

Comment on “Muon-spin-rotation study of the superconducting properties of Mo₃Sb₇”

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In a recent article, Tran *et al.* [*Phys. Rev. B* **78**, 172505 (2008)] reported results from muon-spin-rotation (μ SR) measurements of an Mo₃Sb₇ superconductor. Based on the analysis of the temperature and the magnetic field dependences of the Gaussian relaxation rate, σ_{sc} , 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 μ SR data does not account for the reduction in σ_{sc} 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 μ SR results [*Phys. Rev. B* **78**, 014502 (2008)], all seem to be consistent with the presence of *single* isotropic superconducting energy gap in Mo₃Sb₇.

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I. THE MAGNETIC FIELD DEPENDENCE OF THE μ SR DEPOLARIZATION RATE σ_{sc}

It is commonly accepted that the Gaussian muon-spin-depolarization rate [square root of the second moment of the muon-spin-rotation (μ SR) line] of the superconductor in the vortex state, σ_{sc} , is related to the magnetic penetration depth, λ , in terms of¹

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

Here $A(b)$ is the proportionality coefficient as a function of the reduced magnetic field, b , ($b = B/B_{c2}$, B_{c2} is the upper critical field). Dependence of the proportionality coefficient A on b accounts for the reduction in σ_{sc} caused by increase in overlapping of vortices by their cores with increasing magnetic field. Following Brandt,¹ only at very low fields ($0.13/\kappa^2 \ll b \ll 1$, $\kappa = \lambda/\xi$, ξ is the coherence lengths) one can neglect the dependence of $A(b)$ on the reduced magnetic field and assume it to be constant.

The condition $A(b) = \text{const}$ is 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 [$B_{c2}(0.1 \text{ K}) \approx 2$ T and $B_{c2}(1 \text{ K}) \approx 1.4$ T are taken from Ref. 3]. As follows from Fig. 6 of Ref. 1 for a superconductor with $\kappa > 50$, such as Mo₃Sb₇, $A(b)$ is strongly field dependent in these field regions. This implies, that in order to obtain λ from $\sigma_{sc}(B)$, one needs to account for the reduction in $A(b)$ due to an increasing magnetic field. This can be done by application of the London model (as is made by the authors, but without accounting for some possible limitations of the model, see the discussion below), or by using the approach developed by Brandt in Ref. 1.

Based on the Ginzburg-Landau theory for superconductors with single isotropic *s*-wavelike gap, the equation¹

$$\sigma_{sc} [\mu\text{s}^{-1}] = 4.83 \times 10^4 \times (1 - b) \times [1 + 1.21(1 - \sqrt{b})^3] \lambda^{-2} [\text{nm}] \quad (2)$$

can be used to fit experimental $\sigma_{sc}(B)$ data of Tran *et al.*,² see Fig. 1. As shown in Ref. 1, Eq. (2) gives an error of less than 5% in the field variation in σ_{sc} for an ideal triangular vortex lattice and holds for type-II superconductors with the value of the Ginzburg-Landau parameter $\kappa \geq 5$ in the range of fields $0.25/\kappa^{1.3} \leq b \leq 1$.

As the measured data of Tran *et al.*² can be well described by Eq. (2), it suggests that λ is field independent and therefore Mo₃Sb₇ has only one isotropic energy gap. We also want to note that in our recent paper,⁴ which was published 3 months before the submission of Tran *et al.*,² the magnetic field dependence of σ_{sc} in Mo₃Sb₇ was measured in fields up to four times higher ($\mu_0 H = 0.2$ T, $b \approx 0.1$) and was found to be consistent with Eq. (2).

In reference to the interpretation of μ SR data, we note that the authors of Ref. 2 have mixed the statement of field-dependent λ and σ_{sc} . The muon-spin-depolarization rate of the superconductor in the vortex state σ_{sc} is *always* field dependent while dependence of λ on the magnetic field is the characteristic of unconventional superconductors (such as cuprates,⁵⁻⁷ pnictides,⁸ double gap MgB₂,⁹ etc.). In a single gap *s*-wave superconductor the magnetic penetration depth is independent of the magnetic field.^{6,10,11} In Ref. 5, which was cited by Tran *et al.*² in order to justify the unconventional two-gap superconductivity in Mo₃Sb₇, Sonier refers to the field-dependent penetration depth λ , but not the muon-spin-depolarization rate σ_{sc} .

II. MODIFIED LONDON MODEL

The fit of σ_{sc} vs B data by means of the modified London model, as used by Tran *et al.*,² is in favor of the “one-gap” picture. Note that the London model is based on the assump-

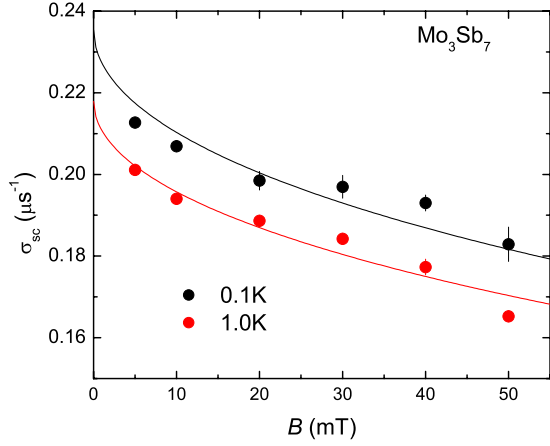


FIG. 1. (Color online) The fit of $\sigma_{sc}(B)$ obtained by Tran *et al.* (Ref. 2) by assuming field-independent λ . See text for details.

tion that λ is field independent. A comparison of this model with the measured $\sigma_{sc}(B)$ would be contradictory as the key argument by Tran *et al.*² is field dependent λ .

