Muon-spin rotation measurements of the penetration depth in Li$_2$Pd$_3$B

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Measurements of the magnetic field penetration depth $\lambda$ in the ternary boride superconductor Li$_2$Pd$_3$B ($T_c \approx 7.3$ K) have been carried out by means of muon-spin rotation ($\mu$SR). The absolute values of $\lambda$, the Ginzburg-Landau parameter $\kappa$, and the first $H_{c1}$ and the second $H_{c2}$ critical fields at $T = 0$ obtained from $\mu$SR were found to be $\lambda(0) = 252(2)$ mm, $\kappa(0) = 27(1)$, $\mu_0 H_{c1}(0) = 9.5(1)$ mT, and $\mu_0 H_{c2}(0) = 3.66(8)$ T, respectively. The zero-temperature value of the superconducting gap $\Delta_0 = 1.31(3)$ meV was found, corresponding to the ratio $2\Delta_0/k_B T_c = 4.0(1)$. At low temperatures $\lambda(T)$ saturates and becomes constant below $T \approx 0.27 T_c$, in agreement with what is expected for s-wave BCS superconductors. Our results suggest that Li$_2$Pd$_3$B is a s-wave BCS superconductor with the only one isotropic energy gap.

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I. INTRODUCTION

The discovery of superconductivity in the ternary boride superconductors Li$_2$Pd$_3$B and Li$_2$Pt$_3$B has attracted considerable interest in the study of these materials. It is believed now that superconductivity in both above mentioned compounds is most likely mediated by phonons. It stems from photoemission, nuclear magnetic resonance (NMR), and specific heat experiments. Moreover, the observation of a Hebel-Slichter peak in the $^1$B spin-lattice relaxation rate in Li$_2$Pd$_3$B strongly supports singlet pairing. However, experimental results concerning the structure of the superconducting energy gap are still controversial. On the one hand, NMR data of Li$_2$Pd$_3$B (Ref. [4]) and specific heat data of Li$_2$Pd$_3$B and Li$_2$Pt$_3$B (Ref. [6]) can be well explained assuming conventional superconductivity. On the other hand, recent measurements of the magnetic field penetration depth $\lambda$ suggest unconventional behavior of both compounds, namely double-gap superconductivity in Li$_2$Pd$_3$B and nodes in the energy gap in Li$_2$Pt$_3$B. This contradiction is serious and shows that further experimental investigations of these compounds are needed.

In this paper, we report a systematic study of magnetic field penetration depth $\lambda$ in Li$_2$Pd$_3$B by means of transverse-field muon-spin rotation (TF-$\mu$SR) (the detailed description of TF-$\mu$SR technique in connection with $\lambda$ studies can be found, e.g., in Ref. [8]). Measurements were performed down to 30 mK in a series of fields ranging from 0.02 T to 2.3 T. For all magnetic fields studied (0.02 T, 0.1 T, 0.5 T, 1 T, and 2.3 T) no sign of a second superconducting gap was detected. All our results may be well explained by assuming conventional superconductivity with the only one isotropic energy gap. The absolute values of $\lambda$, the Ginzburg-Landau parameter $\kappa$, and the first ($H_{c1}$) and the second ($H_{c2}$) critical fields at $T = 0$ obtained from $\mu$SR were found to be $\lambda(0) = 252(2)$ mm, $\kappa(0) = 27(1)$, $\mu_0 H_{c1}(0) = 9.5(1)$ mT, and $\mu_0 H_{c2}(0) = 3.66(8)$ T, respectively. The zero-temperature value of the superconducting gap $\Delta_0$ was found to be 1.31(3) meV that corresponds to the ratio $2\Delta_0/k_B T_c = 4.0(1)$.

II. EXPERIMENTAL DETAILS

The Li$_2$Pd$_3$B polycrystalline sample was prepared by two-step arc-melting. First, a binary Pd$_3$B alloy was prepared by conventional arc-melting from the mixture of Pd(99.9%) and B(99.5%). The alloying of Li was done in the second arc-melting, in which a small piece of Pd$_3$B alloy was placed on a Li(> 99%) plate. Once the Pd$_3$B alloy melted, the reaction with Li occurred and developed very fast, forming a small button specimen (around 300 mg). Since the loss of Li was inevitable, the Li concentration in the final sample was estimated from the weight change. The deviation of the Li concentration from the stoichiometric one was less than 1% for the specimens used in this experiment.

