Further analysis of flux trapping experiments on hydrides under high pressure

J. E. Hirsch[a] and F. Marsiglio[b]
[a] *Department of Physics, University of California, San Diego, La Jolla, CA 92093-0319*
[b] *Department of Physics, University of Alberta, Edmonton, Alberta, Canada T6G 2E1*

It has been claimed that measurements of trapped magnetic flux in hydrides under pressure provide evidence for superconductivity in these materials [1, 2]. In recent work we have questioned that claim [3]. Here we point out that recent experiments [4] provide further evidence supporting our questioning of that claim. We also present calculations for a thin disk and compare these with those previously calculated for a long cylinder. The results are qualitatively similar, and therefore reinforce our previous conclusions. We also address recent criticism of our paper Ref. [3] by Bud'ko [5] and by Talantsev et al [6].

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I. INTRODUCTION

In a recent perspective, M. I. Eremets claims [7] that experiments on trapped flux reported in Refs. [1, 2] provide "conclusive evidence" of superconductivity in hydrides under pressure. Instead, in Ref. [3] we argued that the evidence presented in Ref. [1] was not consistent with superconductivity. In this paper we show that our arguments presented in Ref. [3] are further strengthened by experimental results on trapped flux for materials known to be superconductors presented in Ref. [4]. We also address recent criticisms of our paper Ref. [3] by Bud'ko [5] and by Talantsev et al [6].

Essentially all the results presented in Ref. [1] were under the ZFC protocol. Ref. [1] claimed that their experimental results were fitted by the Bean model and lent strong support to the claim [8] that hydrides under pressure are high temperature superconductors. However, in Ref. [3] we pointed out that the theoretical fit based on the Bean model presented in Ref. [1] was incorrect, and that in fact a correct theoretical treatment leads to the conclusion that the experimental data are inconsistent with what is expected for a superconductor. The crucial point we made in Ref. [3] is that the trapped moment for low field should increase *quadratically* with field in the ZFC protocol and *linearly* with field in the FC protocol. The experimental results in Ref. [1] under the ZFC protocol increase linearly with field as shown in Fig. 1, hence are inconsistent with the expected behavior of superconductors.

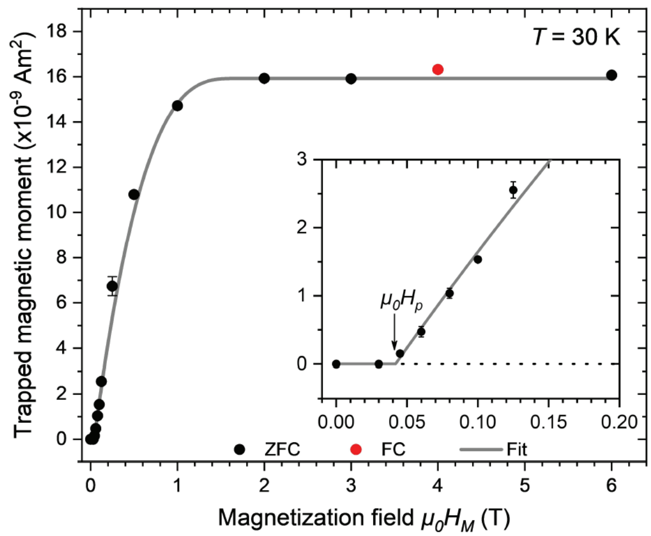

FIG. 1: Trapped magnetic moment for H_3S at 30K, reproduced from Fig. 2a of Ref. [1]. Except for one point (in red), all the data were obtained under the ZFC protocol [1]. The points are experimental data from Ref. [1], the lines are theoretical fits to the data reported in Ref. [1] obtained from Eqs. (1)-(3) of Ref. [1].

