

Muon spin relaxation study of the magnetic penetration depth in MgB₂

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(Received 27 August 2001; published 13 February 2002)

The magnetic vortex lattice (VL) of polycrystalline MgB₂ has been investigated by transverse-field muon spin relaxation (TF- μ SR). The evolution of the TF- μ SR depolarization rate σ , which is proportional to the second moment of the field distribution of the VL, has been studied as a function of temperature and applied magnetic field. The low-temperature value σ exhibits a pronounced peak near $H_{ext}=75$ mT. This behavior is characteristic of strong-pinning-induced distortions of the VL which put into question the interpretation of the low-field TF- μ SR data in terms of the magnetic penetration depth $\lambda(T)$. An approximately constant value of σ , such as expected for an ideal VL in the London limit, is observed at higher fields of $H_{ext}>0.4$ T. The TF- μ SR data at $H_{ext}=0.6$ T are analyzed in terms of a two-gap model. We obtain values for the gap size of $\Delta_1=6.0(3)$ meV [$2\Delta_1/k_B T_c=3.6(2)$], $\Delta_2=2.6(2)$ meV [$2\Delta_2/k_B T_c=1.6(1)$], a comparable spectral weight of the two bands, and a zero-temperature value for the magnetic penetration depth of $\lambda_{ab}\approx 100$ nm. In addition, we performed μ SR measurements in zero external field. We obtain evidence that the muon site (at low temperature) is located on a ring surrounding the center of the boron hexagon. Muon diffusion sets in already at rather low temperature of $T>10$ K. The nuclear magnetic moments can account for the observed relaxation rate and no evidence for electronic magnetic moments has been obtained.

DOI: 10.1103/PhysRevB.65.094512

PACS number(s): 74.70.Ad, 76.75.+i, 74.60.-w

The recent discovery of superconductivity at 39 K in the binary compound MgB₂ (Ref. 1) has triggered an enormous scientific effort in order to understand the mechanism that leads to such a high critical temperature in a seemingly classical superconductor. Based on band structure calculations Kortus *et al.*² conclude that a sizable electron-phonon coupling in combination with the high phonon frequencies due to the light mass of boron can reproduce the high critical transition temperature. The observation of a large B isotope effect confirms the important role of the phonons for superconductivity in this compound.^{3,4}

Subsequently, a large body of experimental work focused on the study of the symmetry of the superconducting order parameter. Measurements of the ¹¹B nuclear spin lattice relaxation rate,⁵ inelastic neutron scattering measurements,⁶ specific heat,⁷ high-resolution photoemission,⁸ and scanning tunneling spectroscopy⁹ are strongly in favor of a conventional BCS *s*-wave pairing state with moderately strong electron-phonon coupling. Even if superconductivity is phonon mediated, an analysis beyond a simple isotropic model may be required. Two different order parameters arising from two different sheets of the Fermi surface were calculated by Liu *et al.*¹⁰ and these predictions are in good agreement with the analysis of specific heat¹¹ and Raman¹² data in terms of a two-gap model.

Recent muon-spin-rotation (μ SR) and low-field ac-susceptibility measurements have been interpreted in terms of a quadratic temperature dependence of the magnetic penetration depth. From their data, the authors concluded that MgB₂ is an unconventional superconductor with an energy gap that has nodes at certain points in *k* space.¹³ These first μ SR experiments have been performed in a comparably small external magnetic field of 45 mT. At such a low mag-

netic field the vortex lattice (VL) is rather soft and therefore can easily be disordered by pinning induced distortions. In such a case random distortions of the VL will lead to a significant increase of the second moment of the magnetic field distribution, such as probed by the transverse-field (TF) μ SR technique. As a result, the magnetic penetration depth cannot be reliably deduced from the TF- μ SR data, at least not in a straightforward manner.

Here we present a more extensive set of TF- μ SR data that spans a wider range of applied magnetic fields. We present evidence that pinning-induced distortions of the vortex lattice are important at low magnetic fields for $H_{ext}\leq 0.3$ T, whereas they become less important at higher fields. We analyze the temperature dependence of the TF- μ SR depolarization rate at 0.6 T in terms of the two-gap model. Using this model we obtain a good fit to our experimental data. The obtained parameters for the gap sizes, the relative density of states of the bands, and the absolute value of the magnetic penetration depth are in reasonable agreement with values that have been previously reported (see, for example, Table I in Ref. 14).

