Flux trapping in superconducting hydrides under high pressure

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High temperature conventional superconductivity in hydrogen-rich materials under high pressure has been reportedly found in twelve different compounds in recent years. However, the experimental evidence on which these claims are based has recently been called into question. Here we discuss the measurement of trapped magnetic flux, that should establish definitively that these materials are indeed high temperature superconductors. Its absence would confirm claims to the contrary.

I. INTRODUCTION

The paper “Conventional superconductivity at 203 kelvin at high pressures in the sulfur hydride system,” published in 2015 [1], launched a revolution in the field of superconductivity. Following in its footsteps [1, 2], eleven other hydrogen-rich compounds under high pressure have been found to be high temperature conventional superconductors based on experimental evidence: $PH_x$ above 100 K [3], $LaH_x$ at 250 K [4, 5], above 260 K [6] and above 550 K [7], $YH_x$ at 243 K [8–10], $ThH_x$ at 161 K [11], $PrH_x$ at 9 K [12], $LaYH_x$ at 253 K [13], $CSH$ at room temperature [14], $CeH_x$ above 120 K [15], $SnH_x$ at 70 K [16], $BoH_x$ around 20 K [17], and $CaH_x$ around 215 K [18, 19]. Many more such materials have been determined to be conventional high temperature superconductors based on theoretical evidence [20]. As a consequence, a tectonic shift has taken hold in the field of superconductivity during the last five years [21–27]: (1) no longer are the highest temperature superconductors expected to be found among so-called “unconventional superconductors” [28], as had been the case at least since the discovery of the cuprates in 1986 [29], and (2) no longer is the conventional theory of superconductivity regarded to be unhelpful in the search for new superconducting materials [30]. Quite the contrary, materials theory of the high pressure hydrides has been the driving force and guiding light in the long and finally successful quest to find superconductivity close to and even at room temperature [14].

Despite this overwhelming evidence, remaining gadflies in the ointment are that (a) some of the experimental evidence that shows that these materials are high temperature superconductors has recently been called into question [31–37], and (b) the internal consistency of the conventional theory of superconductivity as well as its ability to explain the most fundamental property of superconductors, the Meissner effect, have recently been called into question [38, 39]. In this paper we propose that it should be straightforward to present incontrovertible evidence of the reality of conventional superconductivity in high pressure hydrides, if indeed it is real, thus dispensing with both of these criticisms in one fell swoop.

II. THE CONCEPT

No experimental evidence for magnetic flux expulsion, i.e. the Meissner effect, the hallmark of superconductivity, has been reported so far for any hydride, rather the opposite [40]. But, it is a fact that there are some bona-fide superconductors that do not expel any magnetic flux [41]. Instead, we propose that the experimental observation that would definitively prove the existence of superconductivity in these materials is flux trapping. Not all superconductors will trap magnetic flux. However, we show in this paper that if the one experiment [42] that was claimed to “unequivocally confirming the existence of superconductivity” [43] in a hydride was indeed valid, it follows that hydrides should trap very significant magnetic flux, easily detectable experimentally even under the very challenging experimental conditions of high pressure experiments.

When a superconductor traps flux, it exhibits a magnetic moment in the absence of an externally applied magnetic field. That magnetic moment is essentially time independent. It originates in electric current flowing in the material that does not decay with time: therefore, this current must be a supercurrent, that only superconductors can carry. Thus, the measurement of trapped magnetic flux in a material suspected to be a superconductor establishes that the material is indeed a superconductor. For example, as discussed by K. A. Müller and coworkers, in early experiments on high $T_c$ cuprates the samples were cooled in a field which was then switched off and a remnant magnetization was observed, and the authors concluded that “This remanent magnetization results from flux trapping, and it is a proof of superconductivity in its own right.” [44]

Not all superconductors trap magnetic flux. Those that do are type II superconductors and in addition are “hard superconductors”, which means there is a strong pinning force that prevents magnetic vortices from moving, which would dissipate energy and lead to supercurrent and magnetic field decay. Many conventional superconductors are hard superconductors, as are many unconventional superconductors such as cuprates and pnictides. They can trap magnetic fields that are much larger than the lower critical field $H_{c1}$ which is typically under
100 Oe. For example, \( YBCO \) can trap magnetic fields in excess of 17 T at 29 K [45], and of more than 2 T at 77 K [46]; \( Nb_{0.7}Ti_{0.3} \) traps fields of up to 0.4 T at 4.2 K [47]; \( GdBa_{2}Cu_{3}O_{7-\delta} \) traps fields in excess of 3 T at 50 K [48]; \( SmBaCuO \) traps fields of more than 8 T at 40 K [49]; \( MgB_{2} \) traps fields of up to 3 T at 5 K [50]; \( Ba_{0.6}K_{0.4}Fe_{2}As_{2} \) traps fields of up to 1 T at 5 K [51].

