Comment on “Muon-spin-rotation study of the superconducting properties of Mo$_3$Sb$_7$"

Khasanov, R; Shengelaya, A; Savić, I; Baines, C; Keller, H

Abstract: In a recent article, Tran et al. [Phys. Rev. B 78, 172505 (2008)] reported results from muon-spin-rotation (SR) measurements of an Mo3Sb7 superconductor. Based on the analysis of the temperature and the magnetic field dependences of the Gaussian relaxation rate, $\gamma_{sp}$, it was suggested that Mo3Sb7 is a superconductor with two isotropic s-wavelike energy gaps. This relates to results found previously in the specific-heat measurements by several of the same authors in Acta Mater. 56, 5694 (2008). The purpose of this Comment is to point out that from the analysis made by Tran et al., the presence of two superconducting energy gaps in Mo3Sb7 cannot be justified. The analysis of SR data does not account for the reduction in $\gamma_{sp}$ with increasing temperature. The specific-heat data can be satisfactorily described within the framework of the one-gap model by assuming the presence of a small residual specific-heat component. The experimental data of Tran et al., as well as our earlier published SR results [Phys. Rev. B 78, 014502 (2008)], all seem to be consistent with the presence of single isotropic superconducting energy gap in Mo3Sb7. © 2010 The American Physical Society

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Comment on “Muon-spin rotation studies of the superconducting properties of Mo$_3$Sb$_7$”

R. Khasanov,$^1$ P.W. Klamut,$^2$ A. Shengelaya,$^3$ I.M. Savić,$^4$ C. Baines,$^1$ and H. Keller$^5$

$^1$Laboratory for Muon Spin Spectroscopy, Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland  
$^2$Institute of Low Temperature and Structure Research of the Polish Academy of Sciences, Okólna 2, 50-422 Wroclaw, Poland  
$^3$Physics Institute of Tbilisi State University, Chavchavadze 3, GE-0128 Tbilisi, Georgia  
$^4$Faculty of Physics, University of Belgrade, 11001 Belgrade, Serbia and Montenegro  
$^5$Physik-Institut der Universität Zürich, Winterthurerstrasse 190, CH-8057 Zürich, Switzerland

In a recent article Tran et al. [Phys. Rev. B 78, 014502 (2008)] report on the result of the muon-spin rotation ($\mu$SR) measurements of Mo$_3$Sb$_7$ superconductor. Based on the analysis of the temperature and the magnetic field dependence of the Gaussian relaxation rate $\sigma_{sc}$ they suggest that Mo$_3$Sb$_7$ is the superconductor with two isotropic $s-$wave like gaps. An additional confirmation was obtained from the specific heat data published earlier by partly the same group of authors in [Acta Mater. 56, 5694 (2008)]. The purpose of this Comment is to point out that from the analysis made by Tran et al. the presence of two superconducting energy gaps in Mo$_3$Sb$_7$ can not be justified. The analysis of $\mu$SR data does not account for the reduction of $\sigma_{sc}$ with increasing temperature, and, hence, yields inaccurate information on the magnetic penetration depth. The specific heat data can be satisfactory described within the framework of the one-gap model with the small residual specific heat component. The experimental data of Tran et al., as well as our earlier published $\mu$SR data [Phys. Rev. B 78, 014502 (2008)] all seem to be consistent with is the presence of single isotropic superconducting energy gap in Mo$_3$Sb$_7$.

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The magnetic field dependence of the $\mu$SR depolarization rate $\sigma_{sc}$. It is commonly accepted that the Gaussian muon-spin depolarization rate (square root of the second moment of the $\mu$SR line) of the superconductor in the vortex state ($\sigma_{sc}$) is directly related to the magnetic penetration depth $\lambda$ in terms of:

$$\sigma_{sc} = A(b) \cdot \lambda^{-2}.$$  \hspace{1cm} (1)

Here $A(b)$ is the proportionality coefficient ($b = B/B_{c2}$ is the reduced magnetic field, $B_{c2}$ is the upper critical field). One needs to stress, however, that the proportionality coefficient $A(b)$ is not constant. Its dependence on $b$ accounts for reduction of $\sigma_{sc}$ due to stronger overlapping of vortices with their cores with increasing magnetic field. As shown by Brandt$^2$ only for very low fields $(0.13/\kappa^2 \ll b \ll 1, \kappa = \lambda/\xi, \xi$ is the coherence length) one can neglect the dependence of $A(b)$ on the reduced magnetic field and assume it to be constant.

