expected for immobile muonic atoms. Although the μ^-SR spectrum is found to consist of the muonic Mg and the muonic ^{11}B signals (see Fig. 1), only the muonic Mg signal should be affected with a small LF. This is because the μ^- spin of the muonic ^{11}B *feels* the F^+ hyperfine coupling field, which is too strong to see the surrounding nuclear magnetic field. Therefore, we fitted the ZF-μ^-SR spectrum with a combination of an exponentially relaxing static Gaussian Kubo-Toyabe signal (G_{zz}^{KT}) for the muonic Mg and an exponentially relaxation signal for the muonic ^{11}B:

$$A_0 P_{ZF}(t) \quad = \quad A_{KT} G_{zz}^{KT}(t, \Delta) \exp(-\lambda t) + A_B \exp(-\lambda_B t), \tag{2}$$

where the $\exp(-\lambda t)$ term corresponds to any additional contribution to the μ^- spin depolarization of the muonic Mg: that is, the lifetime difference between muonic Mg and muonic ^{11}B [12, 13], the change in the background signal due to a long lifetime component created in

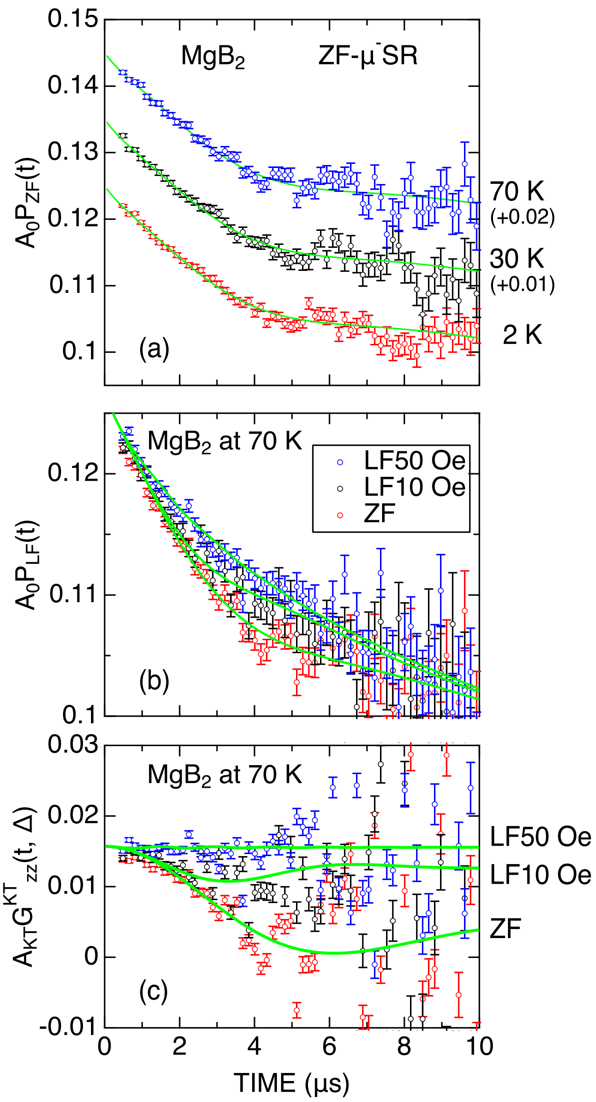

Figure 2. (a) The ZF-μ^-SR spectra for MgB_2 recorded at 0, 30, and 70 K, (b) the ZF- and LF-μ^-SR spectra for MgB_2 recorded at 70 K, and (c) the extracted static Gaussian Kubo-Toyabe component in the ZF- and LF-μ^-SR spectra. Solid lines represent the best fit using Eqs. (2) and (3). In (a), the spectrum at 30 K and 70 K were shifted upward by 0.01 for clarity of display.