A polycrystalline MgB₂ sample has been prepared from a stoichiometric mixture of Mg (99.98%, Johnson Matthey) and natural boron powder (~ 60 mesh, 99.5%, Aldrich). The reaction was carried out at 850 °C for 2 days using sealed Ta capsules under Ar atmosphere that were in turn encased in evacuated silica ampoules. After grinding under argon atmosphere, the sample was pressed into a pellet of 10 mm diameter using a cold isostatic press under a pressure of 0.3 GPa. The pressed pellet was annealed at the same temperature for 1 day. The x-ray powder diffraction pattern is characteristic of MgB₂ with lattice parameters of $a=3.085\,29(8)$ Å and $c=3.523\,84(7)$ Å. No indication for additional impurity phases was found. dc-susceptibility measurements using a

commercial superconducting quantum interference device (SQUID) magnetometer reveal a sharp superconducting transition at $T_c = 38.2(1)$ K.

The μ SR experiments have been performed at the π M3 muon beam line at the Paul-Scherrer-Institut in Villigen, Switzerland. Disk-shaped pellets of MgB_2 (10 mm in diameter and about 2 mm thick) were cooled in an externally applied magnetic field H_{ext} to temperatures below T_c in order to introduce a homogeneous flux line lattice (FLL). 100% spin-polarized positive muons are then implanted into the bulk of the sample with the initial muon spin polarization transverse to the external field. The muons thermalize rapidly without any significant loss in polarization and come to rest about 100–200 μm below the surface typically at interstitial lattice sites. The implanted muons are randomly distributed throughout the field profile of the FLL since the magnetic penetration depth λ significantly exceeds the lattice constants. Each muon starts to precess in its local magnetic field $B_{loc}(r)$ with the Larmor frequency $\omega_\mu = \gamma_\mu B_{loc}(r)$ where $\gamma_\mu = 851.4$ MHz/T is the gyromagnetic ratio of the muon. The time evolution of the so-called muon-spin-polarization function $P_\mu(t)$ is measured by monitoring the decay positrons which are preferentially emitted along the muon spin direction at the instant of decay (half lifetime 2.2 μs). $P_\mu(t)$ is oscillatory in character with a rapidly decreasing amplitude. Under certain conditions (see discussion below), the depolarization of the muon spin precession provides a measure of the field distribution within the vortex state and hence of the magnetic penetration depth λ .

For polycrystalline samples the depolarization is approximately of Gaussian form $P_\mu(t) \propto \exp(-\sigma^2 t^2/2)$, where the depolarization rate σ is proportional to the second moment of the field distribution $\sigma \propto \langle \Delta B^2 \rangle^{1/2}$. For an isotropic type-II superconductor (SC) the second moment is directly related to the magnetic penetration depth λ :

$$\langle \Delta B^2 \rangle_{iso} = 0.0371 \lambda^{-4}$$

and thus

$$\sigma [\mu\text{s}^{-1}] = 7.904 \times 10^4 \times \lambda^{-2} [\text{nm}].$$

However, recent measurements of the upper critical field on single crystals,¹⁵ c -axis-oriented thin films,¹⁶ and aligned MgB_2 crystallites¹⁷ give evidence for a sizable anisotropy of the superconducting properties with anisotropy ratios ranging from about 1.6 to about 2.5. In this case both components of the magnetic penetration depth affect the value of the relaxation rate. The value of the anisotropy therefore needs to be known in order to deduce the components of the magnetic penetration depth. For the special case of a large anisotropy with $\gamma = \lambda_c / \lambda_{ab} > 5$ Barford and Gun¹⁸ have shown that the measured effective penetration depth λ_{eff} is independent of the anisotropy ratio since it is solely determined by the in plane penetration depth λ_{ab} :

$$\lambda_{eff} = f_{anisotropy} \lambda_{ab},$$

with

$$f_{anisotropy} = 1.23,$$

$$\sigma [\mu\text{s}^{-1}] = 7.086 \times 10^4 \times \lambda_{ab}^{-2} [\text{nm}].$$

For anisotropies between 1 and 5 the correction factor $f_{anisotropy}$ varies between 1 and 1.23. For $\gamma = 2.5$, $f_{anisotropy} = 1.19$, which is already close to the high-anisotropy limit.

