

Dear Editors,

We thank the Referees for their detailed feedback, recommendations, and the time and effort they put into reviewing this manuscript. We are pleased that both Referees recommended publication in SciPost Physics based on the novelty and importance of the results after their comments were addressed. We answer each point below. Our response is colored red, and the modified text of the manuscript is colored blue.

Referee 1

The manuscript comprises a comprehensive study of magnetic field penetration to a single-crystalline Mo₈Ga₄₁ superconductor. It brings well presented, original and interesting results. In the manuscript sometimes shortcuts are present, some parts are confusing. I would recommend the publication after the authors clarify/correct the issues listed below.

RESPONSE: We thank the referee for this very positive comment and publication recommendation.

A1) use proper expression for the ratio of the energy gap and the critical temperature (involve Boltzmann constant), especially when you refer to specific numbers.

RESPONSE: In the current literature on superconductivity, it is quite common to set Boltzmann's constant to 1 because what matters is the dimensionless ratio of the gap amplitude to the thermal energy, $k_B T$. However, we followed the Referee's recommendation and added the Boltzmann constant, k_B , where needed.

A2) try to avoid mixing SI and CGS units in the same text. CGS is obsolete - it is easy to use mT instead of Oe.

RESPONSE: This is a typical case where the so-called "practical" system of units is used. Here, the applied field strength, H , is in oersted, and the magnetic induction, B , is in tesla. In SI, the former is measured in A/m, an inconvenient and rarely used unit. We could use all CGS units and express the B-field in gauss, but tesla is better for its factor of 10000. We note that our magnetometer software produces data in CGS units.

A3) In some parts of the text there are references to numbers of the sections (Section II, Section III.B) even if the sections are not numbered. It seems that it is some residuum from an older manuscript layout. Specially referring to Section III.B in a third section and second subsection is confusing.

RESPONSE: We have reviewed the proper numbering of sections and subsections, as suggested.

B1) The authors claim that in Ref.9 the larger gap gives the ratio $\Delta/k_B T_c = 1.857$. However, this was calculated considering the smallest value from the distribution of the larger gaps (1.45 meV), moreover, local T_c of the junction with this energy gap was not determined. On the contrary, the temperature dependence of the two energy gaps with clearly identified critical temperature presented in Ref.9 gives the value of Δ 1.6, that leads to the ratio of 2.06, which is larger than the weak-coupling limit.

RESPONSE: While the text of Ref.9 quotes 1.6 meV and 0.9 meV for the two gaps, and these values appear in the field-dependence of the gaps, Fig.2(d), the temperature dependence, Fig.2(b), shows the gaps of 1.46 meV and 0.9 meV at $T \rightarrow 0$. Furthermore, the text says (bottom left column of page 3): “For the larger gap of $\text{Mo}_8\text{Ga}_{41}$, $2\Delta/k_B T_c$ is found to be 3.5, which is close to the expected value for a weak-coupling BCS superconductor.” We also note that the two-gap fitting of the tunneling spectra involves a fairly large phenomenological Dynes parameter Γ , which signifies some issues with the tunneling barrier. It is known that larger Γ tends to push the gap peaks to larger values. Of course, this is not our place to analyze Ref.9 in detail. We therefore revised our text in the introduction to read:

A scanning tunneling microscopy (STM) study of $\text{Mo}_8\text{Ga}_{41}$ crystals suggests a weak-coupling superconductivity with two gaps of 1.6 meV and 0.9 meV, resulting the ratio, $\Delta(0)/T_c$, of 2.1 and 1.2, and concludes that this behavior is similar to MgB_2 \cite{Sirohi2019}.

B2) The authors claim that in Ref.16 the larger from the two gaps has the ratio even smaller than the weak-coupling limit. This is not true. Actually, it is around 5, which is really unexpectedly far above the weak-coupling limit.

RESPONSE: The Referee is correct. We now mention this in the revised and extended text, please see below a combined response to B2 and B3.

B3 - The authors comment on the results presented in Ref.17, stating that the data can be fit well both with single-gap and two-gap fit with minor differences. This is true, but the main message of the paper is that none of the options is valid, because the single-gap fit leads to the ratio much lower than the weak-coupling limit and the two-gap fit is not consistent with the heat capacity measurement performed on the same crystal. Instead,

the paper is a warning that the measurement of the superfluid density itself, without knowing the complex picture, might be misinterpreted e.g. for a two-gap feature. This context is not mentioned in the Introduction.