We pointed out in Ref. [3] that the theoretical fit that had been performed in Ref. [1] giving a linear behavior that matched the data (Eqs. (1)-(3) of Ref. [1]), as seen in the inset of Fig. 1, resulted from an incorrect application of the Bean model: the authors of Ref. [1] had used the equations appropriate for a FC protocol (their Eqs. (1)-(3)) and a threshold field $H_p = 0.042T$ to match their experimental ZFC data. In Figs. 3 and 4 of our paper Ref. [3] we showed what a correct application of the theory yields: the ZFC results are in drastic disagreement with the experimental data. We also showed (Fig. 4 of Ref. [3]) that using the equations appropriate to the FC protocol, with a finite value of H_p which is inconsistent with the FC protocol, the experimental results, which are linear in field, are matched. That is what was done by the authors of Ref. [1], depicted in Fig. 1 here as "Fit". Such a treatment is of course both wrong and internally inconsistent for fitting ZFC measurements.

In the published version of Ref. [1], namely Ref. [2], that was submitted for publication 21 September 2022, the incorrect theoretical analysis of the preprint version Ref. [1] (its Eqs. (1)-(3)) is missing, and no attempt is made to fit the measurements theoretically, instead, arbitrary lines are drawn through the points to guide the readers' eyes.

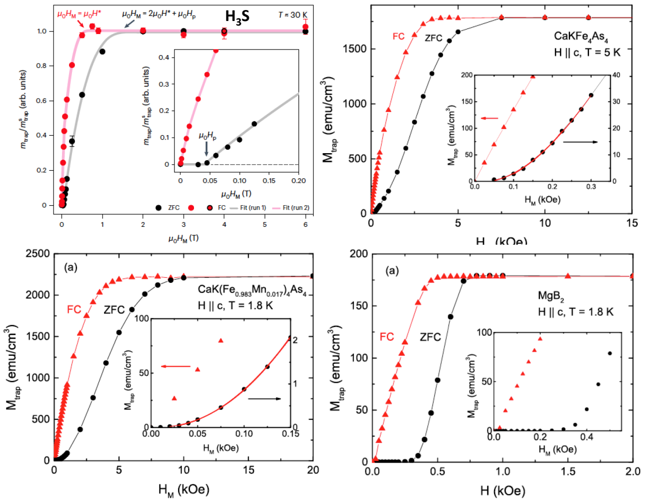

FIG. 2: Magnetic moments measured under FC and ZFC protocols after removal of the applied magnetic field reported in Ref. [4] for three materials known to be superconductors (upper right, lower right, lower left panels) and for H_3S reported in Ref. [2]. The insets show the behavior for low field, the lines are guides to the eyes. The figures were reproduced from Refs. [2] (upper left panel) and [4] (three other panels).

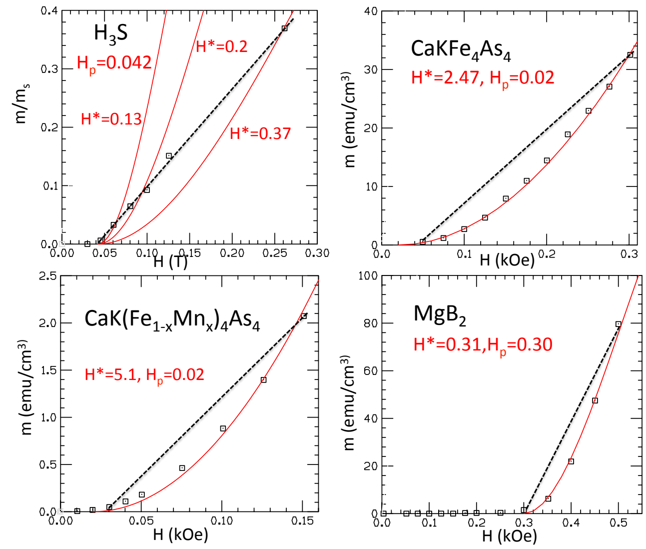

FIG. 3: Low field magnetic moments for the four cases shown in Fig.2. The points were obtain from digitization of the figures showing the measured data in Ref. [4]. The red lines were obtained from our model using the parameters shown in the figure. The dashed lines were drawn from the first point where the moment starts to deviate from zero to the last point in the figure.