It should be stressed that the above equations are valid only for the case of an ideal FLL and in the so-called London limit for $\kappa = \lambda / \xi \gg 1$. With $\xi_{ab} \approx 7$ nm (Ref. 17) and $\lambda_{ab} \approx 100$ nm the London limit is applicable for MgB_2 . Another point of concern that needs to be addressed is the question as to how much pinning effects introduce distortions of the VL. It has been shown theoretically and experimentally that random distortions of the VL can significantly enhance the μ SR depolarization rate and thus lead to a sizable underestimation of the value of the magnetic penetration depth.¹⁹ Furthermore, the influence of the pinning effects will be temperature dependent and therefore can easily account for an unconventional temperature dependence of the μ SR depolarization rate in the presence of a conventional SC order parameter. The influence of pinning on the μ SR depolarization rate can be tested via its dependence on the applied magnetic field. The London model predicts that the second moment of the magnetic field distribution of a perfect VL should be independent of H_{ext} . The distortions of the VL due to pinning will be largest for small external fields where the vortices are far apart and their mutual interaction is comparably weak. At higher fields the vortex-vortex interaction is enhanced and it is more likely to maintain the long-range order of the vortex lattice. A prominent example is the cuprate high- T_c superconductors, where one typically observes a peak in $\sigma(H)$ at low field that is followed by a plateau at high field where $\sigma(H_{ext})$ is almost constant (see, e.g., Ref. 20 for $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ and Ref. 21 for $\text{Tl}_2\text{Ba}_2\text{CuO}_{6+\delta}$).

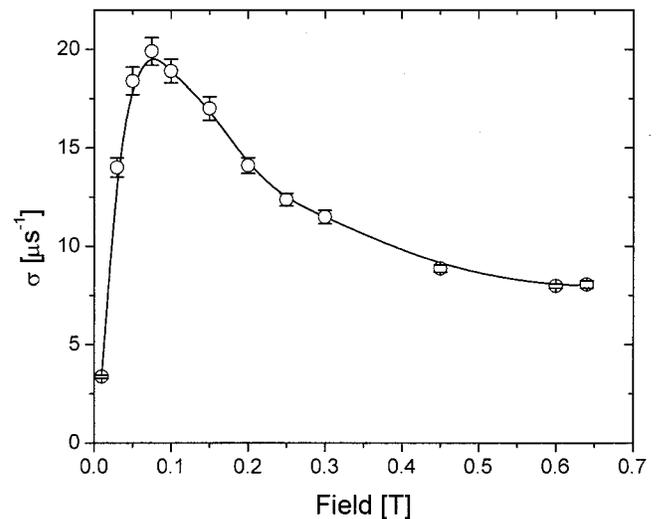


FIG. 1. Field dependence of the depolarization rate σ at $T = 5$ K. The observed behavior is characteristic of a pinning-induced disorder of the flux-line lattice.