We want to stress however, that the London model uses some simplifications and assumptions which are valid only for an extreme type-II superconductor ($\lambda \gg \xi$) for fields in the region $0 \ll B \ll B_{c2}$. The application of this model in order to describe the experimental μ SR data needs to be justified for each particular case, which the authors have not done. On the other hand, the results of numerical calculations by Brand,¹ which are valid for any type-II superconductor and over the whole field region (from 0 up to B_{c2}), are free from these imperfections.

III. DEPENDENCE OF THE INVERSE SQUARED MAGNETIC PENETRATION DEPTH λ^{-2} ON TEMPERATURE

The coefficient $A(b)$, defined in Eq. (1), varies with temperature, whose dependence needs to be accounted for in experiments. This is caused by the temperature dependence of the second critical field B_{c2} and therefore, that of b

$= B/B_{c2}(T)$. The detailed description of $\sigma_{sc}(T)$ to $\lambda^{-2}(T)$ reconstruction procedure, also in application to Mo_3Sb_7 , is given in Refs. 4, 11, and 12.

Figure 2(a) shows $\lambda^{-2}(T)$ normalized to its value at $T=0$ for Mo_3Sb_7 superconductor. The solid black and the red circles refer to $\lambda^{-2}(T)$ reconstructed from $\sigma(T)$ of Tran *et al.*² and that reported in Ref. 4, respectively. The lines correspond to the fit of $\lambda^{-2}(T)$ from Ref. 4 by assuming Mo_3Sb_7 is a superconductor with a *single s-wavelike* energy gap within the clean (solid line) and the dirty (dashed line) limit. 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.

Results of the unnormalized $\lambda^{-2}(T)$ are shown in Fig. 2(c). The solid line corresponds to the “clean-limit” two-gap fit of $\lambda^{-2}(T)$ reconstructed from $\sigma(T)$ of Tran *et al.*² The fit results in *similar* values of the big and the small gap ($\Delta_{0,1} = \Delta_{0,2} \approx 0.41$ meV), thus implying that there is no need to introduce more than one gap parameter in order to describe $\lambda^{-2}(T)$ of Mo_3Sb_7 superconductor.

IV. DEPENDENCE OF σ_{sc} ON TEMPERATURE

As mentioned above, the temperature dependence of σ_{sc} is not the same as the one of λ^{-2} . Even by assuming the opposite, the authors did not present the results of the one-gap fit with the superconducting energy gap being a free parameter. Instead, they fixed the ratio $2\Delta_0/k_B T_c$ to the weak-coupling BCS value of 3.53. Figure 2(b) reveals that the one-gap fit can satisfactory describe even the “uncorrected” $\sigma_{sc}(T)$ data (note that the one-gap fit requires less parameters than is needed for the two-gap model).

We also want to point out the difference between $\sigma_{sc}(T)$ shown in Fig. 2(b) and that reported by Tran *et al.* in Fig. 3 of Ref. 2. The reasons for this are: (i) Tran *et al.* have systematically overestimated the values of σ_{sc} in the vicinity of T_c . For example, $\sigma_{sc}(2.1 \text{ K}) \approx 0.01 \mu\text{s}^{-1}$ (see Fig. 3 in Ref. 2) while according to Fig. 2 of the same reference it must be at 0. (ii) The errors in σ_{sc} were not propagated correctly. Following the relation $\sigma_{sc} = (\sigma^2 - \sigma_{nm}^2)^{0.5}$ (σ_{nm} is the nuclear