II. COMPARISON WITH RECENT EXPERIMENTS

The key point made in our paper Ref. [3] was that the magnetic moment measured under the ZFC protocol should rise quadratically rather than linearly with field if it is indeed a signature of trapped flux resulting from superconducting currents. This is strongly supported by measurements recently published by Bud'ko and coauthors [4]. Figure 2 shows the behavior of trapped moment versus field for both ZFC and FC protocols for three materials known to be superconductors, and for H_3S. It can be seen that under the ZFC protocol the linear behavior of the moment seen for H_3S is qualitatively different from the supralinear behavior seen in the three other examples. In other recent experiments by Bud'ko and coauthors with a sample of $CaKFe_4As_4$ of much smaller dimensions, approaching the dimensions of the hydride samples used in Refs. [1, 2], the dependence of trapped moment on magnetic field for ZFC was also seen to be clearly supralinear [10].

In Fig. 3 we show fits using our equations [3] to the low field behavior of the moment under the ZFC protocol for the four cases shown in Fig. 2. It can be seen that our model, which gives quadratic behavior for low field, fits the three known superconductors very well, but doesn't fit at all the H_3S behavior, no matter what the choice of parameters is. For illustration we have also drawn straight lines connecting the first point where the magnetic moment starts to deviate from zero to the last point in the figures. It can be seen that for the three known superconductors the data deviate strongly from the straight dashed lines, instead for H_3S the data are very well described by the straight dashed line. This establishes the qualititative difference between the behavior of the ZFC moment for H_3S versus that of known superconductors, as was pointed out in Ref. [3].

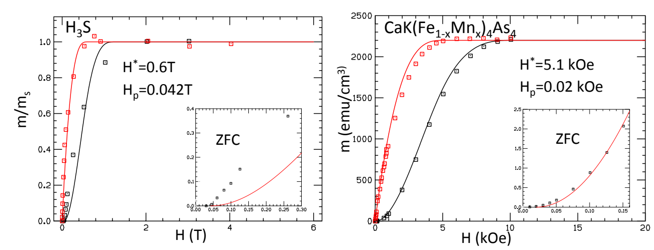

FIG. 4: Comparison of fits for a standard superconductor (right panel) (data from Ref. [4]) and for H_3S (data from Ref. [2]) using our model (black ZFC, red FC). For H_3S we used a value of $H^* = 0.6$ to approximately match the data for FC. It can be seen that it does not match the ZFC data neither at small (inset) nor at large fields. In contrast, for the standard superconductor the data are approximately fitted by the model both for small and large fields, both for FC and ZFC.

In Fig. 4 we show fits of our model for H_3S and for a standard superconductor, $CaK(Fe_{1-x}Mn_x)As_4$, data from Ref. [4], over a wide field range. For H_3S we picked parameters so as to match the FC measurements. It can be seen that the ZFC data for H_3S deviate from the calculated values both for small and large fields. Instead, for the standard superconductor on the right panel our model gives good agreement with the measured values

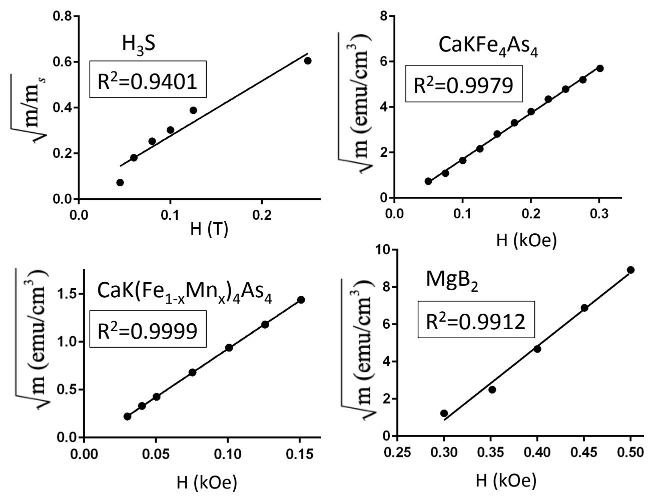

FIG. 5: Linear regression fit of square root of trapped moment versus magnetic field. It can be seen that for the three standard superconductors the fit is very good, indicating quadratic behavior of trapped moment versus field as predicted by our model and general physical arguments. Instead, for H_3S the fit is not good.

