

Absence of a boron isotope effect in the magnetic penetration depth of MgB₂D. Di Castro,^{1,*} M. Angst,¹ D. G. Eshchenko,^{1,2} R. Khasanov,^{1,2} J. Roos,¹ I. M. Savić,³ A. Shengelaya,¹ S. L. Bud'ko,⁴ P. C. Canfield,⁴ K. Conder,⁵ J. Karpinski,⁶ S. M. Kazakov,⁶ R. A. Ribeiro,⁴ and H. Keller¹¹Physik-Institut der Universität Zürich, CH-8057 Zürich, Switzerland²Paul Scherrer Institute, CH-5232 Villigen PSI, Switzerland³Faculty of Physics, University of Belgrade, 11001 Belgrade, Yugoslavia⁴Ames Laboratory and Department of Physics and Astronomy, Iowa State University, Ames, Iowa 50011, USA⁵Laboratory for Neutron Scattering, ETH Zürich and PSI Villigen, CH-5232 Villigen PSI, Switzerland⁶Solid State Physics Laboratory, ETH, CH-8093 Zürich, Switzerland

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The magnetic penetration depth $\lambda(0)$ in polycrystalline MgB₂ for different boron isotopes (¹⁰B/¹¹B) was investigated by transverse field muon spin rotation. No boron isotope effect on the penetration depth $\lambda(0)$ was found within experimental error: $\Delta\lambda(0)/\lambda(0)=0.8(8)\%$, suggesting that MgB₂ is an adiabatic superconductor. This is in contrast to the substantial oxygen isotope effect on $\lambda(0)$ observed in cuprate high-temperature superconductors.

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Since the discovery of superconductivity with transition temperature $T_c \approx 39$ K in the binary intermetallic compound MgB₂,¹ a large number of experimental and theoretical investigations were performed in order to explain the mechanism and the origin of its remarkably high transition temperature. Experiments were done revealing the important role played by the lattice excitations in this material.²⁻⁵ In particular, the substitution of the ¹¹B with ¹⁰B has been demonstrated to shift T_c to higher temperatures,^{2,3} as expected for a phonon mediated pairing mechanism.

However, MgB₂ differs from conventional superconductors in several important aspects, including, for instance, the unusually high T_c and the anomalous specific heat.⁶ Calculations^{7,8} based on the Eliashberg formalism support the experimental results,^{6,9-11} revealing MgB₂ to be a two-band superconductor with two superconducting gaps of different size, the larger one originating from a 2D σ -band and the smaller one from a 3D π -band. The electronic σ -states are confined to the boron planes and couple strongly to the in-plane vibration of the boron atoms (E_{2g} phonon mode). This strong pairing, confined only to parts of the Fermi surface, is the principal contribution responsible for superconductivity and mainly determines T_c . The π -states on the remaining parts of the Fermi surface form much weaker pairs. The double-gap structure explains most of the unusual physical properties of MgB₂, such as the high critical temperature, the total T_c isotope-effect coefficient ($\alpha \approx 0.32^3$), the temperature dependent specific heat,⁶ tunneling,¹⁰ and upper critical field anisotropy H_{c2}^{ab}/H_{c2}^{lc} .¹²

An interesting point to be clarified concerns the nature of the electron-lattice coupling. It was proposed¹³⁻¹⁵ that MgB₂ is a nonadiabatic superconductor. Alexandrov¹³ suggested that, because of the large coupling strength of the electrons to the E_{2g} phonon mode, the many-electron system is unstable and breaks down into a small polaron system, similar to the cuprate high temperature superconductors (HTSC), where the charge carriers are trapped by local lattice distortions. Cappelluti *et al.*¹⁴ proposed that the small value of the