A counterpart of flux trapping is that the superconductor will effectively screen large externally applied magnetic fields, fields that are much larger than the lower critical field \( H_{c1} \) at the given temperature. Both the ability to trap magnetic flux and to screen external magnetic fields follow from the existence of a large critical current, which in turn depends on the existence of strong pinning centers. Therefore, a superconducting material will be able to trap large magnetic fields if and only if it can screen large externally applied magnetic fields.

The ability of sulfur hydride (\( H_{3}S \)) under high pressure to screen a large externally applied magnetic field was established in a seminal nuclear resonant scattering (NRS) experiment by Troyan et al. [42]. An applied magnetic field of 0.68 T was completely excluded from the interior of a thin superconducting disk for temperatures below 50 K, and some screening occurred even up to temperatures of 120 K. This indicates that (if the experiment was valid) the material will trap flux in the same range of temperature, of magnitude similar to the flux that is excluded when a magnetic field is applied. The resulting magnetic moment of the sample should be easily detectable.

In the following two sections we discuss the experiment in quantitative detail for sulfur hydride, and in the last section we summarize our conclusions. Since all high temperature superconducting hydrides are expected to be governed by the same physics, our analysis should apply to any of the 12 high pressure superconducting hydrides so far discovered and any more to come.

### III. THE NRS EXPERIMENT

Figure 1 shows the geometry of the NRS experiment [42]. A magnetic field of magnitude \( H_{ext} = 0.68 \) T was applied to an \( H_{3}S \) sample pressurized in a diamond anvil cell (DAC), and the experiment established that the magnetic field did not penetrate in the region occupied by a Sn-119 foil immersed in the sample. Figure 1 shows two scenarios under which this can (in principle) be explained.

In the first scenario ((a), top panel), the system is in the Meissner state, and the current that screens the magnetic field in the interior flows in a region within the London penetration depth of the surface. In Ref. [35] we showed that this is possible only if \( H_{3}S \) is a ‘nonstandard superconductor’ [31, 33–35], with properties markedly different from those of standard superconductors; in particular both a much higher critical current and a much higher lower critical field \( H_{c1} \) compared to those of standard superconductors are required to explain the data. However, given that theory has convincingly shown [20–25, 27] that hydride superconductors are described by conventional BCS-Eliashberg theory that describes standard superconductors [53], the scenario (a) has to be discarded as implausible. That leaves scenario (b) as the only alternative.

In scenario (b), the lower critical field \( H_{c1} \) is much smaller than the applied field of 0.68 T, as would be the case in any standard superconductor where \( H_{c1} \) is at most a few hundred Oe. Therefore, the magnetic field penetrates the system in the mixed state in an external annulus of radial thickness up to approximately 5 \( \mu m \) as shown in Fig. 1(b). There are Abrikosov vortices in the annulus and a supercurrent flows that screens the magnetic field in the interior region of diameter 20 \( \mu m \) where the Sn foil resides, that thus remains completely field-free in the temperature range 5 K to 50 K, as established by the experiment [42].

The state depicted in Fig. 1(b) is not a thermodynamic equilibrium state. The supercurrent that flows exerts a radial inward force on the vortices, that should cause them to drift inward to the lower energy equilibrium state where there is a uniform distribution of vortices throughout the superconductor [35]. The fact that this doesn’t happen shows conclusively that there is a large pinning force that prevents the vortices from moving, so
that the system remains in this non-equilibrium state at least through the duration of the experiment. The measurements performed in Ref. [42] were measured during a ~ 25 min interval for each temperature, and at least six different temperatures were measured that showed zero magnetic field in the Sn foil in the temperature range 5 K to 50 K. This establishes that the non-equilibrium state shown in Fig. 1(b)) is stable over a period of at least two and a half hours (and probably much longer) in that temperature range.

We will assume in what follows that the Bean critical state model [52–54], widely used to describe hard superconductors, is appropriate to describe the physics of this system. However, even if there were some deviations from the Bean model we argue that this would not alter our conclusions.

Within the Bean model, the system shown in Fig. 1(b) is in the so-called ‘critical state’. The magnitude of the supercurrent density that flows (the current density is assumed to be independent of radius) is the critical current density of this superconductor at the given temperature, which is determined by the strength of the pinning force. The thickness of the region over which supercurrent flows and vortices exist (5 μm in Fig. 1(b)) is determined by the condition that the critical current flowing in that region nullifies the magnetic field in the interior region where no supercurrent flows.

