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COMMENT

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Comment

Comment on "Room-temperature superconductivity in a carbonaceous sulfur hydride" by Elliot Snider et al.

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Introduction. – On October 14, 2020, Snider *et al.* reported the discovery of the first room temperature superconductor, carbonaceous sulfur hydride, hereafter called CSH [1]. If this is true, it represents a major scientific breakthrough. Many researchers throughout the world have been devoting intensive research efforts and resources to this topic for the last 14 months under the assumption that the result is correct. To date the result has not been independently reproduced. In this paper we show that ac magnetic susceptibility results reported in [1] do not seem supported by valid underlying data. This calls the conclusion of ref. [1] that the material is a superconductor into question.

The findings of sharp drops in the measured ac magnetic susceptibility as a function of temperature was claimed in ref. [1] to be "a superior test of superconductivity", demonstrating the existence of superconducting transitions. The susceptibility data reported in [1] were obtained from the subtraction of two independent measurements, namely "raw data" and "background signal", according to the equation

$$\text{data} = \text{raw data} - \text{background signal}. \tag{1}$$

According to the caption of fig. 2(a) of [1], "*The background signal, determined from a non-superconducting C-S-H sample at 108 GPa, has been subtracted from the data*". However, neither of these independent measurements (raw data and background signal) were given in the paper [1] nor in supplemental material for the six pressures for which results were published.

More than a year later, in a paper posted on arXiv in December 2021 [2], two of the authors of ref. [1] reported the measured raw data and the numerical values of the data for the six curves of susceptibility data published in ref. [1]. Here we analyze this information and its relationship with the published data in ref. [1]. We find that there is an unexpected disconnect between the raw data and the data published in ref. [1]. Some partial results were reported earlier in refs. [3,4].

Figure 1 shows the susceptibility data for the six pressure values for which susceptibility data were given in ref. [1], termed "Superconducting Signal" in ref. [2]. Figure 2 shows the raw data for the six pressure values, termed "Measured voltage" in ref. [2]. The sharp drops in the curves as the temperature is lowered are interpreted to signal superconducting transitions [1,2].

It should be pointed out that the top left panel of fig. 1, for 138 GPa, was reported in ref. [1] as "raw data", however it is reported as "Superconducting Signal", *i.e.*, "data", in ref. [2]. It is notable that the results for 138 GPa are *qualitatively* different from all the other cases: for temperatures below the drop, the susceptibility rises sharply, while it is flat in all the other cases. No explanation is given in ref. [2] for this fact, nor for why the results for 138 GPa were reported in ref. [1] as "raw data" when in fact they are "data" obtained after subtracting a "background signal" from the measured raw data, nor for why that particularly anomalous curve was chosen to be shown in the inset of Extended Data fig. 7(d) of ref. [1].

The background signal. – According to eq. (1) and ref. [1], the data ("Superconducting Signal") are obtained from the raw data ("Measured voltage") by subtracting an independently measured background signal at a lower pressure, namely 108 GPa according to ref. [1], for which no superconductivity is expected. The numerical values of this background signal have not been reported by the authors. However, we can obtain them from eq. (1) as

$$\text{background signal} = \text{raw data} - \text{data}. \tag{2}$$

Figure 3 shows the resulting background signal in the different temperature ranges. The vertical scale in each case was chosen so that the curve fits in the graph. In order to compare the slopes of the different parts, we replot the

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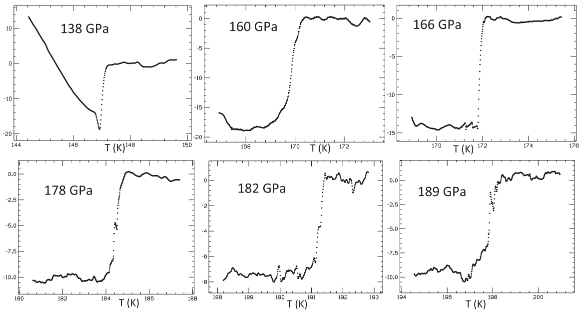

Fig. 1: Ac susceptibility data ("Superconducting Signal") *vs.* temperature for the six pressure values reported in ref. [1]. The numerical values were taken from the tables for "Superconducting Signal" given in ref. [2]. The ordinate gives the value of the signal in nV.