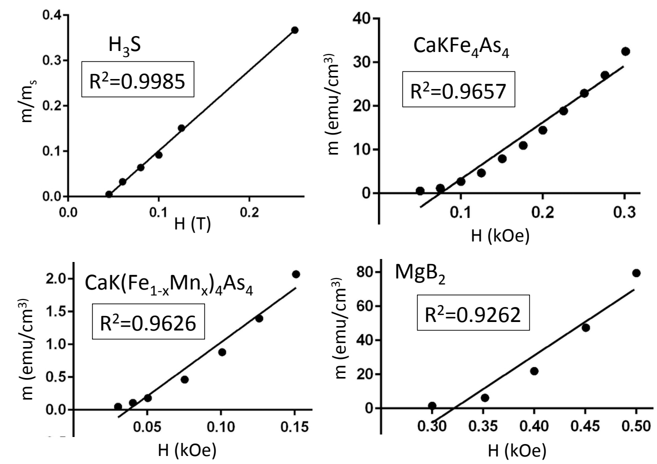

FIG. 6: Linear regression fit of trapped moment versus magnetic field. It can be seen that for the three standard superconductors the fit is not very good, indicating that the trapped moment under ZFC is not linear with magnetic field, as predicted by our model and general physical arguments. Instead, for H_3S the linear fit is excellent.

both for ZFC and FC data over the entire field range.

However it should be pointed out that our model is an approximation to reality, in particular it assumes that the critical current density is independent of field strength. This appears to correspond to the real situation for the material shown on the right panel in Fig. 4, but can fail in other cases. For example, for $CaKFe_4As_4$ our model fits the overall field dependence well with $H^* = 3.5$, somewhat larger than the value used in Fig. 3. This can be accounted for by allowing for variation in the critical current as function of field.

However, the prediction that the behavior of the ZFC moment for low fields is supralinear and approximately quadratic is a robust prediction that should be universally valid. For hard superconductors that trap magnetic flux, the penetration depth upon application of the field is an increasing function of the applied field, and the magnitude of the trapped moment is proportional to both the penetration depth under ZFC and to the maximum field applied, hence necessarily supralinear and approximately quadratic. Instead, in the FC protocol the penetration depth is the radius of the sample independent of the magnitude of the field, hence the trapped moment should be linear in field as observed.

In order to quantify the deviation of the ZFC data for H_3S from the expected behavior of a superconductor, we show in Figs. 5 and 6 linear regression fits [11] to the square root and first power of the trapped moment versus magnetic field respectively for the four cases considered here. The coefficient R^2 measures how well the data fit the regression model, with $R^2 = 1$ being a perfect fit. It can be seen in Fig. 5 that R^2 is larger than 0.99 for the three standard superconductors indicating an excellent fit to quadratic behavior of magnetic moment versus magnetic field, while for H_3S R^2 differs substantially from unity, indicating the observed behavior is not consistent with quadratic dependence of moment on field. Conversely, as seen in Fig. 6, a linear fit of magnetic moment vs field fits the H_3S data extremely well, with $R^2 = 0.9979$, and does substantially worse for the three standard superconductors. This provides strong evidence that for standard superconductors the behavior of magnetic moment versus field under ZFC is quadratic while it is linear for H_3S.

III. DEPENDENCE ON ASPECT RATIO OF THE SAMPLES

The model used to compute the magnetic moment under FC and ZFC conditions assumes tall cylinders where demagnetization effects are accounted for with a demagnetization factor [3]. It is very difficult to treat the most general geometry, but the case of thin disks has also been investigated in the past [12–14], and below we reproduce some of these results to compare with those of a tall cylinder.

For the field-cooled case, we have for an applied field H_M (see Eq. (18) of Ref. [14])

$$\frac{m}{m_s} = \frac{2}{\pi}\left[\cos^{-1}\left(\frac{1}{\cosh(H_M/H^*)}\right) + \frac{\tanh(H_M/H^*)}{\cosh(H_M/H^*)}\right], \quad (1)$$

where $m_s = \pi J_c R^3 h/2$ and $H^* = J_c h/2$, and the disk has thickness h and radius $R \equiv d/2$, with critical current density magnitude J_c.