The experiment shown in Fig. 1 [42] was performed under zero field cooling conditions. The system was cooled to a temperature of 5 K, then the magnetic field was applied, then the NRS measurements were performed at increasing temperature. If instead the system is cooled in a magnetic field, the magnetic field should remain in the interior and vortices will be uniformly distributed in the system. If the external magnetic field is then removed, the same pinning forces that prevented the flux from penetrating in Fig. 1 will prevent the flux from leaving the sample: the flux will remain trapped, as shown schematically in Fig. 2. We discuss this quantitatively in the following section.

**IV. TRAPPED FLUX**

When the system is cooled in a large applied magnetic field the magnetic field should remain in the interior of the sulfur hydride rather than being expelled. This is always true for type II superconductors cooled in fields larger than the lower critical field $H_{c1}$. For sulfur hydride, this is true even for applied magnetic fields as small as 20 Oe, as was demonstrated in Ref. [1], Fig. 4(a). If the external field is then removed, the field will remain trapped over essentially the entire volume of the superconductor in the presence of strong pinning centers, as depicted in Fig. 2.

According to the Bean model, the current flowing is the critical current $J_c$. The maximum external field $H_m$ that can be completely screened out from the center of a long cylinder of radius $r_0$ is given by

$$H_m = \frac{4\pi}{c r_0} J_c$$

where $J_c$ is the critical current density. For the situation of interest here, shown in Fig. 3(a), the applied magnetic field, $H_{ext}$, is less than $H_m$, so using Eq. (1), and neglecting demagnetization, the critical current is given by

$$J_c = \frac{c}{4\pi d} H_{ext}$$

with $H_{ext} = 0.68$ T the applied magnetic field and $d = 5$ μm the width of the outer annulus, as depicted in Fig. 3(a). This yields for the critical current density

$$J_c = 1.08 \times 10^7 Amp/cm^2.$$ 

This estimate agrees with the estimated critical current at low temperatures given in Ref. [1].

The same model then predicts that if we field-cool the system in an applied magnetic field of 0.68 T and subsequently remove the external field, the applied field will remain trapped over a region of at least 20 μm in diameter, as shown in Fig. 3(b), with the same critical current $J_c$ flowing in the outer layer. This trapped field should remain over a time period of the same order as the time period over which the system can keep the applied field out in the zero-field-cooled case, which as discussed above was established to be greater than 2.5 hours in the NRS experiment.

**FIG. 2:** Field configuration expected for field cooling and then removing the applied magnetic field.

**FIG. 3:** (a) Exclusion of external magnetic field $H_a = 0.68$ T from the interior of the sample in the NRS experiment [42], corresponding to the situation shown in Fig. 1(b). (b) Flux trapping resulting when the system is cooled in the presence of an external field $H_r = 0.68$ T and then the external field is removed, corresponding to the situation shown in Fig. 2. The figure assumes no demagnetization, which is exact for a long cylinder.
Let us now estimate the magnetic moment that results from this trapped field. We can use the measured magnetic moment in sulfur hydride reported in Ref. [1] for an applied field of 20 Oe, shown in their Fig. 4(a). The ZFC curve shows that the system goes from diamagnetic to paramagnetic when the transition occurs around 200 K, and the change in magnetic moment of the sample is approximately

\[ m \sim 10^{-6} \text{emu}, \quad (4) \]

as given by the difference in ZFC magnetic moment above and below \( T_c \) in Fig. 4(a) of Ref. [1]. The positive signal above \( T_c \) seen in Fig. 4(a) of Ref. [1] results from a paramagnetic background contribution that presumably does not change when the sample goes through \( T_c \).

Therefore, these data tell us that a magnetic field of 20 Oe gives rise to a magnetic moment \( m \) in the sample that cancels the magnetic field throughout the interior (except within a London penetration depth of the surface). This implies that if a magnetic field of 0.68 T is trapped throughout the volume of the sample, it should generate a magnetic moment approximately \( 10^{-6} \times (6800/20) \text{ emu} = 3.4 \times 10^{-4} \text{ emu} \). Finally, we have to correct for the fact that in the outer layer of thickness 5 \( \mu m \) the magnetic field decays as shown in Fig. 3(b). This gives then for the estimated trapped magnetic moment due to the trapped field:

\[ m_{tr} \sim 3.4 \times 10^{-4} \times \left( \frac{12.5}{15} \right)^3 \text{ emu} = 2.0 \times 10^{-4} \text{ emu} \quad (5) \]

The trapped magnetic moment versus temperature is shown in Fig. 4, right panel. The left panel reproduces the measured magnetic field in the interior of \( H_3S \) under zero field cooling [42].

