Muon-spin rotation measurements of the penetration depth of the Mo3Sb7 superconductor

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Abstract: Measurements of the magnetic-field penetration depth lambda in superconductor Mo3Sb7 (Tc = 2.1 K) were carried out by means of muon-spin rotation. The absolute values of lambda, the Ginzburg-Landau parameter kappa, and the first Hc1 and the second Hc2 critical fields at T=0 are \lambda(0)=720(100) nm, \kappa(0)=55(10), \mu_0Hc1(0)=1.8(3) mT, and \mu_0Hc2(0)=1.9(2) T. The zero-temperature value of the superconducting energy gap \Delta(0) was found to be 0.35(1) meV, corresponding to the ratio 2\Delta(0)/kBTc=3.83(10). At low temperatures \lambda-2(T) saturates and becomes constant below T =0.3Tc, in agreement with what is expected for s-wave BCS superconductors. Our results suggest that Mo3Sb7 is a superconductor with the single isotropic energy gap.

DOI: https://doi.org/10.1103/PhysRevB.78.014502

Posted at the Zurich Open Repository and Archive, University of Zurich
ZORA URL: https://doi.org/10.5167/uzh-13707
Accepted Version

Originally published at:
DOI: https://doi.org/10.1103/PhysRevB.78.014502
Muon-spin rotation measurements of the penetration depth of the Mo$_{3}$Sb$_{7}$ superconductor


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Measurements of the magnetic field penetration depth $\lambda$ in superconductor Mo$_{3}$Sb$_{7}$ ($T_{c} \simeq 2.1$ K) were carried out by means of muon-spin-rotation. The absolute values of $\lambda$, the Ginzburg-Landau parameter $\kappa$, the first $H_{c1}$ and the second $H_{c2}$ critical fields at $T = 0$ are $\lambda(0) = 720(100)$ nm, $\kappa(0) = 55(9)$, $\mu_{0}H_{c1}(0) = 1.8(3)$ mT, and $\mu_{0}H_{c2}(0) = 1.9(2)$ T. The zero temperature value of the superconducting energy gap $\Delta(0)$ was found to be $0.35(1)$ meV corresponding to the ratio $2\Delta(0)/k_{B}T_{c} = 3.83(10)$. At low temperatures $\lambda^{-2}(T)$ saturates and becomes constant below $T \simeq 0.3T_{c}$, in agreement with what is expected for $s$–$wave$ BCS superconductors. Our results suggest that Mo$_{3}$Sb$_{7}$ is a BCS superconductor with the isotropic energy gap.

PACS numbers: 74.70.Ad, 74.25.Op, 74.25.Ha, 76.75.+i

Recently, the attention was devoted to Mo$_{3}$Sb$_{7}$. This compound was originally discovered more than forty years ago and only recently was found to become a type-II superconductor with the transition temperature $T_{c} \simeq 2.1$ K. The properties of Mo$_{3}$Sb$_{7}$ in a superconducting state are rather unusual. Specific heat, resistivity and magnetic susceptibility experiments of Candolfi et al. suggest that Mo$_{3}$Sb$_{7}$ can be classified as a coexistent superconductor – spin-fluctuating system. As discussed in Ref. 4, factoring in the effect of spin fluctuations leads to renormalized values of the electron-phonon coupling constant and the Coulomb pseudopotential, which, being substituted to the McMillan expression, lead to $T_{c}$ between 1.4 K and 2.0 K. This is substantially closer to the experimentally observed $T_{c} \simeq 2.1$ K than $T_{c} \approx 10$ K, which would be obtained without taking into account the effect of spin fluctuations.

There is currently no agreement on the symmetry of the order parameter of Mo$_{3}$Sb$_{7}$. The recent specific heat experiments of Candolfi et al. suggest that the order parameter in Mo$_{3}$Sb$_{7}$ is of conventional $s$–wave symmetry. Tran et al., based again on the results of specific heat measurements, have reported the presence of two isotropic $s$–wave like gaps with $2\Delta_{1}(0)/k_{B}T_{c} = 4.0$ and $2\Delta_{2}(0)/k_{B}T_{c} = 2.5$ [$\Delta(0)$ is the zero-temperature value of the superconducting energy gap]. In contrast, Andreev reflection measurements of Dmitriev et al. reveal that the superconducting gap is highly anisotropic. The maximum to the minimum gap ratio was estimated to be $\Delta_{max}/\Delta_{min} \simeq 40$ and $s + g$–wave symmetry of the order parameter was proposed in a qualitative analysis.