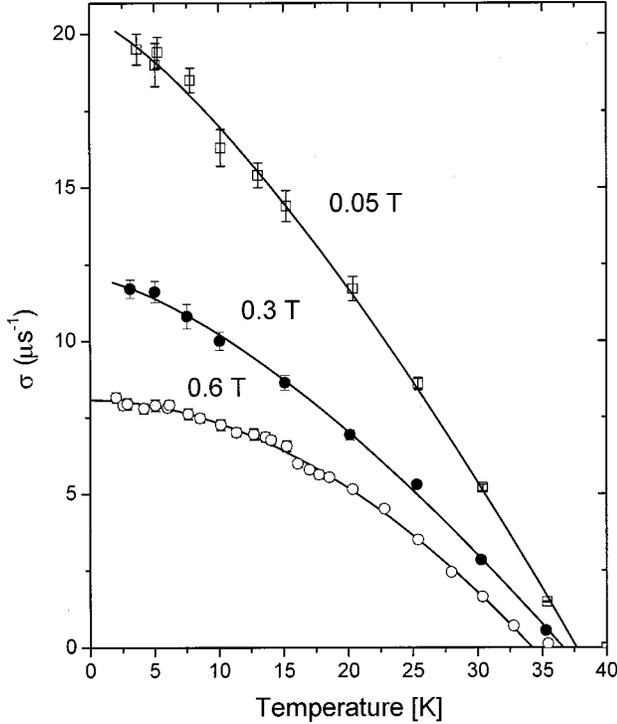


FIG. 2. Temperature dependence of σ for applied external fields of 50 mT (open squares), 0.3 T (solid circles), and 0.6 T (open circles). Solid lines are the best fits obtained with the function $\sigma(T) = \sigma(0)[1 - (T/T_c)^\alpha]$. The T dependence can be seen to change systematically as a function of applied field with the value of α changing from $\alpha = 1.33$ at 50 mT to $\alpha = 1.92$ at 0.6 T. In particular, this implies that the T dependence of $\sigma(T)$ at low T becomes weaker as the applied magnetic field increases.

Figure 1 shows the low-temperature μ SR depolarization rate $\sigma(H, T = 5 \text{ K})$ as a function of the applied magnetic field for $10 \text{ mT} < H_{ext} < 0.64 \text{ T}$ (the largest available field of the spectrometer at PSI). Each point has been obtained by field-cooling the sample from above T_c to 5 K. The value of $\sigma(H, 5 \text{ K})$ increases almost linearly below 50 mT; it goes through a pronounced maximum around 75 mT before it rapidly decreases again from $20 \mu\text{s}^{-1}$ at the peak position to about $8 \mu\text{s}^{-1}$ at 0.64 T. As outlined above, the observed peak in $\sigma(H)$ is characteristic of pinning-induced disorder of the FLL. This finding implies that the μ SR depolarization rate obtained at such a low magnetic field cannot be interpreted in terms of the magnetic penetration depth, at least not in a straightforward manner. Only at high magnetic fields for $H_{ext} \geq 0.4 \text{ T}$ is the value of $\sigma(H, 5 \text{ K})$ almost independent of H_{ext} such as is expected for an ideal FLL in the London limit. Figure 2 shows the evolution of the TF- μ SR depolarization rate as a function of temperature for different external fields of $H_{ext} = 0.05 \text{ T}$, 0.3 T, and 0.6 T. Shown by the solid lines are the best fits obtained with the function $\sigma(T) = \sigma(0)[1 - (T/T_c)^\alpha]$ which is based on the empirical two-fluid model. We ascribe no particular physical meaning to this function; we simply intended to parametrize our data in order to describe the changes in the T dependence of $\sigma(H_{ext}, T)$ with external magnetic field. The T dependence

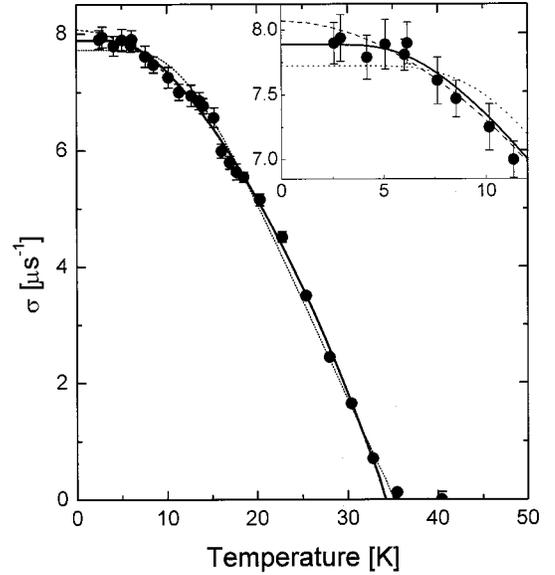


FIG. 3. Temperature dependence of σ for an applied external field of 0.6 T. The solid line shows the best fit using a two-gap model (Ref. 14). The dashed line is the result of a T^2 fit to the data. Also shown by the dotted line is a fit assuming an isotropic single gap. The inset shows the low-temperature region on a larger scale.

can be seen to change appreciably as a function of applied field with $\alpha = 1.33(8)$ at 50 mT, $\alpha = 1.44(7)$ at 0.3 T, and $\alpha = 1.92(5)$ at 0.6 T. The T dependence of $\sigma(T)$ at low T therefore weakens as the applied magnetic field is increased.