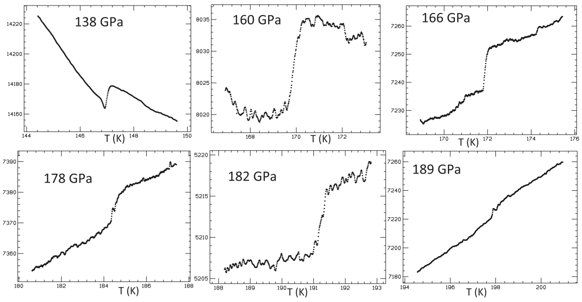

Fig. 2: Raw data ("Measured voltage") for ac susceptibility data *vs.* temperature for the six pressure values reported in ref. [1]. The numerical values were taken from the tables for "Measured voltage" given in ref. [2]. The ordinate gives the value of the voltage in nV.

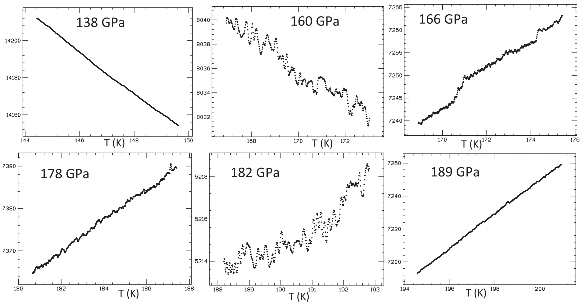

Fig. 3: Background signal for ac susceptibility data *vs.* temperature for the six pressure values reported in ref. [1], obtained from eq. (2), using the numerical values for raw data ("Measured voltage") and data ("Superconducting Signal") given in ref. [2]. The ordinate gives the value of the voltages in nV.

curves in fig. 4 using the same voltage interval in the vertical scale for all panels, namely 68 nV. It can be seen that there are large differences in the magnitude of the slopes, and that two curves have negative slopes and four have positive slopes.

Since the background is presumably a single background signal measured at 108 GPa for the entire temperature range, we would like to replot it as a single curve over the entire temperature range. However, the data for susceptibility reported in ref. [1] were shifted vertically so that they have values close to zero above the sharp jumps, as seen in fig. 1. As a consequence, in obtaining the background signal from eq. (2) there is an unknown vertical shift. To plot all the panels of fig. 4 on the same graph, we shifted the portions vertically to obtain the best possible smooth curve. The result is shown in fig. 5.

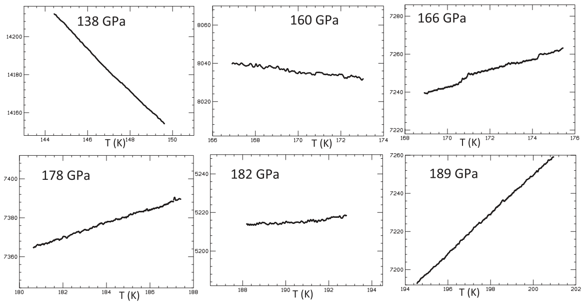

Fig. 4: Same curves as in fig. 3 now using the same range of voltage on the vertical axis, 68 nV, in order to allow visual comparison of the slopes of the curves in the different panels.

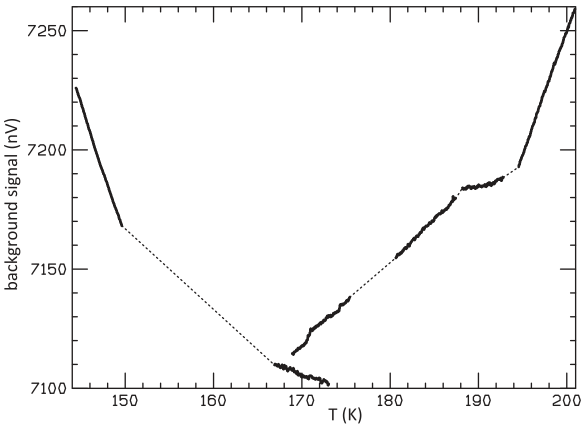

Fig. 5: Background signal for the entire temperature range. We have shifted the different portions vertically so that they can fit on the same curve with minimal changes in the slope. Neighboring portions of the curve are connected with straight dotted lines for visualization.

As can be seen in fig. 5, it is impossible that the background signal resulted from a single measurement, because the temperature ranges given in the panels of fig. 4 for 160 GPa and 166 GPa overlap, and the background signal curve has opposite slope in both panels. In addition, it can be seen that there are large changes in the slope in the region between 180 K and 200 K, also indicating that the different portions of the curve were not obtained in a single measurement *vs.* temperature.