The symmetry of the superconducting order parameter can be probed by measurements of the magnetic penetration depth $\lambda$. A fully gapped, isotropic pairing state produces a thermally activated behavior leading to an almost constant value of the superfluid density $\rho_{s} \propto \lambda^{-2}$ for $T \lesssim 0.3T_{c}$. Presence of line nodes in the gap leads to a continuum of low lying excitations, which result in a linear $\lambda^{-2}(T)$ at low temperatures. In two-gap superconductors with highly different gap to $T_{c}$ ratios the inflection point in $\lambda^{-2}(T)$ is generally present.

In this paper, we report the study of the magnetic field penetration depth in superconductor Mo$_{3}$Sb$_{7}$ by means of muon-spin rotation. Measurements were performed down to 20 mK in a series of fields ranging from 0.02 T to 0.2 T. Our results are well explained assuming conventional superconductivity with the isotropic energy gap in agreement with the recent specific heat experiments of Candolfi et al. The zero-temperature value of the gap was found to be $\Delta(0) = 0.35(1)$ meV corresponding to the ratio $2\Delta(0)/k_{B}T_{c} = 3.83(10)$.

The Mo$_{3}$Sb$_{7}$ single-crystal samples were grown through peritectic reaction between Mo metal and liquid Sb. The transverse field muon-spin rotation (TF-µSR) experiments were performed at the πM3 beam line at Paul Scherrer Institute (Villigen, Switzerland). For our experiments the ensemble of some sub- and millimeter size single crystals were mounted onto the silver plate to cover the area of approximately 50 mm$^{2}$. The silver sample holder was used because it gives a nonrelaxing muon signal and, hence, only contributes as temperature independent constant background. The crystals were oriented so that the magnetic field was preferably applied along the 001 crystallographic direction. The Mo$_{3}$Sb$_{7}$ samples were field cooled from above $T_{c}$ down to $\simeq 20$ mK in magnetic fields ranging from 20 mT to 0.2 T. A full temperature scan (from 20 mK up to 2.5 K) was performed in a field of $\mu_{0}H = 0.02$ T.

Figure 1 shows the muon-spin precession signals in $\mu_{0}H = 0.02$ T above ($T = 2.2$ K) and below ($T = 0.05$ K) the superconducting transition temperature of Mo$_{3}$Sb$_{7}$. The difference in the relaxation rate above and below $T_{c}$ is due to the well-known fact that type-II superconductors exhibit a flux-line lattice leading to spatial inhomogeneity of the magnetic induction. As is shown by Brandt, the second moment of this inhomogeneous field distribution is related to the magnetic field penetration depth $\lambda$ in terms of $\langle \Delta B^{2} \rangle \propto \sigma_{sc}^{2} \propto \lambda^{-4}$. 

PREPRINT (May 24, 2009)
The analysis of TF-μSR data was carried out in the time-domain by using the following functional form:

\[
A(t) = A_0 \exp \left[ -\frac{\sigma_{nm}^2 + \sigma_{sc}^2}{2} t^2 \right] \cos(\gamma_\mu B_{int} t + \phi) + A_{bg} \cos(\gamma_\mu B_{ext} t + \phi),
\]

where the first term denotes the contribution from the Mo$_3$Sb$_7$ sample and the second term is the background contribution from the Ag sample holder. $A_0$ and $A_{bg}$ are the initial asymmetries arising from the sample and the background, $B_{int}$ and $B_{ext}$ are the internal field inside the sample and the applied external field seen in the Ag backing plate, $\gamma_\mu = 2.8 \times 135.5342$ MHz/T is the muon gyromagnetic ratio, and $\phi$ is the initial phase. $\sigma_{sc}$ and $\sigma_{nm}$ are the muon-spin relaxation rates caused by the nuclear moments and the additional component appearing below $T_c$ due to nonuniform field distribution in the superconductor in the mixed state.

