Clear evidence against superconductivity in hydrides under high pressure

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Short Report

Keywords: Meissner effect, superconductors, sulfur hydride

Posted Date: October 26th, 2021

DOI: <https://doi.org/10.21203/rs.3.rs-1001084/v1>

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Clear evidence against superconductivity in hydrides under high pressure

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PACS numbers:

The Meissner effect, magnetic field expulsion, is a hallmark of superconductivity. Associated with it, superconductors exclude applied magnetic fields. Recently Minkov et al. presented experimental results reportedly showing “definitive evidence of the Meissner effect” in sulfur hydride and lanthanum hydride under high pressure [1]. Instead, we show here that the evidence presented in that paper does not support the case for superconductivity in these materials. Together with experimental evidence discussed in earlier papers, we argue that this clearly indicates that hydrides under pressure are not high temperature superconductors.

The era of high temperature superconductivity in hydrides under high pressure was spawned by the reported discovery of superconductivity in sulfur hydride in 2015 by Eremets and coworkers [2], with critical temperature 203 K, higher than any other critical temperature known before. Since then, it has been reported that superconductivity at high temperatures occurs also in 11 other hydrides under pressure [3, 4]. These experimental works are strongly motivated and guided by theoretical predictions of superconductivity in these materials based on the conventional BCS-electron-phonon theory of superconductivity [5–8].

Instead, we have recently argued that the experimental evidence presented so far does not provide conclusive proof of superconductivity in any of these materials [9–18]. Others have also questioned experimental [19–21] and theoretical [22, 23] evidence for superconductivity in some of these materials. Therefore, the very existence of high temperature superconductivity in hydrides under pressure is now in question.

In the initial paper [2], Eremets and coworkers presented magnetic evidence of superconductivity in sulfur hydride, that was questioned in ref. [12]. Recently, Eremets and coworkers provided new magnetic evidence for superconductivity in sulfur hydride as well as in lanthanum hydride [1], and argued that it provides definitive evidence for superconductivity. Instead, we argue in this paper that these new measurements together with the old measurements provide conclusive evidence *against* the existence of superconductivity in these hydrides.

To have confidence that experimental results reflect true physics of the material being studied, it is essential that measurements are reproducible, not only within one lab and experimental group but also in different settings. Unfortunately, no other group has reported mea-

surements of magnetization in sulfur hydride (nor any other hydride) under high pressure.

The experimental results on sulfur hydride reported by the Eremets group in 2015 [2] and 2021 [1] report critical temperatures of 203 K and 196 K respectively, i.e. they are very close to one another. The size of the samples used is also reported to be similar, approximately 80 μm [2] and 85 μm [1] in diameter and “a few μm ” in thickness in [2] and between 2.1 μm and 3.1 μm in thickness in [1]. However, the measured magnetic moment of the samples under an applied magnetic field of the same magnitude differ by a factor of 5, with the 2015 sample having the larger magnetic moment. No explanation for this large discrepancy is given in [1]. These magnetic moments are measured not by field cooling but by applying a magnetic field to an already cold sample, and for sufficiently small fields so that the field should not penetrate the sample, hence such large differences are not expected for samples of similar sizes.

More importantly, the magnitude of the lower critical field reported in 2015 [2] was 30 mT, and the same quantity in 2021 is 1.9 T [1]. That is a difference of a factor of 60. The magnitude of the London penetration depth was reported to be 125 nm in 2015 [2] and 12.7 nm in 2021 [1], a difference of a factor of 10.

Thus, for the past 6 years the physics community was asked to believe that sulfur hydride had been proven to be a 203 K superconductor based on magnetic measurements that were wrong by these very significant factors. The magnetic evidence reported by Eremets and coworkers in 2015 [2] was regarded by many to be the strongest evidence that hydrides under pressure are high temperature superconductors [24]. Now, Eremets et al. themselves are telling us that that evidence was deeply flawed [1]. In other words, the entire edifice of high temperature superconductivity in pressurized hydrides was built on deeply faulty foundations.