We have outlined above, that the TF- μ SR data obtained at $H_{ext} = 0.6 \text{ T}$ can be most reliably interpreted in terms of the magnetic penetration depth. Figure 3 displays the temperature-dependent TF- μ SR depolarization rate at $H_{ext} = 0.6 \text{ T}$. The inset shows a magnification of the behavior at low temperature. It occurs that $\sigma(T)$ is almost temperature independent below 5 K, while it exhibits a kink around 7 K where it suddenly begins to decrease with increasing temperature. Such a trend can be rather well reproduced with a two-gap model¹⁴ that assumes that the SC carriers reside in two different energy bands. The coupling between these bands needs to be sufficiently weak such that the magnitude of the energy gaps is different while they both appear simultaneously at the T_c that is determined by the larger gap. Evidence in favor of the two-gap model has been recently obtained by specific heat¹¹ and by Raman measurements.¹² The thick solid line shows the best fit to our experimental data using the two-gap model. The T dependence of the SC condensate density $n_s(T) \sim \sigma(T)$ is assumed to be

$$n_s(T) = n_s^0 - \gamma \delta n_s(\Delta_1, T) - (1 - \gamma) \delta n_s(\Delta_2, T),$$

$$\delta n_s(\Delta, T) = \frac{2n_s^0}{k_B T} \int_0^\infty f(\epsilon, T) [1 - f(\epsilon, T)] d\epsilon.$$

The parameter γ determines the ratio between the density of states of the band with the larger gap with respect to the one with the smaller gap, k_B is the Boltzmann constant, and

$f(\epsilon, T)$ is the Fermi distribution of quasiparticles with ϵ the energy of the normal electrons relative to the Fermi energy:

$$f(\epsilon, T) = (1 + e^{\sqrt{\epsilon^2 + \Delta^2(T)}/k_B T})^{-1}.$$

For $\Delta(T)$, we used the BCS values tabulated by Mühlischlegel.²² The obtained values of the fitting parameters are $\Delta_1 = 6.0(3)$ meV [$2\Delta_1/k_B T_c = 3.6(2)$], $\Delta_2 = 2.6(2)$ meV [$2\Delta_2/k_B T_c = 1.6(1)$], and $\gamma = 1.8(4)$ (the fit is not very sensitive to γ). These values are in reasonable agreement with previous results that have been obtained by applying the two-gap model to experimental data (see, e.g., Bouquet *et al.*¹⁴ and references therein). Taking the fitted low temperature value for $\sigma(T \rightarrow 0) = 7.9 \mu\text{s}^{-1}$ we derive $\lambda_{eff} = 100$ nm and, depending only weakly on the anisotropy, λ_{ab} values between 95 nm and 100 nm, in good agreement with the value of $\lambda_{ab} = 110$ nm, reported by Manzano and Carrington.²³ In the previous μSR experiment which was performed at a rather low external field of $H_{ext} = 450$ Oe a value of $\lambda_{ab} = 85$ nm was obtained which is also rather close to our value. Apparently, the value of $\sigma(5 \text{ K}) = 10 \mu\text{s}^{-1}$ obtained at $H_{ext} = 450$ Oe by Panagopoulos *et al.*¹³ is significantly lower than our value of $\sigma(5 \text{ K}) = 18 \mu\text{s}^{-1}$ at $H_{ext} = 500$ Oe. Such a difference could be explained if the pinning was much weaker in the commercial Alfa Aesar sample used by Panagopoulos *et al.* Another reason may be that the condensate density is significantly reduced in the commercial sample since it is known to contain a certain amount of magnetic impurities. Future experiments of the temperature and the field dependence of the TF- μSR depolarization rate on samples of different purity will be required in order to answer this question.

Let us return to the interpretation of the T dependence of the TF- μSR depolarization rate. Irrespective of the good agreement with the two-gap model, one should keep in mind that a reasonable fit to the experimental data can be obtained also assuming other kinds of scenarios, for example assuming that a single energy gap exists which is strongly anisotropic in k space. In that case the temperature dependence of the magnetic penetration depth would be strongly modified by the presence of impurities. For a conventional anisotropic s -wave gap the impurity scattering would tend to reduce the anisotropy since it mixes different states in k space. The clean-limit scenario of an anisotropic conventional s -wave gap has recently been discussed by Haas and Maki²⁴ who treated the case that the gap is significantly larger in the direction perpendicular to the boron planes than in the parallel one. It was shown earlier by Schneider and Singer that a sizable anisotropy of the SC energy gap with value of $\Delta_{min}/\Delta_{max}$ of the order of 0.5–0.6 will be required in order to account for a T^2 dependence of $\sigma(T)$.²⁵ In the case of an unconventional order parameter that changes sign in k space [for example, the $d_{x^2-y^2}$ -wave order parameter (OP) in high T_c cuprates], the potential scattering would tend to enlarge the nodal regions where the energy gap is zero.²⁶ As a result the absolute value of the magnetic penetration depth would be strongly reduced and the temperature dependence of λ would tend to change from a linear T dependence in the clean limit to a T^2 dependence in the dirty limit. Such a