The analysis was carried out as follows. First, the muon-time spectra were fitted by means of Eq. (1) with all the parameters free. Then, the ratio $A_0/A_{bg} = 0.975$ was obtained as the mean value in the temperature interval of 0.02 K ≤ $T$ ≤ 1.5 K. After that, by keeping the ratio $A_{bg}/A_0$ fixed, the nuclear moment contribution $\sigma_{nm}$ was estimated by analyzing the experimental data above 2.1 K, (i.e. at $T > T_c$, where $\sigma_{sc} = 0$). The final analysis was made with $A_0/A_{bg}$, $\sigma_{nm}$, and $B_{ext}$ as fixed, and $B_{int}$ and $\sigma_{sc}$ as free parameters. This allows to reduce the number of the fitting parameters and, as a consequence, to increase the accuracy in determination of $\sigma_{sc}$.

Within BCS theory the zero-temperature magnetic penetration depth [\(\lambda(0)\)] of isotropic superconductor does not depend on the magnetic field. Contrary, the nonlocal and the nonlinear response of the superconductor with nodes in the gap to the magnetic field, as well as, the faster suppression of the contribution of the smaller gap to the total superfluid density in a case of two-gap superconductor leads to the fact that $\lambda(0)$, evaluated from $\mu$SR experiments, is magnetic field dependent and it increases with increasing field.\(^\text{20,21}\) Such behavior was observed in various hole-doped cuprates,\(^\text{13,22,23}\) and the double-gap NbSe$_2$\(^\text{22,24}\) and MgB$_2$ superconductors.\(^\text{25,26}\)

Consideration of the ideal vortex lattice (VL) of an isotropic $s$-wave superconductor within Ginsburg-Landau approach leads to the following expression for the magnetic field dependence of the second moment of the magnetic field distribution\(^\text{18}\),

\[
\sigma_{sc}[\mu s^{-1}] = a \times (1 - B/B_{c2}) \left[ 1 + 1.21 \left( 1 - \sqrt{B/B_{c2}} \right)^3 \right] \lambda^{-2}[\text{nm}].
\]

Here $B$ is the magnetic induction, which for applied fields in the region $H_{c1} \ll H_{app} \ll H_{c2}$ is $B \simeq \mu_0 H_{app}$, $a$ is the coefficient depending on the symmetry of the VL ($a = 4.83 \times 10^4$ for triangular VL and, as is shown below, $a = 5.07 \times 10^4$ for rectangular VL), $H_{c1}$ is the first critical field, and $B_{c2} = \mu_0 H_{c2}$ is the upper critical field. Equation (2) accounts for reduction of $\sigma_{sc}$ due to stronger overlapping of vortices by their cores with increasing field. According to calculations of Brandt\(^\text{27}\), Eq. (2) describes with less than 5% error the field variation of $\sigma_{sc}$ for an ideal triangular vortex lattice and it holds for type-II superconductors with the value of the Ginzburg-Landau parameter $\kappa = \lambda/\xi \geq 5$ ($\xi$ is the coherence length) in the range of fields $0.25/\kappa^{1.3} \lesssim B/B_{c2} \leq 1$.

Satisfactory fit of Eq. (2) to the experimental data would suggest that there is no significant change of the penetration depth in the range of magnetic fields of
the experimental. We performed an analysis of the magnetic field dependence of the muon depolarization rate \( \sigma_{sc} \) measured at \( T = 20 \) mK (see Fig. 2), which is a good approximation of \( \sigma_{sc}(T = 0, B) \). The solid line in Fig. 2 corresponds to the fit of experimental \( \sigma_{sc}(B) \) by means of Eq. (2). The fit yields \( \lambda(0) = 720(25) \text{ nm} \) and \( B_{c2}(0) = 1.9(2) \text{ T} \). A good agreement between the experiment and the theory (see Fig. 2) allows to conclude that \( \sigma_{sc}(T = 0, B) \) can be consistently fitted within the behavior expected for conventional isotropic superconductor. For comparison in the inset of Fig. 2 we plot the dependence of the inverse squared penetration depth normalized to its value at \( B = 0 \) on the reduced field \( b = B/B_{c2} \) for \( \text{Mo}_3\text{Sb}_7 \) studied here, the two-gap superconductor \( \text{MgB}_2 \) from Ref. 24, and the hole-doped cuprate superconductor \( \text{YBa}_2\text{Cu}_3\text{O}_{6.95} \) from Ref. 22. It is seen that in a case of \( \text{MgB}_2 \) and \( \text{YBa}_2\text{Cu}_3\text{O}_{6.95} \), \( \lambda \) decreases rather strongly with increasing field.