For the zero-field-cooled case, we have been unable to determine the magnetic moment analytically, Instead, following Ref, [14], we adopt the model for the azimuthal current density ($x \equiv r/R$, $\bar{a} \equiv a/R$, and $\bar{b} \equiv b/R$):

$$\bar{J}_\phi(x) \equiv \frac{J_\phi(x)}{J_c} = -\frac{2}{\pi}\left[\tan^{-1}\left(\frac{c_a x}{\sqrt{\bar{a}^2 - x^2}}\right) + 2\tan^{-1}\left(\frac{c_b x}{\sqrt{\bar{b}^2 - x^2}}\right)\right], \quad \text{for} \quad x < \bar{a}$$
$$= -1 + \frac{4}{\pi}\tan^{-1}\left(\frac{c_b x}{\sqrt{\bar{b}^2 - x^2}}\right) \quad \text{for} \quad \bar{a} < x < \bar{b}$$
$$= +1 \quad \text{for} \quad \bar{b} < x < 1 \tag{2}$$

with $c_a \equiv \sqrt{1-(\bar{a})^2}$ and $c_b \equiv \sqrt{1-(\bar{b})^2}$, and the vortex-free radius is $a = R/\cosh[(H_M - H_p)/H^*]$, and $b = R/\cosh[(H_M - H_p)/(2H^*)]$ is an auxiliary radius, above which the critical current flows. To obtain the magnetic moment, Eq. (2) is integrated numerically for $H_M > H_p$ (it is zero otherwise):

$$\frac{m}{m_s} = 3\int_0^1 dx\, x^2 \bar{J}_\phi(x). \tag{3}$$

A comparison for typical parameters is shown in Fig. (7), and illustrates that the results for the two cases (tall cylinder vs thin disk) are similar. The ZFC result for the thin disk (shown in black) increases even less gradually than the result for the tall cylinder, and requires a higher applied field to achieve full saturation. The inset (a log-log plot of the same quantities for the thin disk case) illustrates that the magnetization increases as $(H_M - H_p)^3$ for applied magnetic fields beyond H_p, reinforcing the physically motivated fact that for the ZFC protocol the magnetization has to increase faster than linearly with applied field, independently of the model used.

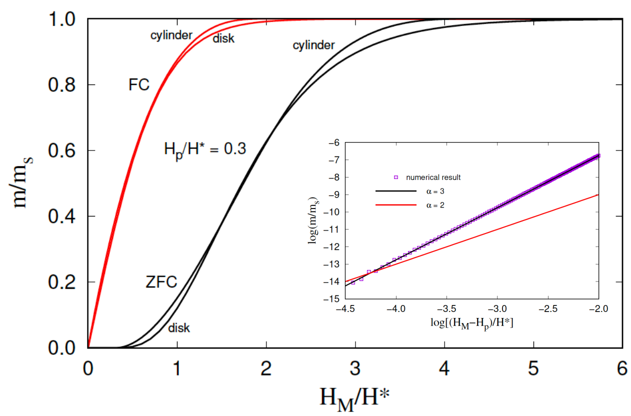

FIG. 7: The normalized magnetization m vs normalized applied field H_M for the two cases, tall cylinder and thin disk, for $H_p/H^* = 0.3$. The main frame illustrates that overall there is little difference between the two cases. The inset shows a log-log plot for the disk case, and shows that the increase in the magnetization beyond H_p is cubic in applied field. The two straight lines drawn in the inset are for $\alpha = 3$ (black) and $\alpha = 2$ (red) where $m \propto (H_M - H_p)^\alpha$, and the numerical results clearly agree with $\alpha = 3$. As determined in Ref. [3], the increase in the magnetization for the tall cylinder case is quadratic in field. In general we expect a supralinear dependence somewhere between these two cases.