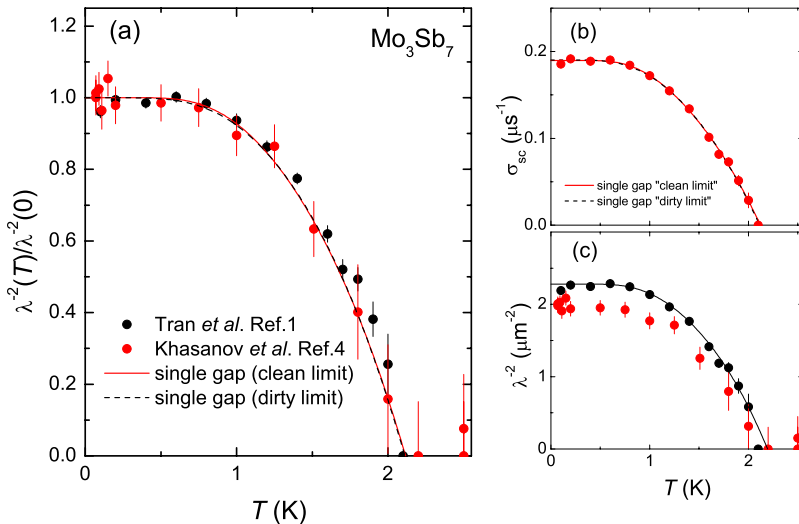


FIG. 2. (Color online) (a) $\lambda^{-2}(T)/\lambda^{-2}(0)$ of Mo_3Sb_7 reconstructed from $\sigma(T)$ data of Tran *et al.* (Ref. 2) (black circles) and that reported in Ref. 4 (red circles). The solid lines represent the single-gap fit of $\lambda^{-2}(T)/\lambda^{-2}(0)$ data assuming Mo_3Sb_7 is a superconductor within the clean (solid curve) and the dirty (dashed curve) limit (after Ref. 4). (b) $\sigma_{sc}(T)$ reconstructed from $\sigma(T)$ data of Tran *et al.* (Ref. 2). The solid line is the single-gap fit of $\sigma_{sc}(T)$. Note the difference of $\sigma_{sc}(T)$ from that reported in Fig. 3 of Ref. 2 (see text for details). (c) $\lambda^{-2}(T)$ reconstructed from $\sigma(T)$ data of Tran *et al.* (Ref. 2) (black circles) and that reported in Ref. 4 (red circles). The solid line corresponds to the two-gap fit revealing $\Delta_{0,1} = \Delta_{0,2} \approx 0.41$ meV (see text for details).

moment contribution), the uncertainty in σ_{sc} should increase when approaching T_c , see Fig. 2(b).

V. ABSOLUTE VALUE OF λ

The 1 nm error in the absolute λ value is unrealistic. The fit was performed within the framework of the modified London model whose validity in application to Mo_3Sb_7 and the conditions of the experiment have not been justified. The fit of $\sigma_{sc}(B)$ by means of 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

More importantly, the authors did not account for *any* possible sources of errors, which in general can lead to much bigger uncertainty in the absolute λ value. This includes: (i) vortex lattice disorder; (ii) different possible symmetries of the vortex lattice (triangular vs. square); (iii) non-Gaussian (asymmetric) line shape of the μSR line which is expected to be seen even in a powder sample of the isotropic (or weakly anisotropic) superconductor; (iv) the background contribution from the Ag backing plate which may be influenced by the magnetic field expelled by Mo_3Sb_7 superconductor, etc. As shown in Ref. 4, the proper consideration of the above mentioned uncertainties leads to the error in the absolute value of λ of Mo_3Sb_7 to be as big as $\sim 100 \text{ nm}$. For these reasons and accounting for the incorrect assumption about temperature-independent proportionality between λ^{-2} and σ_{sc} (see the discussion above), the data of Tran *et al.*² are not an accurate representation of the true magnetic penetration depth.

VI. TEMPERATURE DEPENDENCE OF THE ELECTRONIC SPECIFIC HEAT

One of the arguments pointing to the presence of two superconducting energy gaps in Mo_3Sb_7 was an agreement of the gap values obtained in Ref. 2 with those deduced by Tran *et al.*¹³ in specific-heat experiments. Figure 3 shows the specific-heat data from Ref. 13 together with fits based on the one-gap BCS model. Note that the presence of the small temperature-independent residual electronic specific heat

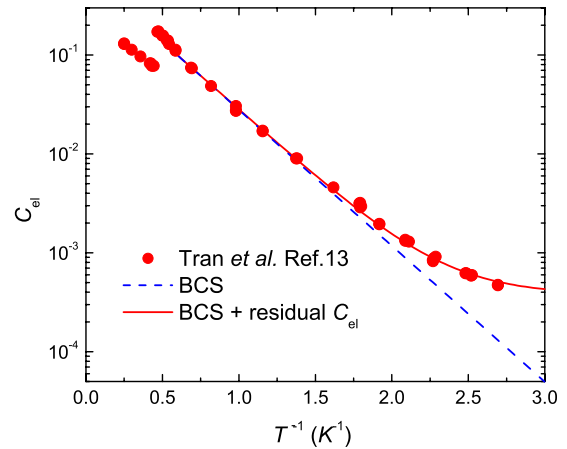


FIG. 3. (Color online) The electronic specific heat C_{el} as a function of T^{-1} , after Ref. 13. Lines correspond to the one gap fit with (the solid line) and without (dashed line) the residual component of the electronic specific heat. Note the logarithmic C_{el} scale.

may be caused by a second nonsuperconducting band and/or by the presence of small inclusions of metallic impurities. These assumptions lead to a good fit of the experimental data to the one-gap model (see Fig. 3, note the logarithmic C_{el} scale).

VII. CONCLUSIONS

The fact that the “two-gap” fits performed by Tran *et al.* in Refs. 2 and 13 result in 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 of the μSR , as well as the specific-heat data of Mo_3Sb_7 superconductor.

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