Fermi energy E_F of the σ bands relative to the phonon energy ω_{ph} violates the adiabatic assumption ($\omega_{ph} \ll E_F$), opening up a nonadiabatic channel that enhances T_c . Both these nonadiabatic models^{13,14} explicitly predict, but not quote, a boron isotope effect (BIE) on the carrier effective mass m^* in MgB₂. Zhao¹⁵ proposed an unconventional phonon mediated mechanism for superconductivity, predicting a boron isotope effect on the inverse squared magnetic penetration depth λ^{-2} of 4%. Similar models¹⁶⁻¹⁸ were already used to explain the large oxygen isotope effect (OIE) on the magnetic field penetration depth λ , a physical quantity directly related to the charge carrier effective mass m^* , observed in HTSC.¹⁹⁻²⁶ The nonadiabatic models are in contrast to the conventional theory of superconductivity (Migdal adiabatic approximation), in which the density of states at the Fermi level, the electron-phonon coupling constant, and the effective supercarrier mass m^* are all independent of the mass M of the lattice atoms.

Here, a muon spin rotation (μ SR) study of the magnetic penetration depth $\lambda(0)$ in polycrystalline MgB₂ for different boron isotopes (¹⁰B/¹¹B) is reported. μ SR is a powerful microscopic tool to measure the magnetic penetration depth λ .²⁷ Indeed, in a polycrystalline type II superconductor with a perfect vortex lattice (VL) the average magnetic penetration depth λ can be extracted from the muon-spin depolarization rate $\sigma(T) \propto \lambda^{-2}(T)$.²⁷ In our measurement, no BIE on $\lambda(0)$ was observed within experimental error [$\Delta\lambda(0)/\lambda(0) = 0.8(8)\%$], in contrast to the substantial OIE observed in cuprate HTSC.¹⁹⁻²⁶ Our results imply that polaronic or nonadiabatic effects in MgB₂ are absent or negligibly small.

To our knowledge, our experiments also provide for the first time a *direct* experimental evidence for the absence of an isotope effect on the penetration depth in a conventional superconductor. Only in a rather *indirect* way, it was shown²⁸ by critical field measurements, that in the conventional strong-coupling superconductor lead there is no isotope effect on the coefficient of the normal electronic specific heat. This confirms the validity of the adiabatic approximation in this system.

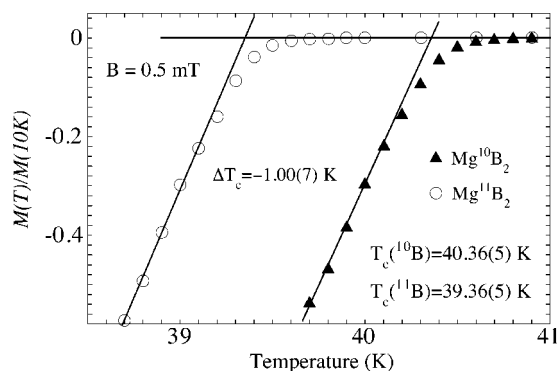


FIG. 1. Normalized field cooled (0.5 mT) magnetization as a function of temperature for Mg^{10}B_2 and Mg^{11}B_2 samples.

The μSR experiments were performed on two polycrystalline MgB_2 samples containing ^{11}B (Mg^{11}B_2) and ^{10}B (Mg^{10}B_2). Full details of the sample synthesis are given in Refs. 2 and 29. In brief, the two samples were synthesized using elemental Mg (99.9% pure in lump form) and isotopically pure boron (99.95% chemical purity, 99.5% isotope purity, <100 mesh) combined in a sealed Ta tube in a stoichiometric ratio. The Ta tube was then sealed in a quartz ampoule, placed in a 950°C box furnace for 24 h, and then removed and allowed to cool to room temperature.