IV. RESPONSE TO BUD'KO'S COMMENT

In Ref. [5] commenting on our paper Ref. [3], Bud'ko claims that our analysis of FC data in Ref. [3] is *"unphysical"* and *"incorrect"*, because for the FC protocol there should be no threshold field H_p, contrary to what our figures 3, 4 and 5 on Ref. [3] show. Of course it is correct that H_p should be zero for FC. The reason we included a finite H_p in our figures in Ref. [3] for FC was to show that under that inconsistent assumption the theoretical expression matches the measured results for ZFC reported in Ref. [1]. But we made very clear in our paper [3] that under the FC protocol the threshold field H_p should be zero, which is the whole point of Bud'ko's Comment [5]. Namely, right after Eq. (4) in Ref. [3] we stated that for the FC protocol Eq. (4) is valid *"with $H_p = 0$"*. After Eq. (7) in [3] we stated *"Figure 3 shows what these expressions predict for the trapped magnetic moment versus magnetization field H_M for the parameters assumed [37] in Ref. [1]"*, and Ref. [37] cited there reads *"In this and the following figures we have assumed the value of H_p from Ref. [1] for ZFC for our FC calculations. However, H_p should be zero for FC."* Thus, there is no error nor *"unphysical"* nor *"incorrect"* statements in our paper, contrary to what Bud'ko states in his Comment [5].

V. RESPONSE TO TALANTSEV ET AL'S COMMENT

In Ref. [6] commenting on our paper Ref. [3], its authors Talantsev, Minkov, Ksenofontov, Bud'ko and Eremets claim that:

(i) Our paper relies on *"the wrong model"*.

(ii) Our paper uses *"selective manipulations *(hide/delete) of calculated datasets"*.

(iii) Our paper *"ignores the reference measurements after the release of pressure"*.

(iv) Our paper *"conducted simulations where all free parameters were fixed"*.

(v) The approach of our paper *"also implies that MgB_2 is not a superconductor"*.

and as a consequence does not provide robust evidence against superconductivity in H_3S. We address these points in what follows. We note that Talantsev et al. fail to address the key point of paper Ref. [3], namely that the linear behavior of ZFC moment versus field reported in Refs. [1, 2] is incompatible with superconductivity.

A. Does our paper rely on the wrong model?

It is unclear why our model is called *"the wrong model"* in Ref. [6]. Our model used here and in Ref. [3] is a straightforward application of Bean's model with the assumption of constant critical current and ignoring corrections for demagnetization. In the previous section we have shown that such corrections will not substantially change the results. The trapped moment under the FC protocol under an applied field H_M is given by

$$m = \int_r^{d/2} \pi r'^2 j_c h dr' = m_s[1 - (\frac{r}{d/2})^3] \quad (4)$$

for a cylindrical sample of diameter d and uniform critical current j_c flowing in the region $r < r' < d/2$, with

$$r = r(H_M) = \frac{d}{2}(1 - \frac{H_M}{H^*}). \quad (5)$$

for $H_M < H^*$, $r = 0$ for $H_M > H^*$. For a long cylinder, $H^* = (2\pi/c)dJ_c$. Instead, under the ZFC protocol the trapped moment after application of a field H_M and subsequent removal is given by

$$m = m_s[1 - 2(\frac{r_1}{d/2})^3 + (\frac{r_2}{d/2})^3] \quad (6)$$

where, for $H_M < H^* + H_p$

$$r_1 = \frac{d}{2}(1 - \frac{H_M - H_p}{2H^*}) \quad (7a)$$

$$r_2 = \frac{d}{2}(1 - \frac{H_M - H_p}{H^*}). \quad (7b)$$

For $H^* + H_p < H_M < 2H^* + H_p$, r_1 is given by Eq. (7a) and $r_2 = 0$, and for $H_M > 2H^* + H_p$, $r_1 = r_2 = 0$. This results from J_c flowing in one direction in the region $r_1 < r' < r_2$ and in the opposite direction in the region $r_2 < r' < d/2$, as illustrated in Fig. 2 of Ref. [3]. It can be easily seen that Eqs. (6), (7) lead to $m \propto (H_M - H_p)^2$ for small $(H_M - H_p)$ (always positive of course! [15]) and Eqs. (4), (5) lead to $m \propto H$ for small H.

B. Does our paper selectively hide and manipulate data sets?

A detailed response disproving this claim is given in Ref. [15].