It is also remarkable that after the reported magnetization measurements in 2015 using a specially designed miniature nonmagnetic DAC cell that could accommodate a SQUID magnetometer [2], no new experimental results using that sophisticated apparatus and technique were reported for a full 6 years, neither by the authors of ref. [2] nor by anybody else. Yet during those years, about 30 new reports of high temperature superconductivity in 12 different pressurized hydrides were published [3, 4].

Fast forward to 2021 [1]. Figure 1 shows on the left panel the reported magnetization versus temperature under an applied magnetic field, and on the right panel the

FIG. 1: Left panel: magnetization versus temperature of sulfur hydride reported in ref. [1], under field cooling (FC) and zero field cooling (ZFC). Right panel: magnetization versus applied magnetic field for sulfur hydride reported in ref. [1].

magnetization versus magnetic field for various values of the temperature [1]. It can be seen that no evidence of a superconducting transition is seen under field cooling on the left panel. While this behavior has been observed in some strongly type II superconductors, it has never been observed for type I or weakly type II superconductors. According to ref. [1], this material is a weakly type II superconductor, with Ginzburg-Landau parameter $\kappa = 6.9$. The reported London penetration depth is remarkably small, 12.7nm , indicating that the material has a large superfluid density and small degree of disorder. Such materials always exhibit a robust Meissner effect, i.e. magnetic flux *expulsion*. We argue that the fact that this material does not show *any* evidence of magnetic field expulsion under zero field cooling, the signature of superconductivity, is clear and direct evidence that the material is not a superconductor.

Ref. [1] claims that the sample is a flat disk of demagnetization factor $N = 0.95$. From this information, and from the observation that a magnetic field at low temperatures of magnitude $H_p \sim 96\text{ mT}$ starts to penetrate the sample, as seen in the lowest curve on the right panel of fig. 1, labeled 20 K, the authors infer that the lower critical field of sulfur hydride is $H_{c1}(T = 0) = 1.9\text{ T} = H_p/(1 - N)$. In figure 2 we plot the behavior of the lower critical field of a standard superconductor as a function of temperature inferred from Ginzburg-Landau theory. The horizontal lines indicate the values of the lower critical field H_{c1} for the values of applied field indicated, $H_p = 10\text{ mT}$, 20 mT , etc. The squares indicate the critical points for each value of the applied magnetic field.

For each value of the magnetic field, for temperatures lower than that critical value, the magnetic field should be completely excluded from the sample, except for the small region within λ_L of the surface. Given that the penetration depth is 12.7 nm and the sample diameter and

FIG. 2: Lower critical field versus temperature for a standard superconductor according to Ginzburg-Landau theory. The zero temperature critical field is 1.9 T . The numerical values of the field given in mT indicate the applied field H_p , that becomes $H_{c1} = 20H_p$ due to the demagnetization factor $1/(1 - N)$ for $N = 0.95$ reported in ref. [1].

height are $85\text{ }\mu\text{m}$ and $2.8\text{ }\mu\text{m}$ respectively, this implies that 99.5% of the sample remains field free except very close to the critical temperature. Therefore, we expect the magnetization versus temperature to be essentially flat below each critical temperature. This is approximately consistent with what is seen for the three curves shown in the left panel of Fig. 1. For those values of the applied field, $H_p = 2\text{ mT}$, 4 mT and 10 mT , the critical temperatures are 194.4 K , 192.8 K and 187.8 K , all close to the zero field critical temperature 196 K .

From the reported values of magnetic moment versus magnetic field shown in the right panel of fig. 1 we can extract the behavior of magnetic moment versus temperature for applied fields larger than the values shown on the left panel of fig. 1. This is shown in fig. 3. The value of the abscissa for each square is the critical temperature for each value of the applied field inferred from fig. 2, and we would expect the magnetic moment to be approximately *constant* for temperatures lower than those values, since the magnetic field is completely excluded from the sample. The behavior seen in fig. 3 is *qualitatively different* from this expectation.