We conclude that with the information given in refs. [1] and [2] we cannot understand how the background signal was obtained, in other words what was measured and subtracted from the "Measured voltage" to obtain the "Superconducting Signal" reported in these references.

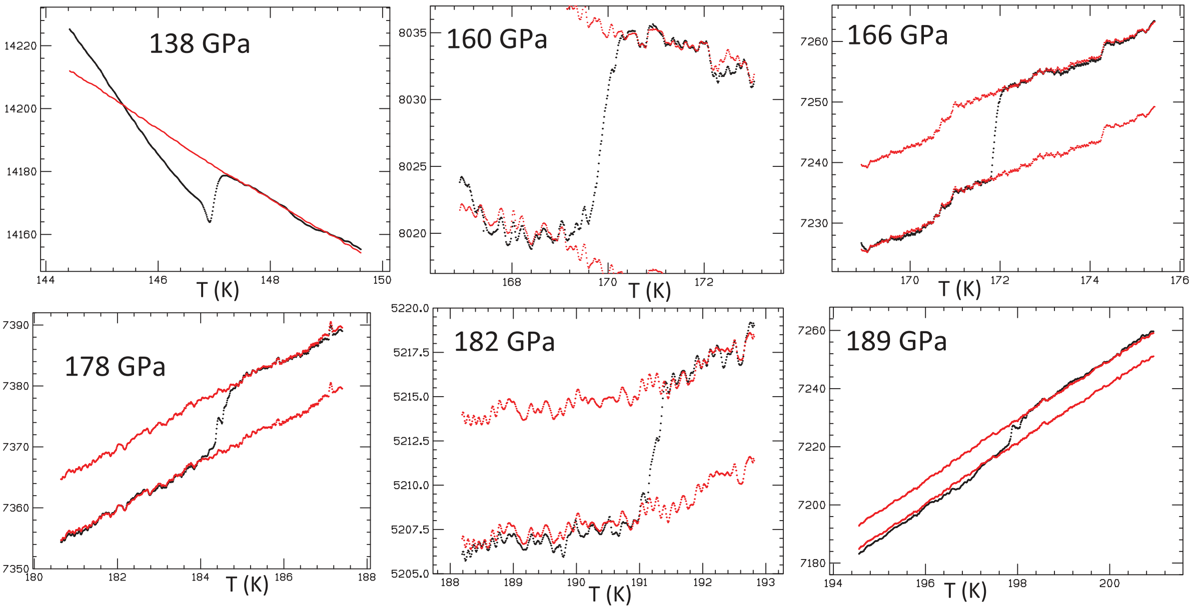

Fig. 6: Comparison of fine structure in the raw data (black points) and background signal (red points, upper curves). The lower red curves are identical to the upper red curves, shifted downward to facilitate comparison with the fine structure in the black curves for temperatures below the drops. The ordinate gives the value of the voltages in nV.