To examine the quality of the samples low field (0.5 mT, field-cooled) magnetization measurements were performed using a commercial Superconducting Quantum Interference Device. Figure 1 shows the temperature dependence of the magnetization for the Mg^{11}B_2 and Mg^{10}B_2 samples in the vicinity of T_c . The high quality of the two samples is revealed by the sharp transition and the high T_c extracted from the intercept of the linear extrapolations (Fig. 1): $T_c(^{10}\text{B}) = 40.36(5)$ K, $T_c(^{11}\text{B}) = 39.36(5)$ K. There is a clear isotope shift of $\Delta T_c = T_c(^{11}\text{B}) - T_c(^{10}\text{B}) = -1.00(7)$ K. The corresponding isotope effect coefficient $\alpha_B = -d \ln(T_c) / d \ln(M_B) = 0.29(2)$ (enrichment corrected) is in good agreement with previous results.^{2,3}

The transverse-field μSR experiments were performed at the Paul Scherrer Institute (PSI), Switzerland, using the $\pi\text{M}3$ μSR facility. The samples used for the magnetization measurements (see Fig. 1) were pressed in disk-shaped pellets with 10 mm diameter and 3 mm thickness and cooled in an external magnetic field B_{ext} perpendicular to the muon spin polarization from well above T_c to temperatures lower than T_c . The measurements were taken in a field of $B_{\text{ext}} = 0.6$ T (the highest available at PSI), high enough to avoid pinning induced distortion of the VL.^{30–32} As shown in Fig. 2 for Mg^{11}B_2 at two different temperatures, the local magnetic field distribution can be very well approximated by a single Gaussian, centered at a field lower than the external one. This again indicates the high quality of the samples and the absence of any normal conducting domains. From the width of the Gaussian field distribution, which is proportional to the muon spin depolarization rate σ , the penetration depth λ , that is the length scale of the variation of the magnetic field, can be extracted using the relation $\lambda^{-2} \propto \sigma$.

In Fig. 3, the temperature dependence of σ for the Mg^{11}B_2 (○) and Mg^{10}B_2 (▲) samples is shown. Below T_c , σ

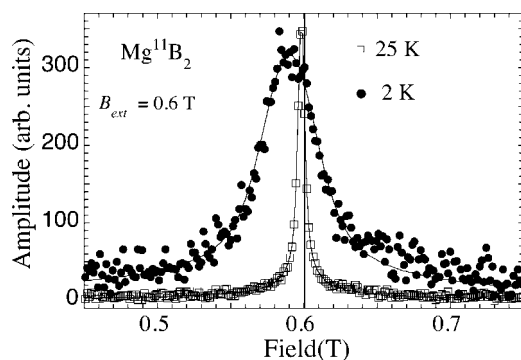


FIG. 2. Local magnetic field distribution, obtained from the Fourier transform of the muon spin precession signal, for Mg^{11}B_2 at 2 (●) and 25 K (□). Solid lines are Gaussian fits to the experimental data. The vertical solid line indicates the external field of 0.6 T.

for both samples starts to increase and saturates at low temperatures $T \leq 6$ K, in agreement with previous μSR measurements.³¹ The data for the two samples close to T_c show a clear isotope shift of $\Delta T_c = -1.2(2)$ K, in agreement with ΔT_c deduced from the low field magnetization measurements (Fig. 1). With decreasing temperature, the values of σ for Mg^{11}B_2 sample are systematically lower than those for the Mg^{10}B_2 sample. However, at low temperature they merge together, indicating that there is no substantial BIE on $\sigma(0)$.

In order to quantify this observation, we performed fits to the experimental data. It was suggested^{31,32} that for the two-gap superconductor MgB_2 , the temperature dependence of σ can be written in the form:

$$\sigma(T) = \sigma(0) - w \cdot \delta\sigma(\Delta_1, T) - (1 - w) \cdot \delta\sigma(\Delta_2, T) \quad (1)$$

with $\delta\sigma(\Delta, T) = [2\sigma(0)/k_B T] \int_0^\infty f(\varepsilon, T) \cdot [1 - f(\varepsilon, T)] d\varepsilon$.