the decay reaction of muonic atoms, and the possible appearance of an internal magnetic field in the superconducting state. Since only the muonic Mg provides information on the nuclear magnetic field and $A_{\rm TF1}$ corresponds to the asymmetry from the muonic Mg, we assume the following relationship:

$$\begin{aligned} A_{\rm KT} &= A_{\rm TF1}(H_{\rm TF} = 50 \text{ Oe at } 50 \text{ K}), \\ \Delta &= 0.317\ \mu\text{s}^{-1}, \end{aligned} \tag{3}$$

where Δ was predicted by a dipole field calculation with dipelec [14] at the Mg site in the MgB_2 lattice. In fact, not only the ZF-μ^-SR spectra but also the LF-μ^-SR spectra are reasonably fitted with Eqs. (2) and (3), as seen in Figs. 2(a) and 2(b). Moreover, the applied LF (= 50 Oe) is

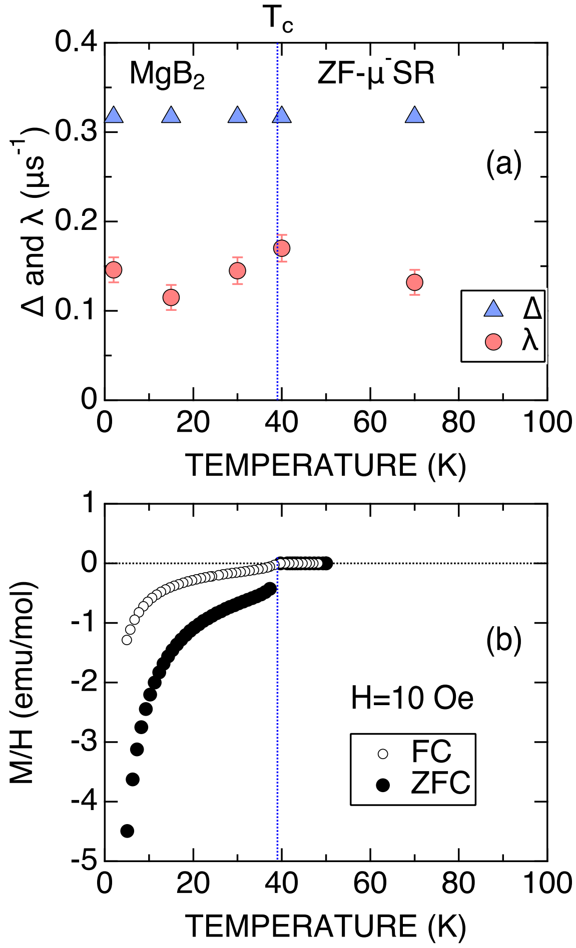

Figure 3. The temperature dependencies of (a) the field distribution width (Δ) and exponential relaxation rate (λ) and (b) magnetization for MgB_2. The data in (a) were obtained by fitting the ZF-μ^-SR spectra with Eqs. (2) and (3). Magnetization was measured both in a field cooling (FC) mode and a zero field cooling (ZFC) mode in $H = 10$ Oe.

fully decoupled a nuclear magnetic field [see Fig. 2(c)], although the data above around 5 μs are scattering due to the relatively low statistics (200 Mevents for each spectrum) for the small A_{KT}. Therefore, the ZF-μ^-SR spectra recorded at $T \leq 70$ K were fitted using common $A_{\mathrm{KT}} = A_{\mathrm{TF1}}$, Δ, A_{B}, and λ_{B}.

Figure 3 shows the temperature dependencies of λ and Δ together with the magnetization measured in $H = 10$ Oe with a SQUID magnetometer (mpms, Quantum Design). Despite a clear superconducting transition in the present MgB_2 sample, λ is almost temperature-independent in the temperature range between 2 and 70 K within the estimation accuracy. In fact, although λ looks to decrease with decreasing temperature below T_c, λ at 70 K is almost the same to λ at 2 K. This suggests the lack of a systematic change in λ below T_c. In overall, the μ^-SR experiment shows either the absence of an additional internal magnetic field below T_c or the presence of an internal magnetic field below T_c that is smaller than the experimental resolution.

4. Conclusion

Even in situations with diffusing μ^+, μ^-SR provides the information on an internal magnetic field from the fixed viewpoint. In fact, the nuclear magnetic field in MgB_2 was clearly detected in the whole temperatures below T_c. However, due to a small asymmetry of the μ^-SR signal, it was difficult to judge whether the additional internal magnetic field appears in MgB_2 below T_c.

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