The sensitivity of the in-situ SQUID magnetometer used in Ref. [1] to obtain the data shown in their Fig. 4(a) was of the order \( 10^{-8} \) emu. The expected magnetic moment resulting from trapped flux after application of a 0.68 T magnetic field, Eq. (5), is more than four orders of magnitude larger. Thus it is trivial to detect this trapped moment using that equipment, or far less sophisticated equipment.

Of course the presence of a background magnetic moment due to the DAC should also be considered. However, that contribution will not disappear above \( T_c \), as the contribution from the trapped magnetic moment resulting from supercurrents does. It can also be measured separately by performing the experiment without the sample, and subtracted off when the measurement with the sample is performed.

V. DISCUSSION

The magnetic moment resulting from trapped flux Eq. (5), shown in Fig. 4, right panel, should in fact be a lower bound, and the real one may well be higher. This is because the NRS experiment only informed us that the Sn foil remained magnetic-field free, but did not inform us whether there was also a magnetic field-free region outside of it, which would be the case if the critical current was larger than Eq. (3). If so, the trapped magnetic moment for applied field 0.68 T should be correspondingly higher. It should also be possible to estimate the maximum value of magnetic moment that can be trapped by this system for larger applied fields by repeating the NRS experiment with larger fields and finding the maximum external field that keeps the Sn film field-free at a given temperature.

In fact, it would seem that it should have been straightforward to repeat the NRS experiment [42] by field-cooling the sample in a field of 0.68T down to low temperatures, removing the external field, and then obtaining the NRS spectra. Observation of quantum beats under those conditions would have provided compelling proof that the magnetic field had remained trapped in the superconducting sample. We urge that this test be done when the NRS experiment is repeated to check its reproducibility.

We also point out that the measurements of magnetization versus magnetic field reported in ref. [1] are inconsistent with the NRS measurements [42]. Fig. 4c of [1] shows a paramagnetic response for fields larger than 0.05T at \( T = 50 K \). This is clearly in contradiction with the observations reported in Ref. [42] that a magnetic field of 0.68T was excluded from at least 67% of the sample volume (figs. 4A and S6 of Ref. [42] at the same \( T = 50K \)). The caption of Fig. 4c in [1] reads “At higher fields, magnetization increases due to the penetration of magnetic vortexes”. Instead, the field applied (up to 0.2T) should have remained largely excluded according to the results in [42]. To resolve this contradiction, one would have to assume that the \( H_3S \) samples used in refs. [1] and [42] were qualitatively different.

In this paper we have neglected demagnetization effects resulting from the finite aspect ratio of the samples. They will not qualitatively affect our conclusions, as can be inferred for example from the measurements and cal-
calculations in Refs. [47, 54].

The measurement described here is much simpler and requires much less sensitivity than the far more sophisticated measurements performed in Refs. [1] and [42] that claimed to establish that sulfur hydride is superconducting below 203 K. Detection of a large magnetic moment resulting from trapped flux that persists over time will confirm that these materials are indeed high temperature superconductors, and is an unavoidable consequence of the physics of these superconductors revealed by the NRS experiment [42], namely that they are hard superconductors with strong pinning forces.

Of course, if for some reason the NRS experiment as performed was flawed, as was suggested as a possibility in Ref. [35], on one hand it would not provide evidence that $H_3S$ is a superconductor, but on the other hand we would also not know for a fact that if these materials are superconductors, then they are hard superconductors, leaving open the possibility that they could be soft superconductors that do not trap magnetic flux. If so, we still suggest it would be revealing to check for the existence of trapped flux in a non-simply connected geometry, by introducing in the center of the sample a non-superconducting material so that the superconducting material would form a ring around it. In that case, quantized flux should certainly be trapped in the non-superconducting material as long as the magnetic field is smaller than $H_{c1}$.

Detecting trapped magnetic flux in the absence of applied magnetic field that persists over an extended time period will establish that these materials are superconductors, by proving that supercurrents that do not decay due to resistance indeed flow in these materials. And, if these materials are indeed superconductors, there can be no doubt that they are conventional superconductors, since they owe their very existence to the conventional theory of superconductivity [56, 57].

On the other hand, failure to detect magnetic moments resulting from trapped flux will establish that either (a) these materials are nonstandard superconductors [33–35], with properties qualitatively different from standard superconductors, that additionally don’t trap magnetic flux neither in simply connected nor in multiply connected geometries, as all other known superconductors do; or (b), far more likely, that these materials are not superconductors [31, 32, 34, 36]. Either of the two possibilities will call the ability of the conventional theory of superconductivity to describe real superconductivity of real materials into question [58].

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