RESPONSE: Reference 17 shows an excellent single gap fit, Fig.7, and the text below says: “*The fit using Eq. (4) is shown in Fig.7 as a dashed line. The single-gap BCS-type model satisfactorily describes the experimental data with $\Delta(0) = 1.80(7)$ meV, $\Delta(0)/kBT_c=2.1\dots$ ”.* This value is a bit larger than 1.76 of the clean limit but could be due to the fact that the authors used a dirty-limit expression for fitting.

However, as the Referee pointed out, the authors of Ref.17 attempt to reconcile their results with previous specific heat measurements by the same group and arrive at some very large gap values, far exceeding the weak coupling limit. However, their two-gap analysis is not self-consistent, and one can find many different pairs of gaps of different amplitudes to fit the data. A full Eliashberg-type analysis deriving temperature-dependent gaps from the self-consistency equations and then evaluating the superfluid density has to be performed. Considering how much the gap(T) changes in the strong-coupled superconductor (new reference: Marsiglio and Carbotte 1991) it is certain that the superfluid density with such large gaps will look very different from the weak-coupling single-gap BCS.

We note that there are a few superconductors (e.g. Rh17S15) where a specific heat jump at T_c is much larger than expected from the weak-coupling theory, but it is unclear if this signifies a strong coupling or if there are other contributions to the free energy from other degrees of freedom.

This part of the introduction is rewritten:

Specific heat measurements reported larger than weak coupling jump at T_{c} \cite{Verchenko2016}. A scanning tunneling microscopy (STM) study of $\text{Mo}_8\text{Ga}_{41}$ crystals suggests weak coupling superconductivity with two gaps of 1.6 meV and 0.9 meV, yielding the ratios $\Delta(0)/T_c$, of 2.1 and 1.2, and concludes that this behavior is similar to MgB_2 \cite{Sirohi2019}. The superfluid density obtained by a muon spin resonance (μSR) study is consistent with a single-gap weak coupling isotropic BCS theory \cite{Verchenko2017}. However, to connect with the specific heat studies of the same group \cite{Verchenko2016}, the same data were fitted with two very large gaps, 4.3 meV and 1.76 meV, resulting in gap ratios of 5.1 and 2.1, respectively. However, these calculations use a non-self-

consistent α -model-type approach with approximate BCS temperature dependencies for both gaps, and using a dirty limit formula. Considering how different the temperature dependence of the gap is in a strongly coupled superconductor \cite{Marsiglio1991}, it is certain that a self-consistent Eliashberg theory would not agree with this analysis.

C) In the section Samples and methods, subsection Lower critical field, the authors claim that the measurements are performed as close to the sample edge as possible and refer also to Ref.17. However, in Ref.17 it is explained that for the measurement of the penetration field in this sample featuring strong pinning (V-shaped profile), a probe is selected that is distant from the edge by the half-width of the sample. This does not mean "as close as possible", actually the selected probe was the forth in a row from the edge (see Fig.1 in Ref.17). I would suggest to elaborate this part - mentioning the sample shape and sizes, and estimate the position of the measurement in respect to the sample edge. Was some other position examined as well?

RESPONSE: We used a cuboid-shaped sample with dimensions: 400 μm *200 μm *190 μm . In Ref.17, it is clear that there is a significant uncertainty in the actual sample edge location and the elevation of the Hall probe above the surface. If they could, they would choose the probe closest to the edge, but the first probe where flux penetration is clearly identified is probe #6, with probe #5 being the "apparent" edge. In our case of NV centers dispersed in a diamond film, we have a much finer spatial resolution. The spot where we could clearly identify the superconducting screening was 10 μm into the sample. We updated the relevant description accordingly.

D) I wonder why the authors used weak-coupling limit for calculation of λ . The other literature refers mostly to moderate or strong coupling (Ref.9: $\Delta/k_{\text{BT}_c} = 2.06$; Ref.15: jump in C_p 2.83>1.43; Ref.16: $\Delta/k_{\text{BT}_c} = 2.1$ if single-gap fit is considered; Ref.17/18: $\Delta/k_{\text{BT}_c} = 2.2$). What would be the difference in the penetration depth and subsequently in the superfluid density (since one needs to feed the value of $\lambda(0)$ for the calculation), if larger coupling was involved?