Field-cooled magnetization ($M_{FC}$) measurements were performed with a SQUID magnetometer in fields ranging from 0.5 mT to 4 T at temperatures between 1.75 K and 10 K. The $M_{FC}(T)$ curve for $\mu_0 H = 0.5$ mT is shown in Fig. 1. The superconducting transition is rather broad indicating that the sample is not particularly uniform, i.e., the superconducting transition temperature may be evaluated only approximately. The midpoint of the transition corresponds to $T_c^{(mp)} = 7.1$ K, while the linear extrapolation of the steepest part of the $M(T)$ curve to $M = 0$ results in $T_c^{(ext)} = 7.6$ K (see Fig. 1).

TF-$\mu$SR experiments were performed at the $\pi$M3 beam line at Paul Scherrer Institute (Villigen, Switzerland).
The positron rate is given by the expression
\[ \dot{N}(t) = N_0 \left( \frac{1}{\tau_\mu} - \frac{1}{\tau_\mu} e^{-t/\tau_\mu} \right) + b g, \]
where \( N_0 \) is the normalization constant, \( b g \) denotes the time-independent background, \( \tau_\mu \) is the muon lifetime, \( a \) is the maximum decay asymmetry for the particular detector telescope (\( a \sim 0.18 \) in our case), and \( P(t) \) is the polarization of the muon ensemble:
\[ P(t) = \int P(B) \cos(\gamma_\mu B t + \phi) dB. \]

The \( P(t) \) and \( P(B) \) distributions inside the \( Li_2Pd_3B \) sample in the normal (\( T > T_c \)) and in the mixed state (\( T < T_c \)) after field cooling in a magnetic field of 0.1 T are shown in Fig. 2. The \( P(B) \) distributions were obtained from the measured \( P(t) \) by using the fast Fourier transform procedure based on the maximum entropy algorithm. In the normal state, a symmetric line at the position of the external magnetic field with a broadening arising from the nuclear magnetic moments is seen. Below \( T_c \) the field distribution is broadened and asymmetric. In order to account for the asymmetric field distribution, \( \mu SR \) time spectra obtained below \( T_c \) were fitted by two Gaussian lines.

\[ P(t) = \sum_{i=1}^{2} A_i \exp\left(-\sigma_i^2 t^2/2\right) \cos(\gamma_\mu B_i t + \phi), \]

where \( A_i, \sigma_i, \) and \( B_i \) are the asymmetry, the Gaussian relaxation rate, and the first moment of the \( i \)-th line, respectively. At \( T > T_c \), the analysis is simplified to a single line with \( \sigma_{nm} \sim 0.1 \) MHz arising from the nuclear moments of the sample. Eq. (3) is equivalent to the field distribution.

\[ P(B) = \gamma_\mu \sum_{i=1}^{2} \frac{A_i}{\sigma_i} \exp\left(-\frac{\gamma_\mu^2 (B - B_i)^2}{2\sigma_i^2}\right). \]
one (upper panel) and two Gaussian lines (lower panel) to the $\mu$SR time spectra. The corresponding $P(B)$ lines are shown in Figs. 2(c). For this distribution the mean field and the second moment are

$$\langle B \rangle = \sum_{i=1}^{2} \frac{A_i B_i}{A_1 + A_2}$$  \hspace{1cm} (5)

and

$$\langle \Delta B^2 \rangle = \frac{\sigma^2}{\gamma \mu} = \sum_{i=1}^{2} \frac{A_i}{A_1 + A_2} \left[ \frac{\sigma^2}{\gamma \mu} + |B_i - \langle B \rangle|^2 \right].$$  \hspace{1cm} (6)

The superconducting part of the square root of the second moment $\sigma_{sc}$ was then obtained by subtracting the contribution of nuclear moments $\sigma_{nm}$ measured at $T > T_c$ as $\sigma_{sc} = \sigma^2 - \sigma_{nm}$. From the known value of $\sigma_{sc}$ the absolute value of $\lambda$ can be evaluated using the following relation

$$\sigma_{sc}[\mu\text{s}^{-1}] = 4.83 \times 10^4 (1 - H/H_{c2}) \left[ 1 + 1.21 \left( 1 - \sqrt{H/H_{c2}} \right)^3 \right] \lambda^{-2}[\text{nm}],$$  \hspace{1cm} (7)

which describes the field variation of $\sigma_{sc}$ for an ideal triangular vortex lattice. \hspace{1cm} Note, that according to Ref. 10, Eq. (7) does not hold for very low magnetic inductions.