Indeed, it can be seen in fig. 3 that the sample is unaware of the fact that it underwent a superconducting transition at its critical temperature, denoted by the square on each curve. The magnetization in each case (except for the selected ones that the authors of ref. [1] chose to show us on the left panel of fig. 1, also reproduced in fig. 3), continues its downward trend as the temperature is lowered further, oblivious to the fact that the magnetic field is no longer in its interior. This could be interpreted as revealing yet another new property of non-standard superconductors [10] not shared by standard conventional or unconventional superconductors: an ability to change the magnetic field in their surroundings

FIG. 3: Magnetic moment versus temperature inferred from the experimental results of ref. [1] shown in Fig. 1, for various values of the applied magnetic field H_p given in mT in the figure. The colored squares indicate the point on the curve for the applied magnetic field that corresponds to the critical temperature given by the phase boundary in Fig. 2.

while keeping the magnetic field in their interior constant (i.e. equal to zero). Alternatively, the behavior shown in fig. 3 conclusively establishes that this sample is not superconducting.

In ref. [11], in connection with an analysis of a nuclear resonant scattering experiment on sulfur hydride [25], we pointed out that a lower critical field $H_{c1} \sim 2.6 T$, a thermodynamic critical field $H_c \sim 10.6 T$, and a London penetration depth $\lambda_L = 10.0 nm$ are completely incompatible with the physics of standard superconductors. Those values are remarkably close to the values $H_{c1} = 1.9 T$, $H_c = 9.8 T$ and $\lambda_L = 12.7 nm$ that Erements et al now postulate [1] are the appropriate values for sulfur hydride.

Let us start by computing the critical current. A magnetic field of magnitude smaller than $H_{c1} = 1.9 T$, the value for H_{c1} estimated for sulfur hydride in ref. [1], should be completely excluded from the interior of a long cylinder. The current density circulating near the surface is, according to London's equation

$$J_c = \frac{c}{4\pi\lambda_L} H_{c1} \quad (1)$$

which for the above given values of H_{c1} and λ_L yields

$$J_c = 1.19 \times 10^{10} \text{ Amp/cm}^2. \quad (2)$$

This value is at least two to three orders of magnitude larger than critical currents of any other known superconductor, whether type I or type II, hard or soft [26]. Instead, ref. [1] estimated a value of the critical current density

$$J_c \sim 7 \times 10^6 \text{ Amp/cm}^2 \quad (3)$$

using magnetization measurements and the Bean model. However, such an estimate can only be valid for a strongly type II superconductor with much smaller H_{c1} and larger λ_L , such that the value of magnetic field given by the expression $H = (4\pi\lambda_L/c)J_c$ is larger than H_{c1} . In such a case the magnetic field penetrates the sample and generates vortices that are pinned near the sample surface, and the strength of the pinning potential determines the critical current according to the Bean model. That calculation is not applicable for the case at hand here to determine the critical current.

The density of states at the Fermi energy $g(\epsilon_F)$ can be obtained from the standard thermodynamic relation

$$\frac{H_c^2(0)}{8\pi} = \frac{1}{2} g(\epsilon_F) \Delta^2 \quad (4)$$

with Δ the energy gap at zero temperature. From the standard BCS relation $2\Delta/k_B T_c = 3.53$ and $T_c = 196 K$ [1] we have $\Delta = 29.8 meV$, and using $H_c = 9.8 T$ yields

$$g(\epsilon_F) = \frac{0.537 \text{ states}}{\text{spin} - eV \text{ \AA}^3}. \quad (5)$$

This is an enormous density of states. For comparison, using density functional theory the density of states of sulfur hydride was estimated to be 0.019 states/(spin-eV $\text{\AA}^3$) [7], twenty eight times smaller.

In ref. [11] we discussed in more detail why such numbers are completely incompatible with standard superconductivity. Barring a qualitatively different superconducting state unlike that of all known superconductors, this implies that the properties measured in ref. [1] interpreted to “unambiguously confirm superconductivity” in fact confirm that the material that was measured was not a superconductor.

The analysis discussed above for sulfur hydride applies equally well to the measurements for LaH_{10} reported in ref. [1]. It is claimed that for LaH_{10} the zero temperature lower critical field, thermodynamic critical field and London penetration depth are $H_{c1} = 1 T$, $H_c = 7.4 T$ and $\lambda_L = 20.7 nm$, comparable to the values reported for H_3S and equally impossible. The resulting critical current density for LaH_{10} is $3.84 \times 10^9 \text{ Amp/cm}^2$, and the resulting density of states is $g(\epsilon_F) = 0.22 \text{ states/spin-eV-}\text{\AA}^3$, much larger than the theoretically estimated value of 0.016 states/spin-eV- $\text{\AA}^3$ [7]. The magnetic moment behavior reported for LaH_{10} has the same unphysical behavior as shown above for H_3S .