Here, Δ_1 and Δ_2 are the zero temperature large and small gap, respectively, w is the relative contribution of the large gap to $\lambda^{-2}(0)$, and $f(\varepsilon, T)$ is the Fermi distribution. For the temperature dependence of the gaps we used the conven-

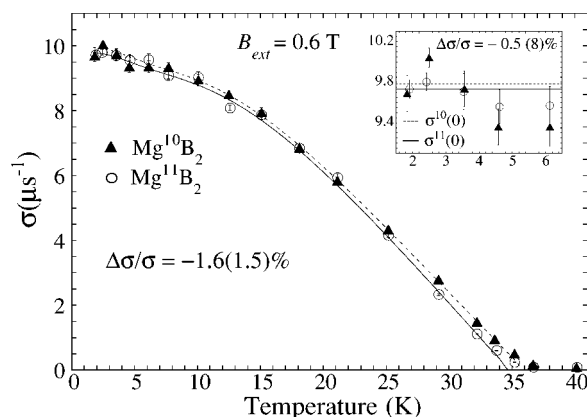


FIG. 3. Temperature dependence of σ at $B_{\text{ext}} = 0.6$ T for the two isotope samples Mg^{10}B_2 (▲) and Mg^{11}B_2 (○). The solid (Mg^{10}B_2) and dotted (Mg^{11}B_2) lines are fits using Eq. (1). Inset: low-temperature region on a larger scale. The dotted and solid horizontal lines represent the weighed average values of $\sigma(0)$ for $T < 7.5$ K for Mg^{10}B_2 and Mg^{11}B_2 , respectively.

tional BCS $\Delta(T)$. In order to improve the ratio of the number of data point vs the number of fit parameters, the two gaps and w were considered as common fitting parameters for the two isotope data. As shown by the solid and dotted lines in Fig. 3, the experimental data are well described by Eq. (1). The fit yields: $\sigma(0)^{11\text{B}}=9.79(10)\mu\text{s}^{-1}$, $\sigma(0)^{10\text{B}}=9.95(11)\mu\text{s}^{-1}$, $w=0.88(2)$, $\Delta_1=4.9(1)$, and $\Delta_2=1.1(3)$. All these values are in very good agreement with previous μSR measurements performed by us on a natural boron MgB_2 sample and by Ohishi *et al.*³² It is interesting to note that the high value of w implies that only a very small contribution to $\sigma(0)$ originates from the π -band, in accordance with the experimental finding that the superfluid density in the π -band is strongly suppressed by an external magnetic field.^{6,9,10,33–35} Below we discuss this issue in more details.

The relative isotope shift of $\sigma(0)$ is

$$\begin{aligned} (\sigma(0)^{11\text{B}} - \sigma(0)^{10\text{B}}) / \sigma(0)^{10\text{B}} &\equiv \Delta\sigma(0)/\sigma(0) = \Delta\lambda^{-2}(0)/\lambda^{-2}(0) \\ &= -1.6(1.5)\% , \end{aligned}$$

corresponding to:

$$\Delta\lambda(0)/\lambda(0) = 0.8(8)\% . \quad (2)$$

For comparison, we calculated the relative isotope shift using a different and model independent procedure, taking the weighed average of the experimental points for $T < 7.5$ K (see inset of Fig. 3), where $\sigma(T)$ saturates. We obtained $\Delta\sigma(0)/\sigma(0) = \Delta\lambda^{-2}(0)/\lambda^{-2}(0) = -0.5(8)\%$. Both the procedures give results compatible with zero BIE on the penetration depth $\lambda(0)$.

Here, it is very important to recall that the two isotope samples used in the experiment were made with the same starting Mg for both the samples, and with ^{10}B and ^{11}B powders of the same mesh size (distribution of grain sizes), and were synthesized under exactly the same conditions. Therefore, we can exclude any influence on σ due to different grain size and to a difference in pinning or vortex dynamical effects.