III. EXPERIMENTAL RESULTS AND DISCUSSION

A. Temperature dependence of the upper critical field

To summarize, the temperature dependence of the second critical field can be well described within the WHH theory. The absolute value of the second critical field at $T = 0$ was found to be $3.58(10)$ T in agreement with results of Badica et al. Ref. 2.

FIG. 4: (Color online) Temperature dependence of the upper critical field $H_{c2}$ of Li$_2$Pd$_3$B. The solid line is the fit using the WHH model. The fit parameters are listed in the text.

The field-cooled magnetization $M_{FC}(T)$ curves for several magnetic fields $H$ are shown in Fig. 3. The transition temperature $T_c$ was taken from the linearly extrapolated $M(T)$ curves in the vicinity of $T_c$ with $M = 0$

line. For each particular field $H$ the corresponding transition temperature $T_c(H)$ was taken as the temperature where $H = H_{c2}(T = T_c)$. The resulting $H_{c2}(T)$ dependence is presented in Fig. 4. It is seen that $H_{c2}(T)$ data can be satisfactory fitted with the model provided by the Werthamer-Helfand-Hohenberg (WHH) theory. The fit yields $H_{c2}(0) = 3.58(10)$ T and $T_c = 7.25$ K. Note, that the value of the transition temperature obtained from the fit lies between $T_{c}^{\text{imp}}$ and $T_{c}^{\text{ext}}$ introduced in Fig. 1 i.e., it is in agreement with low-field magnetization measurements. In the following, the $H_{c2}(T)$ curve presented in Fig. 4 is used to analyze the $\mu$SR data (see Sec. III C).

B. Magnetic field dependence of the second moment of $\mu$SR line

In Fig. 5(a) the temperature dependences of $\sigma_{sc}$ for $\mu_0 H = 0.02$ T, 0.1 T, 0.5 T, 1 T, and 2.3 T are shown. For $\mu_0 H = 0.5$, 1 T, and 2.3 T, $\sigma_{sc}(T)$ was measured down to 30 mK. It is seen, that below 1.5 K $\sigma_{sc}$ is practically temperature independent [Fig. 4(b)]. Bearing in mind, that the temperature dependence of $\sigma_{sc}$ saturates at low temperatures, the values of $\sigma_{sc}(T = 0)$ can be reliably evaluated for all magnetic fields. The results are plotted in Fig. 5 as a function of $H$. For $\mu_0 H = 0.5$ T, 1.0 T, and 2.3 T $\sigma_{sc}(0)$ was obtained from the zero slope linear fit of the $T \leq 1.5$ K data [see Fig. 5(b)] and for $\mu_0 H = 0.02$ T and 0.1 T $\sigma_{sc}(0)$ was assumed to be equal to $\sigma_{sc}$ at the lowest measured temperature ($T \approx 1.55$ K). The solid line in Fig. 5 represents the result of the fit of Eq. (7) to the experimental data with $H_{c2}(0) = 3.66(8)$ T and $\lambda(0) = 252(2)$ nm. As it was already mentioned above, Eq. (7) does not hold for the very low magnetic field. For
this reason the data point for \( \mu_0 H = 0.02 \) T was excluded from the fit. It is seen (Fig. 5) that the theoretical curve perfectly matches all data points for \( \mu_0 H \geq 0.1 \) T. The value of \( \sigma_{sc} \) for \( \mu_0 H = 0.02 \) T lies slightly below the theoretical curve, as expected. It is important to emphasize that the value of \( \mu_0 H_{c2}(0) = 3.66(8) \) T obtained from the fit of \( \sigma_{sc}(0, H) \) data coincides within the error with \( \mu_0 H_{c2}(0) = 3.58(10) \) T, evaluated in the previous section from the directly measured \( H_{c2}(T) \). This good agreement between the values of \( H_{c2} \), obtained from two completely different experiments, clearly demonstrates the validity of our analysis.