C. Does our paper ignore the reference measurements after the release of pressure?

In Refs. [1, 2] it is stated that after a diamond cracked and pressure dropped below 10GPa, no trapped moment was detected in the experiment. In our paper we hypothesized that the measured moments, which were all measured with uncracked diamonds and under much higher pressures, originated from *"magnetic properties of the sample or its environment unrelated to superconductivity"*. The authors of [6] object to the fact that we did not consider the measurements with cracked diamond and low pressure relevant. We do not, because both the properties of the sample and its environment will change when diamonds crack and the pressure drops, so we don't find it surprising nor relevant to the understanding that no moments were measured under those very different experimental conditions.

The authors should perform control experiments, under the same experimental conditions for which moments were detected (in particular high pressures and uncracked diamonds), using samples known not to be superconducting, as well as samples known to be superconducting, to provide relevant information to support or refute their claims [2, 7] that their trapped flux experiments shed light on the question whether the hydrides are superconducting or not.

D. Does our paper fail to deduce the penetration field and other parameters through standard data fitting?

We are unaware of what the authors mean by *"standard data fitting"*. In our calculations both in Ref. [3] and in this paper we varied the parameters to obtain good fits to the data. Our model results vary smoothly with the parameters and each parameter influences the resulting curve in a distinct way, so it is simple to find parameters that will best represent the measured data. As an example consider the three red curves shown in the upper left panel of Fig. 3. It is clear that for other values of H^* the curves will interpolate between the ones given and that no value of H^* will give a good fit to the data.

E. Does the approach of our paper imply that MgB_2 is not a superconductor?

Our model does not imply that MgB_2 is not a superconductor. As discussed above, our model has simplifying assumptions, in particular that the critical current is independent of field. Within that approximation we cannot fit both the FC and ZFC reported data for MgB_2 with the same parameters, but need different parameters in the model. A more elaborate model allowing for non-constant critical current can presumably fix that.

VI. TIME DEPENDENCE

A characteristic feature of magnetic moments that originate in trapped flux in superconductors is that the moment decays with time because of thermally activated motion of vortices, so-called flux creep, with the decay rate increasing as the temperature approaches T_c. Fig. 8 shows the reported behavior of the trapped moment versus time for H_3S, compared with the behavior seen for several standard superconductors, both conventional and unconventional. It can be seen that for H_3S the trapped moment shows essentially no time dependence within error bars [20]. The decay rate $S = (1/m_{trap})(dm_{trap}/dlnt)$ estimated by the authors of Ref. [2] is given in Fig. 8 upper left panel for two values of the temperature, $T = 185K$ and $T = 180K$ as $S = 0.005$ and $S = 0.002$ respectively. For the lower temperature $T = 165K$ the value of S was not given, however the authors included a dashed blue line indicating the rate of decay. We added next to that line a solid blue line showing the rate of decay for 180K, namely S=0.002. It can be seen that the moment decays faster for 165K than for 180K, which is contrary to what is expected. The fact that there is no evidence in these data that the decay rate increases as T approaches T_c, in fact there is no clear evidence that the moment decays at all, is contrary to what is observed in standard superconductors and suggests that the measured moments do not originate in superconducting currents. This was also pointed out in Ref. [20].

VII. DISCUSSION

In the year 2021, we proposed [21] that flux trapping experiments on hydride materials under high pressure claimed to be high temperature superconductors [22] should be performed in order to test those claims, since it was already clear at the time that the only possibility was that they are 'hard' superconductors with strong pinning centers. Minkov et al followed up on our suggestion [1, 2], but ignored the fact that we had added an important caveat in our paper [21] : *"Of course the presence of a background magnetic moment due to the DAC should also be considered...It can also be measured separately by performing the experiment without the sample, and subtracted off when the measurement with the sample is performed."* That control experiment has not been performed according to Refs. [1, 2], leaving open the possibility that the measured "trapped flux" is a response of the experimental apparatus unrelated to superconductivity. That the sample itself is ferromagnetic and therefore gives rise to trapped flux also cannot be ruled out by the reported experiments [1, 2], and would be consistent with the observed linear behavior of trapped moment versus field under ZFC.