To check the reliability of our results, a second measurement on a set (set B) of samples from different source and preparation technique and with smaller Meissner fraction, was performed in a field of 0.4 T. The results are very similar to the first set (set A) shown above: $\Delta\lambda^{-2}(0)/\lambda^{-2}(0) = -1.5(1.7)\%$ as compared to the above $-1.6(1.5)\%$. This shows that our result is intrinsic for MgB_2 and holds for lower fields as well. A summary of the results for both sets of isotope samples is given in Table I. Note that the values of $\sigma(0)^{11\text{B}}$ and $\sigma(0)^{10\text{B}}$ for set B measured in lower fields are larger than the corresponding values for set A. This is not a consequence of flux lattice pinning or vicinity to the lower critical field H_{c1} (0.4 T and 0.6 T are fields well above H_{c1}), but is due to the field dependence of λ^{-2} caused by the suppression of the superfluid density in the π -band, as shown in several previous works.^{6,9,10,33–35} In particular, Cubitt *et al.*,³³ by means of small angle neutron scattering, and, more recently, Lyard *et al.*³⁴ and Angst *et al.*,³⁵ by means of magnetization measurement, show that the superfluid density λ^{-2} , rapidly decreases with increasing magnetic field from 0.1 T

TABLE I. Summary of the BIE results for $\sigma(0)$ obtained from the μSR measurements of two sets of isotope samples.

	$\sigma(0)^{10\text{B}}$ (μs^{-1})	$\sigma(0)^{11\text{B}}$ (μs^{-1})	$\Delta\lambda^{-2}(0)/\lambda^{-2}(0)$	
Set A	9.95(11)	9.79(10)	-0.016(15) ^a	-0.005(8) ^b
Set B	12.91(17)	12.69(13)	-0.015(17) ^a	-0.016(30) ^b

^aFrom fit using Eq. (1).

^bFrom low temperature average (inset Fig. 3).

to 1.2–1.5 T. This field dependence as a result of the two band superconductivity in MgB_2 may be likened to, yet is different from, the field dependence due to nonlocal effects as a consequence of the nodes in the gap in cuprate superconductors. In the latter case, the nonlocal effects cause a correction to the zero field penetration depth,³⁶ and therefore an isotope effect on the zero field penetration depth should be reflected in an isotope effect on the penetration depth in any field. The only exception would be a masking of a zero field penetration depth isotope effect by an opposite isotope effect on the correction terms, in a weird coincidence. The fact that we obtain no significant isotope effect in two different fields would seem to rule out such a scenario. In the case of MgB_2 , however, low field measurements simply probe the superfluid density in σ and π bands together, whereas high field measurements (as done here, particularly in the 0.6 T measurement) probe the superfluid density in the σ bands alone.

We note that we did not attempt to decompose the effective penetration depth λ into λ_{ab} and λ_c . It was shown³⁷ that in anisotropic polycrystalline samples with large anisotropy factor γ , λ is mainly determined by the in-plane penetration depth λ_{ab} : $\lambda = k\lambda_{ab}$, with k varying with γ . In MgB_2 , however, γ has a characteristic behavior as a function of temperature and magnetic field (see, for example, Refs. 12 and 33), due to the presence of two distinct bands, but not yet fully understood in a quantitative way. Particularly with a field dependent anisotropy, an accidental cancellation of non-zero isotope effects on λ_c and λ_{ab} both in 0.4 and 0.6 T can be considered highly unlikely. Therefore, in the rest of the paper we continue to use the effective λ .

It is interesting to contrast the result given in Eq. (2) with the oxygen isotope effect found in cuprate superconductors.^{19–26} At all doping levels a substantial oxygen isotope effect on λ_{ab} was observed, ranging from 2.8(1)%, close to optimal doping,²⁵ up to about 5(1)% in the underdoped regime.^{22,24} The BIE found here [0.8(8)%] is well below the values found in cuprates, and rather compatible with zero effect. Moreover this value is also considerably smaller than predicted in Ref. 15.