The above presented experiments clearly demonstrate, that \( \lambda \), evaluated from \( \mu \)SR measurements, is indeed magnetic field independent, as one would expect in case of a conventional superconductor with isotropic energy gap. On the other hand, in superconductors with nodes in the gap and isotropic double-gap superconductors like MgB\(_2\), \( \lambda \), evaluated in the same way, increases with increasing magnetic field (see e.g., Refs. 13, 14, and 15). Thus, the fact, that the \( \sigma_{sc}(0) \) versus \( H \) dependence is perfectly described by the field independent \( \lambda \) (see Fig. 2), implies that Li\(_2\)Pd\(_3\)B is a conventional single-gap superconductor.

The zero–temperature value of the superconducting coherence length \( \xi(0) \) may be estimated from \( H_{c2}(0) \) as \( \xi(0) = (\Phi_0/2\pi H_{c2}(0))^{0.5} \), which results in \( \xi(0) = 9.5(2) \) nm (\( \Phi_0 \) is the magnetic flux quantum). Using the values of \( \lambda(0) \) and \( \xi(0) \), one can also evaluate the zero-temperature value of the Ginzburg-Landau parameter \( \kappa(0) = \lambda(0)/\xi(0) \approx 27(1) \). This value of the first critical field can also be calculated by means of Eq. (4) from Ref. 11 as \( \mu_0 H_{c1} = 9.5(1) \) mT. It is remarkable that all superconducting characteristics of Li\(_2\)Pd\(_3\)B could be obtained solely from \( \mu \)SR experiments.

To summarize, the magnetic field dependence of the superconducting part of the \( \mu \)SR depolarization rate \( \sigma_{sc} \) is well described within the Ginzburg-Landau theory for anisotropic single-gap superconductors. The zero-temperature values of the first and the second critical fields, the magnetic penetration depth, the coherence length, and the Ginzburg-Landau parameter were found to be \( \mu_0 H_{c2}(0) = 3.66(8) \) T, \( \mu_0 H_{c1} = 9.5(1) \) mT, \( \lambda(0) = 252(2) \) nm, \( \xi(0) = 9.5(2) \) nm, and \( \kappa = 27(1) \), respectively.

C. Temperature dependence of \( \lambda \)

Eq. (4) implies that \( \sigma_{sc} \) depends on \( \lambda \) and the reduced magnetic field \( H/H_{c2} \). It is also clear that \( \sigma_{sc} \) vanishes for \( H \geq H_{c2}(T) \). This means, that in order to obtain the temperature dependence of \( \lambda \) from the \( \sigma_{sc}(T) \) curves, the temperature dependence of \( H/H_{c2} \) must be taken into account. In our calculations, the \( H_{c2}(T) \) curve provided by the solid line in Fig. 4 was used. Due to the known value of \( \kappa = 27(1) \), we are not limited by Eq. (4) but can directly apply the numerical calculations of Brandt. 16 In this case, the results collected in the lowest magnetic field (\( \mu_0 H = 0.02 \) T) can also be used. The resulting temperature dependence of \( \lambda \) is shown in the Fig. 4. Remarkably, all \( \lambda \) points, reconstructed from \( \sigma_{sc} \) measured in vari-
ous magnetic fields, collapse onto a single $\lambda(T)$ curve. One should emphasize that in this reconstruction no adjustable parameter was used. The $\sigma_{sc}(T)$ dependences were obtained by means of $\mu$SR, while the $H_{c2}(T)$ curve was measured in a completely different set of magnetization experiments (see Sec. III A).

FIG. 7: (Color online) $1/\lambda^2$ versus temperature. The solid line represents the theoretical curve provided by the BCS theory for $2\Delta_0 = 4k_BT_c$ with $T_c = T_{c_{ext}} = 7.6$ K (see Fig. 4). The inset shows $\lambda(T)$. The errors are not shown for clarity.