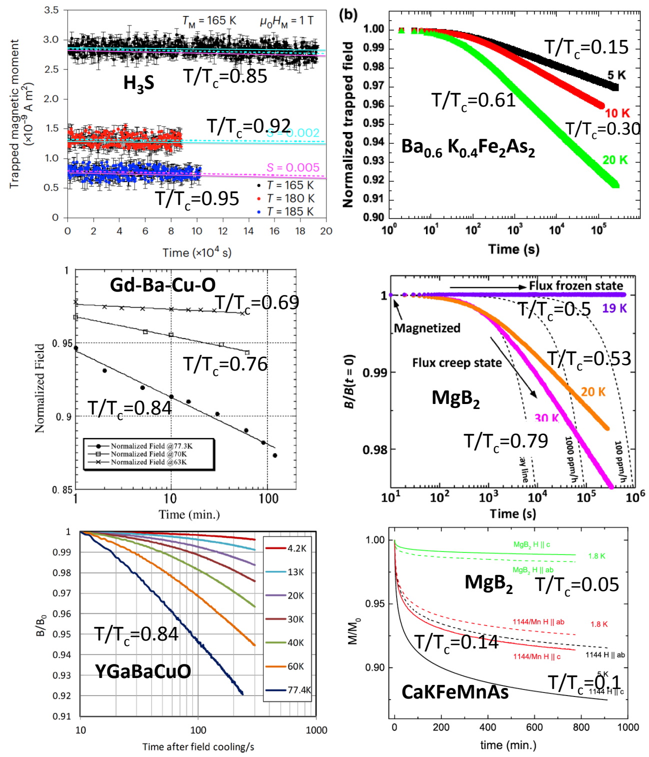

FIG. 8: Decay of trapped moment versus time for H_3S (upper left panel) from Ref. [2], and for various standard superconductors (other panels), from Refs. [5, 16–19] for lower right, upper right, middle left, middle right, lower left panels respectively. For the lower right panel see detailed figure caption for Fig. A9 in Ref. [4]. It depicts the decay for the three materials shown in Fig. 2, all for temperatures much lower than their critical temperatures.

It is interesting to note that a main focus of Ref. [4] was to validate the results presented in Refs. [1, 2] claiming to observe trapped flux in hydrides under pressure, by showing similarity of those results with results for standard superconductors. In fact, we have argued here that Ref. [4] instead provided additional evidence to question that claim. In addition to the anomalous behavior of the moment under ZFC, another notable difference is the decay of the magnetic moment with time shown in Fig. 8. All the materials investigated by Bud'ko et al in Ref. [4] showed clear evidence for flux creep even at temperatures much lower than the critical temperatures, as seen in their Fig. A9, lower right panel of Fig. 8 here. It is strange that the authors didn't report the time evolution of the moments in their standard superconductors at temperatures close to their critical temperatures. Presumably a much faster decay than that shown in the lower right panel of Fig. 8 would be found, as indicated by the temperature evolution in the other panels in Fig. 8, and in particular a *much* faster decay than shown in the upper left panel of Fig. 8, that would have highlighted the clearly anomalous nature of the flux creep behavior, more precisely absence of evidence for it [20], shown by the data reported by Minkov et al for hydrides [1, 2].

In summary, in this paper we reiterate with further evidence that the observed linear behavior of trapped moment versus field under the ZFC protocol reported in Refs. [1, 2] is inconsistent with the inference made in those references that it originates in superconducting currents. Given that the hydrides do not expel magnetic field under FC [23], the only possible explanation for that fact consistent with superconductivity is that they are very hard superconductors with very strong pinning centers. Those pinning centers should inhibit the applied magnetic field under ZFC from penetrating, hence limit the amount of field that can remain trapped when the applied field is removed (if the field can't penetrate it obviously cannot get trapped), hence give rise to non-linear (quadratic or higher power) rather than linear behavior of trapped moment vs field, contrary to what was reported in Refs. [1, 2]. The reported linear behavior of moment vs field in the ZFC protocol is incompatible with superconductivity, thus providing clear evidence against superconductivity in hydrides under pressure, as first pointed out in Ref. [3]. The absence of decay of magnetization with time at temperatures close to the critical temperature provides further evidence that the so-called trapped moments do not originate in superconducting currents.

In this paper we have also corrected the misleading claims about our earlier paper on this topic [3] made in Refs. [5] and [6].

Acknowledgments

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