Theoretically, the zero temperature penetration depth is proportional to a density-of-states weighed average of a tensor involving the Fermi velocities. Detailed calculations within different formalisms have been carried out for MgB_2 (see Refs. 38 and 39). For our purpose it is sufficient to use the simpler London approach considering a free electron model and linking $\lambda(0)$ to the superconducting charge carrier density n_s and effective mass m^* , only considering different

contributions from the σ and the π bands. There is of course a direct connection between the Fermi velocities and the effective mass (a band average), both of which are not bare quantities, but in general renormalized, e.g., due to coupling with the phonons. The London approach has the advantage of facilitating the comparison with theoretical predictions^{13,14} and results obtained on cuprate superconductors,^{19–26} all of which are formulated within this approach.

Unlike the cuprate superconductors with their extremely short coherence lengths, MgB₂ cannot be considered as being in the superclean limit and we need to consider a possible impact of scattering. In a moderately clean superconductor the penetration depth is related to the effective mass m^* by the following relation:⁴⁰

$$1/\lambda^2 = [\mu_0 e^2 / c^2] (n_s / m^*) [1 / (1 + \xi / \ell)], \quad (3)$$

where n_s and m^* are the superconducting charge carrier density and effective mass, respectively, ξ is the coherence length, and ℓ is the mean free path. As already mentioned, the major contribution ($\sim 90\%$) to λ^{-2} in our experimental conditions comes from the σ -band. Therefore n_s , m^* , ξ , and ℓ in Eq. (3) have to be considered as σ -band values. It was estimated^{41,42} that in the σ -band $(\xi / \ell)_\sigma \approx 1/8$, a value which is close to the clean limit ($\xi / \ell \ll 1$). Therefore Eq. (3) may be approximated by $1/\lambda^2 \approx [\mu_0 e^2 / c^2] (n_s / m^*)$. A shift in $1/\lambda^2$ due to the isotope substitution is then given by

$$\frac{\Delta \lambda^{-2}(0)}{\lambda^{-2}(0)} = \frac{\Delta n_s}{n_s} - \frac{\Delta m^*}{m^*}. \quad (4)$$

The contribution from the supercarrier density n_s is negligible, as was already experimentally demonstrated in the case of HTSC.^{20–22} Specifically, for MgB₂, it can be argued that: (i) by changing the isotope only the mass of the nuclei is changed and not the charge carrier density n . Furthermore, MgB₂ is a stoichiometric compound; (ii) x-ray diffraction measurements, performed on the samples used for the μ SR experiments, showed no substantial difference between the lattice parameters of Mg¹¹B₂ and Mg¹⁰B₂. This implies that the band structure is not appreciably modified by the isotope

substitution. Therefore, assuming $\Delta n_s / n_s \approx 0$ in Eq. (4) and neglecting the small π -band contribution, we can estimate the boron isotope effect on the σ -band effective mass m_σ^* :

$$\Delta m_\sigma^* / m_\sigma^* \approx - \Delta \lambda^{-2}(0) / \lambda^{-2}(0) = 1.6(1.5) \% . \quad (5)$$

Here we have used the value of the relative shift on $\lambda^{-2}(0)$ obtained from the fit to Eq. (1). There is no BIE on the σ -band effective mass within experimental error.

Our result then suggests that nonadiabatic or polaronic effects in MgB₂ are absent or negligibly small, and establishes an upper limit [Eq. (2) and Eq. (5)] to any theoretical prediction of such effects.^{13–15} This conclusion is in contrast to cuprate superconductors, where a substantial oxygen isotope effect on m^* , well above the upper limit stated here, was observed.^{19–26} Recent magnetization measurements on MgB₂ under pressure⁴³ show no substantial pressure effect on the magnetic penetration depth λ at low temperature, further supporting the main conclusion of the present work.

In summary, μ SR experiments on polycrystalline Mg¹⁰B₂ and Mg¹¹B₂ samples revealed no substantial boron isotope effect on the magnetic penetration depth at $T=0$ K. From this finding we conclude that there is no substantial BIE on the effective mass m_σ^* of the charge carriers in the σ band. This result suggests that MgB₂ is a conventional phonon mediated superconductor without nonadiabatic or polaronic effects, in contrast to cuprate superconductors.

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