scenario has been proposed in Ref. 13 in order to explain the T dependence of the TF- μSR depolarization rate. Shown by a dashed line is the result of a T^2 fit to our μSR data at 0.6 T. Evidently, this model also allows one to obtain a reasonable fit to the experimental data. For comparison the dotted line also shows the best fit assuming an isotropic single gap which does not describe our experimental data very well. The inset of Fig. 3 shows the most relevant low-temperature range on an enlarged scale. Such a closer inspection of the low- T data suggests that the two-gap model does provide the best fit to the experimental data.

Nevertheless, we would like to emphasize that based on experimental TF- μSR data one has to be very cautious with the attempt to discriminate between different scenarios concerning the nature and the symmetry of the superconducting energy gap. The experimental error of the data points, the problems related to pinning induced disorder of the flux-line lattice, possible structural changes of the vortex configuration as a function of temperature, and the unknown effect of vortex dynamics, especially at large external field and in the vicinity of the SC transition, make it virtually impossible to resolve the rather small differences in the T dependence of the magnetic penetration depth that are expected between the various scenarios. Further μSR experiments, preferably on single-crystalline materials, will be required in order to shed more light on the static and dynamic behavior of the VL in MgB_2 . We would like to stress that in our opinion only μSR experiments on single-crystalline materials of good quality will allow one to obtain reliable μSR results concerning the temperature dependence of $\lambda(T)$. This is in fact a lesson that was learned from studying the magnetic penetration depth of cuprate high- T_c superconductors. Experiments on polycrystalline materials as well as on the first available single crystals seemed to indicate that the magnetic penetration depth has a very weak low- T dependence that is indicative of an isotropic s -wave order parameter.^{20,27} Only after the microwave measurements of Hardy and co-workers²⁸ was it recognized from studies on good single-crystalline materials that the magnetic penetration depth rather follows a linear low- T dependence characteristic of an order parameter that has nodes in k space. Meanwhile it is well established that the order parameter has mainly $d_{x^2-y^2}$ symmetry. It is still not understood why the T dependence of the TF- μSR depolarization rate is so different for polycrystalline and single-crystalline materials.

Finally, in order to check our samples for the presence of magnetic impurity phases, we performed zero-field μSR measurements. Typical μSR time spectra are shown in Fig. 4. The static and randomly oriented nuclear magnetic moments of boron (and to a lesser extent magnesium) give rise to a Kubo-Toyabe-like relaxation, which is characterized by a Gaussian decay at early times followed by a recovery of the asymmetry to 1/3:

$$P_z(t) = \frac{1}{3} + \frac{2}{3} (1 - \Delta^2 t^2) \exp\left(-\frac{1}{2} \Delta^2 t^2\right),$$

where Δ is the rms width of the field distribution arising from the nuclear moments. Besides the signature of these