Adding to these arguments, we argue that Fig. SI1 of the paper [1] is a smoking gun that provides clear

evidence for the faulty analysis and conclusions of ref. [1].

The lower left panel of Fig. SI1 shows the measured magnetic moment versus temperature under an applied magnetic field of 2 mT for the sample precursors, namely $S+NH_3BH_3$ before undergoing the laser heating process that generates the supposedly superconducting sample. The zero field cooling and field cooling curves coincide for temperatures above 200K and diverge below 200K, with the ZFC magnetic moment decreasing and the FC moment increasing as the temperature is further lowered. Precisely the same behavior is observed in the sample after heating, attributed to superconductivity, shown in the left middle panel of Fig. SI1 and in the left panel of Fig. 1 of this paper (red and green curves).

How does the precursor sample know that the critical temperature of the superconducting sample will be close to 200K? Why does an applied magnetic field give rise to a different magnetization for the non-superconducting sample under field cooling and zero field cooling, also seen in the lower middle and right panels of Fig. SI1 for other values of the applied magnetic field?

The authors attribute the difference in the FC and ZFC curves shown in fig. SI1 to “contamination by magnetic pieces” [1, 27]. Clearly such effects, which will not necessarily have the same temperature dependence before and after heating, and hence cannot be simply subtracted out, could also be responsible for the signals interpreted by the authors as superconductivity.

We argue that this observed behavior in the lower panels of Fig. SI1, which obviously is not indicative of superconductivity, but clearly is due to experimental artifacts, strongly suggests that the similar observed behavior shown for the sample after heating in the middle panels in fig. SI1, interpreted by the authors as due to superconductivity, is equally caused by the same experimental artifacts.

We have proposed in ref. [13] that detection of trapped flux would provide convincing evidence for the existence of persistent currents in these materials. It would appear that to do this test would not be more difficult than to perform the measurements reported in refs. [1] and [2]. To rationalize the complete absence of flux expulsion upon field cooling seen in the left panel of fig. 1, one could hypothesize that there is a large concentration of strong pinning centers that prevent the magnetic field from being expelled, even at the large cost of condensation energy implied by the very large H_c . Let us entertain that possibility, even though it doesn’t seem compatible with the reported large value of H_c and small value of λ_L . If after the field cooling process shown on the left panel of Fig. 1 the external magnetic field is turned off, those pinning centers should prevent the interior magnetic field from decaying and a remnant magnetization should be detectable for several hours, days or months thereafter. As shown in refs. [36, 37], the remnant magnetic moment should be given approximately by the difference in the FC and ZFC moments for the same magnetic field.

If such a trapped flux is detected for the sample, *and* it is not detected when the same process is performed for the precursor sample before heating, it will provide convincing evidence that persistent currents flow in the material in the absence of applied magnetic field. Then the unlikely possibility that these materials are ‘nonstandard superconductors’ [10, 11] will have to be seriously considered. Conversely, if no field trapping is observed, it will provide strong confirmation of our arguments that indicate that these systems are not superconducting.

Similarly we have suggested [13] that in a nuclear resonant scattering experiment (NRS) [25] that claimed to show flux exclusion from sulfur hydride from the absence of quantum beats in the NRS spectra under an applied magnetic field, it would be straightforward to verify the presence of trapped field by showing that quantum beats are present after field cooling and removal of the external field.

The magnetic measurements reported in ref. [1] were intended to establish that hydrides under pressure are indeed high temperature superconductors. The paper claims to present *unambiguous* (three times) and *definitive* (once) evidence for high temperature superconductivity in sulfur hydride and lanthanum hydride under pressure. In this paper we argued that instead these recent experimental results establish that these hydrides are not superconductors. Let us summarize our arguments.