FIG. 3: (Color online) (a) Theoretical \( P(B) \) profiles (\( \lambda = 720 \text{ nm}, \xi = 13 \text{ nm}, \text{and } \mu_0H = 0.02 \text{ T} \)) calculated for \( \sigma_g = 0.0006, 0.05, 0.1, \) and 0.15 \( \mu^2\text{m}^{-1}. \) (b) Dependence of the product \( \sigma_{sc} \times \lambda^{-2} \) on the reduced field \( b = B/B_{c2} \) for the triangular (circles) and the rectangular (stars) VL’s. Calculations were made by using the numerical approach of Brandt25 for \( \kappa = 100. \) The solid lines are obtained by means of Eq. (2) with \( a = 4.83 \cdot 10^4 \) for triangular and \( a = 5.07 \cdot 10^4 \) for rectangular VL’s, respectively. The insets are the contour plots of field variation within triangular and rectangular VL’s.

The absolute error in \( \lambda(0) \), estimated from the fit of \( \sigma_{sc}(B) \) by means of Eq. (2), is \( \approx 3.5\% \). There are, however, other sources of uncertainties which could also affect \( \lambda(0) \) and, consequently, increase the error: (i) The use of Eq. (2) to extract \( \lambda^{-2} \) from \( \sigma_{sc}(B) \) requires that the VL is well ordered and it is triangular, as opposed to disordered or rectangular VL. (ii) The fitting function Eq. (1) assumes that the contribution from the sample is described by a single line of Gaussian shape as opposed to asymmetric magnetic field distribution \( P(B) \) generally observed in good quality single crystals.26 (iii) The background signal due to the Ag backing plate was assumed to have a temperature-independent frequency and is non-relaxing at all \( T \). However, when the sample goes superconducting it excludes some field, even in the mixed state. These small fields are very inhomogeneous and could cause as a shift of the background signal to higher fields and an increase of a slow relaxation. In the following we are going to discuss in detail the sources (i) and (ii). We believe that the source (iii) does not play a substantial role here even since at an \( T = 20 \) mK \( B_{int} \) is only 0.16 mT smaller than the applied field and \( B_{ext} \) is constant in the whole temperature range (from 20 mK up to \( T_c \)). It should be mentioned, however, that experience with more robust superconductors has shown that even though the background field does not usually change perceptibly, the excluded flux can affect the background relaxation rate.

To account for possible random deviations of the flux core positions from their ideal ones (VL disorder) and for broadening of \( \mu \text{SR} \) spectra due to nuclear depolarization, the field distribution of an ideal VL \( P_{id}(B) \) is, generally, convoluted with a Gaussian distribution in terms of \( \sigma_{sc}, \sigma_{nm}, \) and \( \sigma_{B} \):

\[
P(B) = \frac{1}{\sqrt{2\pi}\sigma_g} \int \exp \left[-\frac{1}{2} \left( \frac{B-B'}{\sigma_g} \right)^2 \right] P_{id}(B') dB'.
\]

(3)

Here \( \sigma_g = \sqrt{\sigma_{sc}^2 + \sigma_{nm}^2 + \sigma_B^2} \) and \( \sigma_g \) is the contribution to the Gaussian broadening of \( P_{id}(B) \) due to VL disorder. The theoretical \( P(B) \) profiles for various \( \sigma_g's \) are shown in Fig. 3 (a). It is obvious that both, the nuclear moment contribution and the VL disorder broaden the \( P(B) \) profiles, thus requiring that the total second moment of \( \mu \text{SR} \) line needs to be the sum of three components:

\[
\sigma^2_{sc} + \sigma^2_{nm} + \sigma^2_{B}.
\]