RESPONSE: This is the central point of our paper. The stronger coupling values, quoted in the comment, would result in the superfluid density significantly different from the weak coupling. The gap(T) changes dramatically – we now cite a paper by Marsiglio and Carbotte (1991). The gap value affects the superfluid density curve exponentially, so it is very sensitive to it. It is important that we fit the data in the full temperature

range, whereas specific heat relies on the jump at T_c . On the other hand, the value of the London penetration depth is only in the pre-factor of the low-T expansion, and it is obtained from an independent measurement. Furthermore, we must point out that, despite of what is often done in the literature – using the Δ/T_c ratio as a variable, one cannot use the weak-coupling BCS formulas to compute the superfluid density with arbitrary not-self-consistent gaps. A full Eliashberg calculation must be performed. Since we successfully fit the data with the simplest weak-coupling theory, we do not need to consider a stronger coupling because we know that the curve will certainly be different.

E) In the section Results and Discussion, subsection London penetration depth..., there is a sentence I consider to be too simplistic: "There is no indication of a multi-gap behavior, which usually appears as a convex curvature ..." The shape of the superfluid density depends on various parameters - mostly on how far are the two energy gaps apart and what is the weight of their contribution. It is true, that most of the clearly identified two-gap superconductors have this feature, but it is not general. In other words, absence of the positive curvature does not necessarily exclude two-gap superconductivity.

This is correct. To address this verbatim, we introduced a new paragraph:

Addressing the possibility of two distinct gaps, we note that, unlike the not-self-consistent α -model where temperature dependence of the two gaps is assumed to be BCS-like, the self-consistent two-band treatment shows that the smaller gap becomes smaller than what is expected from the BCS. If the interband pairing is not too strong, the resulting superfluid density, $\rho_s(T)$, develops a visible suppression at intermediate and higher temperatures. (In the limit of zero interband pairing, there will be two different T_c values!). None of the reported measurements so-far showed such features in the temperature dependencies of the gaps or the superfluid density. The tunneling study suggested that their results are similar to MgB₂ \cite{Sirohi2019}. However, this is not the case from the superfluid density point of view, because in MgB₂, $\rho_s(T)$ is clearly suppressed at elevated temperatures and this was one of the signatures that helped to identify two-gap physics \cite{Carrington2003,Kim2019}.

F) Description of Fig. 3 looks confusing - the Y-axis is called λ_m , in the figure

caption it says that it shows $\Lambda_m(T) \cdot \lambda(0) + \Delta\Lambda(T)$ and in the text it states "note that the measured $\Delta\Lambda_m(T)$..." Also the notion of saturation is rather confusing, since all the curves in Fig.3 increase at elevated temperatures, while their values overlap.

RESPONSE: We have corrected the description as $\Lambda_m(T) = \lambda(0) + \Delta\Lambda(T)$ and text as $\Lambda_m(T)$ instead of $\Delta\Lambda_m(T)$.

The Referee is correct. We now re-phrased the description and, instead "saturation" use "does not diverge". Specifically, the text now reads:

Note that the measured $\lambda_m(T)$ does not diverge approaching the normal state. Of course, the actual penetration depth diverges at $T \rightarrow T_c(H)$, but in the normal state, the measured depth λ_m cannot exceed the skin depth, $\delta_{\text{skin}} = \sqrt{\rho/\mu_0 \pi f}$, where $\mu_0 = 4\pi \times 10^{-7} \text{ H/m}$ is the vacuum permeability and ρ is the resistivity above T_c . Therefore, the $\lambda_m(T)$ curves in Figure~\ref{fig:Lm} change the behavior entering the normal state and above $T_c(H)$ follow the temperature dependent $\delta_{\text{skin}}(T)$. In fact, such measurements can be used for contactless resistivity measurements \cite{Prozorov2007}.

G) The first sentence of the section Results..., subsection Campbell penetration depth... looks confused, probably with wrong position of the bracket.

RESPONSE: It was reviewed and corrected.

Referee 2

Mo8Ga41 is a potentially unconventional superconductor that has been controversially discussed in the previous literature. The manuscript under consideration reports the very first penetration depth measurement that demonstrates an unexpected but remarkably good agreement with the BCS expression in the clean limit. This interesting result sheds new light on the physics of Mo8Ga41 and potentially meets the SciPost Physics criterion of presenting a breakthrough in an existing research direction. Moreover, a curious peak effect in the critical current is reported and analyzed. I believe that this manuscript could be published in SciPost Physics, but it requires a major revision along the following lines.