Considering the temperature dependence of $\lambda$, one should note that it can be better described by the BCS theory for a moderate coupling with the zero-temperature energy gap $2\Delta_0 = 4.0(1)k_BT_c$ [$\Delta(0) = 1.31(3)$ meV] rather than for the weak coupling limit. This is in agreement with the value $2\Delta_0 = 3.94k_BT_c$ obtained recently from specific-heat measurements. The upward deviations of the experimental data points from the theoretical curve at higher temperatures (see Fig. 7) is expected for nonuniform samples (e.g., for the samples where the transition temperatures are distributed via a certain range $\Delta T_c$). At temperatures close to $T_c$, $\lambda^{-2} \sim (T_c - T)$. Assuming that the critical temperature varies throughout the sample from $T_{c0} - \Delta T_c$ to $T_{c0}$, then the relative variation of $\lambda^{-2}$ for $T < T_{c0} - \Delta T_c$ is proportional to $[1 - \Delta T_c/(T_{c0} - T)]^{-1}$, i.e., it increases with increasing temperature. The value of $\sigma$, resulting from $\mu$SR experiments in the case of nonuniform samples, may be written as $\sigma = 1/V_0 \int \sigma_{local}^2 dV$ ($\sigma_{local}$ is the local value of $\sigma$ in the unit volume $dV$ and $V_0$ is the sample volume). Taking into account that $\sigma_{local}^2 \sim 1/\lambda_{local}^4$, this averaging implies that the parts of the sample with the smallest values of $\lambda$ (the highest values of $T_c$) provide the main contribution to $\sigma_{sc}$. At lower temperatures, however, $\lambda$ become more uniform throughout the sample and $\sigma_{sc}$ can be used for reliable calculations of $\lambda(T)$.

Recently, Yuan et al. studied the magnetic field penetration depth in Li$_2$Pd$_3$B by means of self-inductance technique. It was obtained that a considerable increase of $\lambda$ starts already at temperatures well below 1 K. Since such a behavior contradicts conventional BCS theory, the authors assumed the presence of the second superconducting gap. Our results (see Fig. 7) are quite different. $\lambda(T)$, evaluated from $\mu$SR measurements, is practically temperature independent below 2 K in complete agreement with conventional single-gap theories of superconductivity. We argue that the most probable reason for the above mentioned disagreement in the $\lambda(T)$ dependence comes from the difference in the experimental techniques. The self-inductance technique used in Refs. 3 and 4 provides information about the properties of the surface of the sample. It is likely that in this Li-containing compound the properties of the surface layer of the sample are different from those of the bulk.

To summarize, in the whole temperature range (from $T_c$ down to 30 mK) the temperature dependence of $\lambda$ is consistent with what is expected for a single-gap $s$-wave BCS superconductor. The value of the superconducting gap was found to be $\Delta_0 = 1.31(3)$ meV, that corresponds to the ratio $2\Delta_0/k_BT_c = 4.0(1)$.

IV. CONCLUSIONS

Muon-spin rotation and magnetization studies were performed on the ternary boride superconductor Li$_2$Pd$_3$B ($T_c \approx 7.3$ K). The main results are: (i) The absolute values of $\lambda$, $\xi$, $\kappa$, $H_{c1}$, and $H_{c2}$ at zero temperature obtained from $\mu$SR are: $\lambda(0) = 252(2)$ nm, $\xi(0) = 9.5$ nm, $\kappa(0) = 27(1)$, $\mu_0H_{c1}(0) = 9.5(1)$ mT, and $\mu_0H_{c2}(0) = 3.66(8)$ T. (ii) The values of $H_{c2}(0)$ evaluated from $\mu$SR and magnetization measurements coincide within the experimental accuracy. (iii) Over the whole temperature range (from $T_c$ down to 30 mK) the temperature dependence of $\lambda$ is consistent with what is expected for a single-gap $s$-wave BCS superconductor. (iv) No influence of the applied magnetic field to $\lambda(T)$ was observed. (v) At $T = 0$, the magnetic field dependence of $\sigma_{sc}$ is in agreement with what is expected for a superconductor with an isotropic energy gap.

To conclude, all the above mentioned features suggest that Li$_2$Pd$_3$B is a BCS superconductor with an isotropic energy gap.

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