The condition $A(b) = const$ is definitively not satisfied in Ref.$^2$. The experiments were conducted for reduced fields in the range of 0.0025 $\leq b \leq 0.025$ at $T = 0.1$ K and 0.0036 $\leq b \leq 0.036$ at $T = 1$ K [between (0.1 K) $\approx 2$ T and $B_{c2}(1$ K) $\approx$ 1.4 T are taken from Ref.$^2$. As follows from Fig. 6 of Ref.$^2$ at these regions $A(b)$ for Mo$_3$Sb$_7$ superconductor $(\kappa > 50)$ is strongly field dependent. This implies, in turn, that in order to obtain $\lambda$ from $\sigma_{sc}(B)$ data, one needs to account for reduction of $A(b)$ with increasing magnetic field. It may be done, e.g., within the framework the London model (as is made by the authors, but without accounting for some limitations of the model, see the discussion below), or by using the approach developed by Brandt in Ref.$^2$.

Figure 1 shows the results of the fit of $\sigma_{sc}(B)$ data of Tran et al.$^2$ The Eq. (2) is derived within the framework of Ginzburg-Landau theory for the superconductor with single isotropic $s-$wave like gap$^2$. It 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 $\kappa \geq 5$ in the range of fields $0.25/\kappa^{1.3} \lesssim b \leq 1$.  

$$\sigma_{sc}[\mu s^{-1}] = 4.83 \cdot 10^4 \times \left(1 - b\right) \left[1 + 1.21 \left(1 - \sqrt{b}\right)^3\right] \lambda^{-2}[nm]$$ \hspace{1cm} (2)
An agreement of Eq. (2) with the experimental data of Tran et al. is relatively good (see Fig. 1) thus pointing to the field independent \( \lambda \) and, consequently, to the presence of only one superconducting energy gap in \( \text{Mo}_3\text{Sb}_7 \). We want also to note that in our recent paper\(^5\) which was published 3 month before the submission of Tran et al., \( \sigma_{\text{sc}} \) as a function of magnetic field for \( \text{Mo}_3\text{Sb}_7 \) superconductor was measured up to 4 times higher field \((\mu_0 H = 0.2 \text{ T}, b \simeq 0.1)\) and was found to be consistent with Eq. (2) and, consequently, with the field independent magnetic penetration \( \lambda \).

In reference to the interpretation of \( \mu \text{SR} \) data we note, that the authors of Ref. \(^4\) have mixed, somehow, the statements of field dependent \( \lambda \) and \( \sigma_{\text{sc}} \). The muon-spin depolarization rate of the superconductor in the vortex state \( \sigma_{\text{sc}} \) is always field dependent, while dependence of \( \lambda \) on the magnetic field is the characteristic of unconventional superconductors (like cuprates, pnictides, double gap MgB\(_2\) etc.). In a single gap \( s \)-wave superconductor the magnetic penetration depth is found to be independent on the magnetic field.\(^6\,9\,10\,11\) In the Ref. \(^3\), which is cited by Tran et al.\(^2\) in order to justify the unconventional two-gap superconductivity in \( \text{Mo}_3\text{Sb}_7 \), Sonier refers to the field dependent penetration depth \( \lambda \), but not the muon-spin depolarization rate \( \sigma_{\text{sc}} \).

**The modified London model.** The fit of \( \sigma_{\text{sc}} \) vs. \( B \) data by means of the modified London model, used by Tran et al., is in favor of the “one-gap” picture. Note that the London model is based initially on the statement of field independent \( \lambda \). By pointing to an agreement of this model with the experimental \( \sigma_{\text{sc}}(B) \) data, the authors of Ref. \(^1\) strongly contradict themselves, since the key argument of their paper is, in contrast, the field dependent \( \lambda \).

We want to stress however, that the London model uses some simplifications and assumptions and is strictly valid for the extreme type-II superconductor \((\lambda \gg \xi)\) for fields in the region \( 0 \ll B \ll B_c2 \). The possibility to use this model in order to describe the experimental \( \mu \text{SR} \) data needs to be justified for each particular case. The authors have not done that. On the other hand, the results of the numerical calculations of Brand\(^4\) which are valid for any type-II superconductors and in the full field region (from 0 up to \( B_{c2} \)), are free from these imperfections.

**Dependence of the magnetic penetration depth \( \lambda \) on temperature.** For the experiment conducted in constant magnetic field one needs, in addition, to account for dependence of the coefficient \( A(b) \), which relates the muon-spin depolarization rate \( \sigma_{\text{sc}} \) to the penetration depth \( \lambda \) [see Eq. (1)] on temperature. It is caused by the temperature dependence of the the second critical field \( B_{c2} \) and, as a consequence, that of \( b = B/B_{c2}(T) \). Obviously, this needs to be considered in order to reconstruct \( \lambda(T) \) from \( \sigma_{\text{sc}}(T) \) obtained experimentally. The detailed description of the reconstruction procedure (also in application to \( \text{Mo}_3\text{Sb}_7 \)) is given in Refs. \(^4\,11\) and \(^12\).