RESPONSE: We thank the referee for this very positive comment and overall recommendation to publish this paper in SciPost Physics.

- 1) One major puzzle is the simple BCS behavior of the superfluid density, as opposed to several previous observations of the "less conventional" superconductivity in Mo₈Ga₄₁. Some of these reports could be affected by the ambiguous fitting of the muon data or by surface effects, but bulk probes such as heat capacity should be more tenable. The large jump in the specific heat at T_c [PRB'2016] indicates some physics beyond BCS's weak-coupling limit. This observation directly contradicts the conventional scenario advocated by the present work. A plausible explanation for this discrepancy should be given.

RESPONSE: This is a question similar to B3 of the Referee 1. We note that there are a few superconductors (e.g. Rh₁₇S₁₅) where specific heat jump at T_c is much larger than expected from the weak-coupling theory. It is unclear if this signifies a strong coupling or not. Perhaps, there are other contributions to the free energy (entropy) from other degrees of freedom. However, such large gap taken at a face value is simply incompatible with the measurements of superfluid density or tunneling. A full Eliashberg-type analysis of the superfluid density and temperature-dependent specific heat (not just its jump at T_c) is needed to address this important question.

- 2) One plausible explanation is the sample dependence. In this regard, details of the sample characterization are essentially missing, and it is not even clear whether exactly the same crystal(s) as in Ref. [14] have been used. I strongly encourage the authors to provide additional characterization data, such as resistivity, magnetization, heat capacity that could be compared to the previous publications. It would also be natural to supply Fig. 8 with the B_{c2} values from the existing literature. Do they match?

RESPONSE: The samples used in this study were from the same batch as used described in Ref.[14], as can be seen from the shared co-authors between two works. No variation of properties was found within the batch. The resistivity and magnetization measurements were reported in the original paper, Ref.[14].

- 3) Another possibility is that the superconductivity of Mo₈Ga₄₁ is not of the simple BCS type, but a more complex scenario mimics the BCS temperature dependence of the superfluid density, such that the good match in Fig. 2 is merely accidental. This idea may not be so odd if the physics of the normal state is considered. Mo₈Ga₄₁ is far from being a simple metal. Its resistivity exceeds the MIR limit and shows a rather peculiar temperature dependence below 100 K. The mean-free

path should be quite short. Would one really expect the clean limit to be applicable in this case? Should not the dirty limit be more appropriate? What is the mean-free path in your sample?

RESPONSE: It is not easy to estimate the mean mean free path in this multiband system. However, the $RRR=17$ is quite good. We also see from our Fig.2 that the dirty limit would result in distinctly different superfluid density. Finally, Ref.[17] reports well-resolved quantum oscillations, which is impossible in a dirty system.

- 4) The lower critical field was determined by the NV-center magnetometry. The B_{c1} value at 4.25 K is somewhat lower than reported in PRB'2016: 85 Oe vs. 110-115 Oe. What is the reason for this difference? How was the sample surface prepared?

RESPONSE: Our NV center magnetometry is a local probe that looks at the field of first penetration. The magnetization measurement, such as in PRB 2016, is a bulk probe, and there is a significant issue of an unknown effective demagnetizing factor of a non-ellipsoidal sample. Furthermore, it is extremely hard to distinguish the moment when flux starts to penetrate the sample. All this results in a large uncertainty.

We screened numerous samples to identify the best one with well-defined edges. It's important to note that the same sample was utilized for both NV magnetometry and tunnel diode oscillator measurements to ensure consistency.

- 5) On page 5 there is a typo in the description of the critical current at 2 T and 4 T in the FC and ZFC regimes. The sentence with "the ZFC curves reverse this order" should probably read as "the FC curves reverse this order". According to Fig. 6, the order is reversed below 4 K only. Why does it not happen at higher temperatures? Would it be possible to shed more light on the vortex state of Mo₈Ga₄₁? For example, is there a vortex liquid, and in which temperature range?

RESPONSE: We have corrected the typo.

The Referee is correct. We clarified the statement and write:

Surprisingly, the FC curves reverse this order below roughly 4 K, so $j_c(2\text{T}) < j_c(4\text{T})$, showing an increase of j_c with increasing magnetic field.

Regarding the vortex state studies. Indeed, this system shows interesting behavior. We plan to conduct ac and dc magnetic measurements and measure magnetoresistance to fully investigate vortex physics in this fascinating superconductor. Of course, this will take some time to complete.