- (1) The measurements show that if the materials are superconductors, they have critical current values that are at least three orders of magnitude larger than those of all standard superconductors.
- (2) Their lower critical field and thermodynamic critical field are between one and two orders of magnitude larger than for any standard superconductor.
- (3) Their density of states is between one and two orders of magnitude larger than what is expected for a material with such composition.
- (4) Their London penetration depth is at least a factor of 2 smaller than that of any known superconductor, including very pure elements.
- (5) They show absolutely no evidence for magnetic field expulsion in the presence of a magnetic field (figs. 2c and 2d in ref. [1]). This is completely unprecedented for weakly type II superconductors with such small values of the London penetration depth that imply very high values for the superfluid densities.
- (6) The measured magnetic moment versus temperature under zero field cooling conditions, except for very small magnetic field values, shows monotonic behavior of approximately constant slope with absolutely no signature of a superconducting transition at the expected values of the critical temperature.
- (7) The measured magnetic moment interpreted as due to superconductivity is five times smaller than the magnetic moment interpreted as due to superconductivity in the 2015 paper [2] for a sample of similar size.
- (8) The measured magnetic moment versus tempera-

ture in the presence of a small magnetic field for precursor nonsuperconducting samples shows similar behavior to the behavior allegedly signaling superconductivity in the alleged superconducting samples.

(9) Detection of trapped flux that persists for long periods after field cooling and removal of the external field would show that the materials can carry persistent currents, but it has never been done. Its absence would confirm that the materials are not superconductors.

It should also be pointed out that ref. [1] misleadingly claims that “the Meissner effect” was demonstrated, even in the title. The fact is, the Meissner effect is magnetic field *expulsion*, not magnetic field *exclusion*. Magnetic field exclusion was known to researchers in 1911, magnetic field expulsion was only discovered in 1933. Ref. [1] showed zero evidence for magnetic field expulsion. Similarly ref. [25] claimed to report “direct observation of the expulsion of the magnetic field” from sulfur hydride, when in fact no field cooling was even performed.

In other recent papers we have analyzed various other reported evidence for superconductivity in pressurized hydrides and concluded that every experiment was flawed, namely:

(1) ac magnetic susceptibility reported to show a superconducting transition in sulfur hydride [28] was shown to be due to an experimental artifact [17].

(2) Optical reflectance measurements that reportedly showed that sulfur hydride is a superconductor [29] were shown to be flawed [15].

(3) The reported observation of a Meissner effect in sulfur hydride using nuclear resonant scattering [25] was shown to be flawed [11].

(4) Magnetic susceptibility measurements for a room temperature superconducting hydride [30] were shown to be flawed [18].

(5) Magnetic susceptibility measurements reported for lanthanum hydride showed weak and very broad peaks [31], inconsistent with the width of the presumed transi-

tion shown in resistivity measurements of that material [12, 32, 33].

(6) Resistance measurements for a room temperature superconducting hydride [30] and several other hydrides [2, 33–35] were shown to be anomalously sharp, and/or with an unchanging width in the presence of an applied magnetic field, indicating that the drops in resistance were not due to superconductivity [9, 10, 14].

In addition, an alternative theory of superconductivity predicts that high temperature superconductivity in this class of materials should not exist [38, 39].

The field of high temperature superconductivity in hydrides was launched in 2015 by the publication of ref. [2] by Eremets and coworkers. Now, the same author and coworkers present evidence that (a) invalidates the evidence for superconductivity presented in the 2015 paper and (b) is in itself deeply flawed, as discussed in this paper. From this, together with our analysis of the totality of magnetic evidence and other experimental evidence for superconductivity in hydrides discussed in our earlier papers, we argue that the inevitable conclusion is that hydrides under pressure are not high temperature superconductors. Whatever the origin of their anomalous behavior is, whether intrinsic or due to experimental artifacts or both, it is not due to superconductivity.

Acknowledgments

We are grateful to D. Semenov for calling ref. [1] to our attention, and to V. S. Minkov, S. L. Bud’ko and M. I. Eremets for clarifying correspondence on their paper [1]. JEH would like to thank S. Shylin for informative discussions and sharing of information on ref. [2]. FM was supported in part by the Natural Sciences and Engineering Research Council of Canada (NSERC), and by an MIF from the Province of Alberta.

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