This implies that neglecting the VL disorder leads to overestimation of \( \sigma_{sc} \) and, as a consequence, to underestimation of \( \lambda \). On the other hand, \( \sigma_{sc} \), obtained from the fit of asymmetric \( P(B) \) line by using symmetric Gaussian function [see Eq. (1)], becomes underestimated.22 In a case of \( \text{Mo}_3\text{Sb}_7 \) the main contribution to \( \sigma_g \) comes, most probably, from the nuclear moment term \( \sigma_{nm} \approx 0.178 \mu^2\text{m}^{-1} \), which is comparable with \( \sigma_{sc} \) in the whole field range, rather than from \( \sigma_B \), which for good quality single crystals corresponds, typically, to 10–20%, of \( \sigma_{sc} \) (see, e.g., Ref. 22 and references therein). Such big \( \sigma_{nm} \) also leads to the fact that the shape of \( P(B) \) profile is close to the Gaussian one as is demonstrated by the solid blue line in Fig. 3.

Dependences of \( \sigma_{sc} \) on the reduced field \( b = B/B_{c2} \) for triangular and rectangular VL’s, obtained by using numerical calculations of Brandt18 are shown in Fig. 6 (b). Solid lines correspond to Eq. (2) with \( a = 4.83 \cdot 10^4 \) (red line) and \( 5.07 \cdot 10^4 \) (blue line). As is seen, in a case of rectangular VL \( \sigma_{sc}(b) \) can still be satisfactorily described by means of Eq. (2) with the only 5% bigger value of the coefficient \( a \). This implies that the uncertainty related to different VL symmetries would result in \( \approx 2.5\% \) additional error in \( \lambda \). To summarize, by taking into account the above presented arguments we believe that the true overall uncertainty in the absolute value of \( \lambda(0) \) is about 10–15\%, and \( \lambda(0) = 720(100) \text{ nm} \).

The zero-temperature value of the superconducting coherence length \( \xi(0) \) may be estimated from \( B_{c2}(0) \) as \( \xi(0) = |\Phi_0/2\pi B_{c2}(0)|^{1/2} \), which results in \( \xi(0) = 13(1) \text{ nm} \) (\( \Phi_0 \) is the magnetic flux quantum). Using the
values of $\lambda(0)$ and $\xi(0)$, the zero-temperature value of the Ginzburg-Landau parameter is $\kappa(0) = \lambda(0)/\xi(0) = 55(10)$. The value of the first critical field can also be calculated by means of Eq. (4) from Ref. 18 as $\mu_0 H_{c1}(0) = 1.8(3)$ mT. Note that the zero-temperature values of $\lambda(0)$, $\xi(0)$, $\kappa(0)$, and the first and the second critical fields are in agreement with those reported in the literature.\textsuperscript{15,6,7,9}