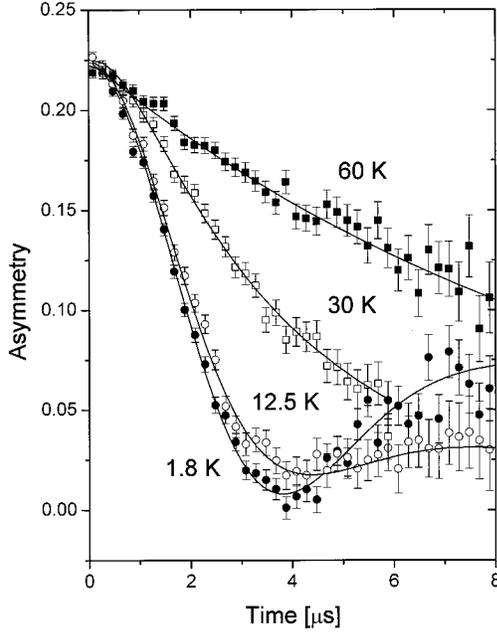


FIG. 4. Zero-field μ SR spectra obtained at 1.8 K, 12.5 K, 30 K, and 60 K. The Gaussian-like decay together with the almost complete recovery of the asymmetry to 1/3 observed at 1.8 K is characteristic for the interaction of the muon spin with randomly oriented static nuclear magnetic moments. The suppression of the 1/3 tail observed in the spectrum at 12.5 K indicates a low mobility of the muons with hopping rates of less than Δ^{-1} . In the limit of fast diffusion ($\Delta\tau_c \ll 1$) the time evolution of the muon polarization changes to an exponential shape such as for the spectrum at 60 K.

static nuclear magnetic moments, we obtained no evidence for any kind of additional magnetic moments (neither static nor slowly fluctuating within the characteristic time window of the μ SR experiment of $10^{-6} \text{ s} < \tau < 10^{-9} \text{ s}$). The spectrum obtained at 1.8 K shows a recovery of the polarization to 1/3 in the time range shown. The spectrum obtained at 12.5 K shows a significant suppression of the 1/3 tail, whereas the short-time behavior of $P_z(t)$ is not much changed from the static case. This behavior is characteristic for a slow motion of the muon with hop rates less than Δ^{-1} . Best-fit results were obtained by using a dynamic Kubo-Toyabe function of the form²⁹

$$P(t) = P_z(t) \exp(-\nu t) + \nu \int_0^t P_z(t') \exp(-\nu t') P_z(t-t') dt'$$

where $\nu = 1/\tau_{hop}$ is the jump rate (τ_{hop} is the time between jumps in the diffusion process, i.e., the residence time in

each potential well). A series of experiments done with a special setup which allows extension of the time range up to $20 \mu\text{s}$ shows that even at low temperatures the muon is not completely static, but shows very slow dynamics with hop rates of the order of $0.1 \mu\text{s}^{-1}$. At 60 K the time evolution of the muon polarization is well described by an exponential decay, indicating sufficiently fast motion of the muon ($\Delta\tau_c \ll 1$). Fast muon diffusion could in principal also lead to a motional narrowing of the field distribution arising from the flux-line lattice in the transverse field measurements. In the superconducting state we observe jump rates which increase from about $0.2 \mu\text{s}^{-1}$ at 12.5 K to about $3 \mu\text{s}^{-1}$ at 40 K just above T_c . Even at 40 K the diffusion path of the muons on a microsecond time scale will be shorter than about 1 nm. Such a distance is negligible as compared to the characteristic length scales of the vortex lattice, the magnetic penetration depth, and the vortex-vortex spacing that are of the order of 100 nm and 60 nm (at 0.6 T), respectively.

For at least a qualitative explanation of the muon dynamics one needs to know the muon stopping site within the MgB_2 lattice. Since Mg is almost completely ionized in the MgB_2 compound, the positive muon will be strongly repelled by the positively charged Mg ions. This should make the boron plane the most likely stopping site. In this plane the site in the center of the boron hexagon has the highest point symmetry D_{6h} (so-called b site). The calculated value of the second moment of the field distribution on the b site gives only $\Delta \sim 0.3 \mu\text{s}^{-1}$ and thus poorly agrees with the measured value of $\Delta = 0.45 \mu\text{s}^{-1}$. Much better agreement can be obtained for a muon site k , which is located about 0.5 \AA away from the center of the boron hexagon. Site b is surrounded by sextets of such k sites (along the center to boron line and, rotated by $\pi/6$, of sites m) which form a ring around the b site. Similar interstitial sites were identified as the muon sites in the hexagonal compounds UNi_2Al_3 (Ref. 30) and GdNi_5 (Ref. 31). There it was argued that the potential barrier between neighboring sites in the ring must be shallow, thus preventing localization of the μ^+ at individual m or k sites even at low temperatures. Hopping of μ^+ along a ring of interstitials as reported recently for GdNi_5 (Ref. 31) may explain our observed low-temperature behavior.

We would like to thank Alex Amato and Dierk Herlach (PSI) for the technical support during the μ SR experiments. This work was supported by the BMFT. C.N. gratefully acknowledges financial support by the Deutsche Forschungsgemeinschaft and helpful discussions with J.I. Budnick and G. Solt.

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