Figure 2 shows \( \lambda^{-2}(T) \) normalized on its value at \( T = 0 \) for \( \text{Mo}_3\text{Sb}_7 \) superconductor. The inset represents \( \lambda^{-2}(T) \) data. The solid black and the red circles refer to \( \lambda^{-2}(T) \) reconstructed from \( \sigma_{\text{sc}}(T) \) of Tran et al.\(^2\) and that reported in Ref. \(^1\), respectively. The inset of Fig. 2 implies that within the whole temperature region the difference between the absolute \( \lambda^{-2} \) values obtained in both sets of experiments does not exceed 10\% (5\% in \( \lambda \) value), which may be caused by the different sample shape (single crystalline samples in Ref. \(^4\) vs. fine powder in Ref. \(^1\), as well as the sample preparation procedures.

The lines in the main panel of Fig. 2 correspond to the fit of \( \lambda^{-2}(T) \) data from Ref. \(^1\) by assuming that \( \text{Mo}_3\text{Sb}_7 \) is a superconductor with the single \( s \)-wave like energy gap within the clean (solid line) and the dirty (dashed line) limit. It is obvious that both sets of the experimental data are in agreement with the each other as well as with the “single-gap” fitting curves from Ref. \(^4\).

**The absolute value of \( \lambda \).** The 1 nm error in the absolute \( \lambda \) value is unrealistic. The fit was performed within the framework of the certain (modified London) model, the validity of which, in application to \( \text{Mo}_3\text{Sb}_7 \) and the conditions of the experiment, was not justified. The fit of the \( \sigma_{\text{sc}}(B) \) data by using Eq. (2) [see Fig. 1] results in \( \lambda(0.1 \text{ K}) = 673(3) \text{ nm} \) which is 8 nm higher than \( \lambda(0.1 \text{ K}) = 665(1) \text{ nm} \) reported by Tran et al.\(^2\). In addition, the authors did not account for any other possible sources of uncertainties as, e.g.: i) vortex lattice disorder; ii) different possible symmetry of the vortex lattice (triangular vs. squared); iii) non-gaussian line shape of the \( \mu \text{SR} \) line which is expected to be seen even in a powder sample of the isotropic (weakly anisotropic) superconductor; iv) the background contribution from the Ag backing plate which may be influenced by the magnetic field expelled by \( \text{Mo}_3\text{Sb}_7 \) superconductor, etc. None of them
were discussed by Tran et al. in Ref.\textsuperscript{1} For these reasons and accounting for the uncertain assumption about temperature independent proportionality between $\lambda^{-2}$ and $\sigma_{sc}$ (see the discussion above), we call the penetration depth data presented by Tran et al. “inaccurate”.

**Temperature dependence of the electronic specific heat.**

One of the arguments pointing to the presence of two superconducting energy gaps in Mo$_3$Sb$_7$ was an agreement of the gap values obtained in Ref.\textsuperscript{1} with that deduced by Tran \textit{et al.}\textsuperscript{13} in specific heat experiments ($\simeq 13\%$ and $\simeq 5\%$ difference in the absolute values of the big and the small gap, respectively). Fig. 3 represents the specific heat data from Ref.\textsuperscript{13} together with the fits based on the “one-gap” BCS model. Note that, the simplest assumption about the presence of small temperature independent residual electronic specific heat, which may be easily caused by the presence of small inclusions of metallic Mo, leads to good agreement of the “one-gap” fit with the experimental data (see Fig. 3 note the logarithmic $C_{el}$ scale).

**Conclusions.** The fact that the "two-gap" fits performed by Tran \textit{et al.} in Refs.\textsuperscript{1} and \textsuperscript{13} lead to reasonable agreement between the proposed description and the experiment is obvious. Using a model with more parameters would always yield a more satisfactory fit. However, there is neither a statistical nor a physical justification for introducing more than one gap parameter in the description. The “one-gap” model provide already a statistically sound fit to the $\mu$SR as well as specific heat data.

In our opinion, the presence of two superconducting energy gaps in Mo$_3$Sb$_7$ may not find its justification in the experimental data presented by Tran \textit{et al.} in Refs.\textsuperscript{1} and \textsuperscript{13}. The field dependence of the muon-spin depolarization rate $\sigma_{sc}$ is well described by assuming the field independent magnetic penetration depth $\lambda$. The temperature dependences of $\lambda^{-2}$ and the electronic specific heat are consistent with what is expected for a BCS superconductor with the single $s$–wave like energy gap.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure3.png}
\caption{(Color online) The electronic specific heat $C_{el}$ as a function of $T^{-1}$ after Ref.\textsuperscript{13}. Lines correspond to the one gap fit with (the solid line) and without (dashed line) the residual electronic specific heat. Note the logarithmic $C_{el}$ scale.}
\end{figure}

* Corresponding author: rustem.khasanov@psi.ch
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