Here $f = [1 + \exp(E/k_BT)]^{-1}$ is the Fermi function. The temperature dependence of the gap was approximated by $\Delta(T) = \Delta(0) \tanh[1.82[1.018(T_c/T - 1)]^{0.51}]$. As is seen, both, the dirty- and the clean-limit approaches, describe the experimental data reasonably well. The fitted curves are almost indistinguishable from each other. The results of the fits are $T_c = 2.11(2)$ K, $\Delta(0) = 0.35(1)$ meV and 2.12(2) K, 0.39(1) meV for the dirty- and the clean-limit calculations, respectively. The corresponding gap to $T_c$ ratios were found to be $[2\Delta(0)/k_BT_c]_{\text{dirty}} = 3.83(10)$ and $[2\Delta(0)/k_BT_c]_{\text{clean}} = 4.27(11)$. There are few reasons why, we believe, Mo$_3$Sb$_7$ studied here is in the dirty limit: (i) The maximum gap value obtained in Andreev reflection experiments was found to be $\Delta(0) \approx 0.32$ meV which is close to $\Delta(0) = 0.35(1)$ meV observed within our dirty limit calculations. (ii) The residual resistivity ratio measured on similar single crystals was found to be rather low $\rho_{300K}/\rho_{0K} = 1.4(2)$. Note that the isostructural compound Ru$_3$Sb$_7$ has $\rho_{300K}/\rho_{0K} = 144(1)$ which is bigger by more than 2 orders of magnitude. (iii) A reasonable assumption about Fermi velocity $v_F \approx 10^6$ m/s can be made within the free electron approximation by taking $E_F \approx 6.5$ eV.\textsuperscript{23} This allows us to readily estimate the BCS-Pippard coherence length $\xi_0 = h v_F/\pi \Delta(0) \approx 600$ nm which is approximately 50 times bigger than $\xi = 13(1)$ nm obtained experimentally. Considering that $\xi = \xi_0^{-1} + l^{-1}$ ($l$ is the mean free path) we obtain that the coherence length $\xi$ in Mo$_3$Sb$_7$ is limited by $l$ and, correspondingly, $l \approx \xi \approx 13$ nm.

Now we go to comment shortly the results of the specific heat experiments of Tran et al.\textsuperscript{24} It was shown that the analysis of the electronic specific heat data within the framework of the phenomenological alpha model suggests the presence of two gaps with $2\Delta_1(0)/k_BT_c = 4.0$ and $2\Delta_2(0)/k_BT_c = 2.5$ and the relative weight of the bigger gap of 0.7.\textsuperscript{24} The $\lambda^{-2}(T)$ curve simulated by using these parameters and $\Delta(0) = 716$ nm, is shown in Fig. 4 by blue dashed line. Both contributions were assumed to be in the dirty limit [see Eq. (4)] and the similar alpha model, but adapted for calculation of the superfluid density, was used.\textsuperscript{14,15} The agreement between the simulated curve and the experimental data is rather good. We should emphasize, however, that in two-gap superconductor the contribution of the smaller gap to the total superfluid density decreases very fast with increasing field, thus leading to strong suppression of $\lambda^{-2}$.\textsuperscript{15,25,26} This is inconsistent with the data presented in the inset of Fig. 2 revealing that in Mo$_3$Sb$_7$ $\lambda^{-2}(B) = \text{const.}$

To conclude, the superconductor Mo$_3$Sb$_7$ ($T_c \approx 2.1$ K) was studied by means of muon-spin rotation. The main results are: (i) The absolute values of the magnetic field penetration depth $\lambda$, the Ginzburg-Landau parameter $\kappa$, and the first $H_{c1}$ and the second $H_{c2}$ critical fields at $T = 0$ were found to be $\lambda(0) = 720(100)$ nm, $\kappa(0) = 55(8), \mu_0 H_{c1}(0) = 1.8(3)$ mT, and $\mu_0 H_{c2}(0) = 1.9(2)$ T. (ii) Over the whole temperature range (from $T_c$ down to 20 mK) the temperature dependence of $\lambda^{-2}$ is consistent with what is expected for a single-gap $s$-wave BCS superconductor. (iii) The ratio $2\Delta(0)/k_BT_c = 3.83(10)$ was

![Figure 4](image-url)
found, suggesting that Mo$_3$Sb$_7$ superconductor is in the intermediate-coupling regime. (iv) The magnetic penetration depth $\lambda$ is field independent, in agreement with what is expected for a superconductor with an isotropic energy gap. (v) The value of the electronic mean-free path was estimated to be $l \approx 13$ nm. This relatively short value suggests that strong scattering processes play an important role in the electronic properties of Mo$_3$Sb$_7$.

This work was performed at the Swiss Muon Source (S$
u$S), Paul Scherrer Institute (PSI, Switzerland). The authors are grateful to A. Amato and D. Herlach for providing the instrumental support during the $\mu$SR experiments, and A. Maisuradze for calculating $\sigma_{sc}(b)$ dependences. The work was supported by the K. Alex Müller Foundation and in part by the SCOPES grant No. IB7420-110784.

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