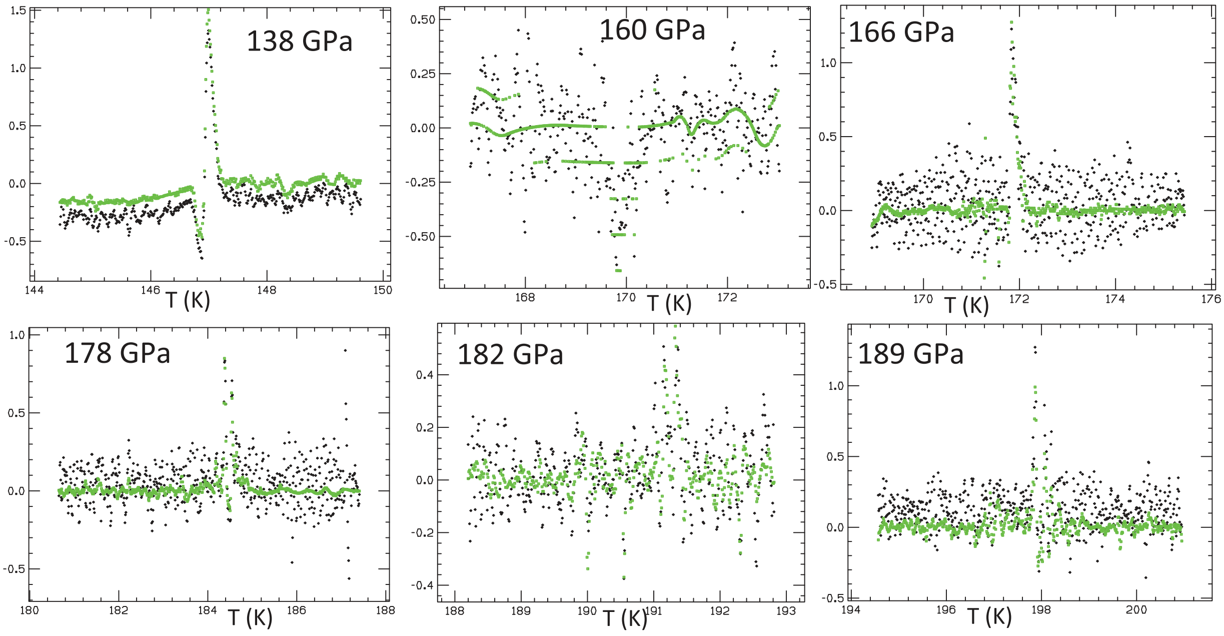

Fig. 7: Comparison of susceptibility increments given by eq. (3) (in nV) for neighboring points in temperature between raw data (black points) and data (green points). All values are obtained from the tables in ref. [2].

Fine structure of the background signal. – We had already reported in refs. [3,4] that the fine structure in the inferred background signal for three pressure values was very similar to the fine structure in the raw data. We find that this is also the case for the additional data reported in ref. [2]. We show the comparison for all the pressure values in fig. 6. In contrast to refs. [3,4] we use here the numerical values for data reported in ref. [2], while in refs. [3,4] we used the values obtained from analysis of the published vector graphic images since the numerical values had not been yet reported by the authors.

For the case of 138 GPa we only show one background signal curve because unlike the other cases the slope changes substantially below the jump. This is also the only case for which a background signal is also provided in ref. [2], albeit only graphically, in the upper panel of their fig. 7. The background signal shown there closely

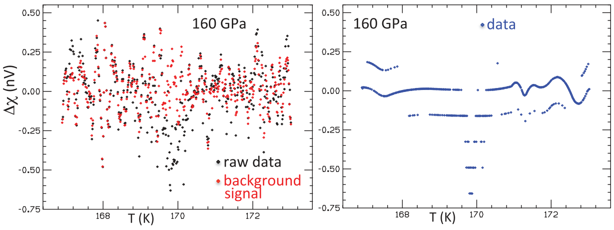

Fig. 8: Left panel: susceptibility increments for raw data (black points) and background signal obtained through eq. (2) (red points). Right panel: susceptibility increments for data, the difference between raw data and background signal shown in the left panel.

matches the background signal shown in fig. 6 upper left panel that we obtained from eq. (2).

It can be seen in fig. 6 that the fine structure in all the red curves (background signal) closely tracks the fine structure in the black curves (raw data). This is not understandable if the background signal originated in a different independent measurement at a different pressure, as claimed in ref. [1].

Comparison of susceptibility increments in raw data and in data. – To attempt to understand the relationship between the reported data ("Superconducting Signal") and raw data ("Measured voltage") we considered the susceptibility increments

$$\Delta\chi_i \equiv \chi_i - \chi_{i-1}, \tag{3}$$

where χ_i is either the data or the raw data for point i. In the tables given in ref. [2] the data and raw data are all given for the same list of temperature values, which facilitates comparison. Figure 7 shows comparison of the susceptibility increments for raw data and data for the six pressure values.

Recall that the data are supposedly obtained from the raw data through eq. (1). An independently measured background signal is subtracted from the raw data to arrive at the published data, denoted by "Superconducting Signal" in the tables of ref. [2]. However, we cannot understand fig. 7 in light of eq. (1). In particular, for 160 GPa, 166 GPa, 178 GPa and 189 GPa the range of values of $\Delta\chi$ for the raw data is *much* larger than the range of values of $\Delta\chi$ for the data. According to eq. (1) we would expect exactly the opposite: given a range of values for $\Delta\chi$ for the raw data and another one for the independently measured background signal, the resulting range of values of $\Delta\chi$ for the difference, *i.e.*, the data, should be larger than for both. Instead, it is substantially smaller.

Data for 160 GPa. – The discrepancy between what we expect to see and what we see is particularly glaring for 160 GPa.

For that case, the $\Delta\chi$ increments for the data in fig. 7 follow well defined lines with no scatter at all. We cannot understand how this behavior can result from a physical measurement of a voltage and subtraction of a physical measurement of another voltage at a different pressure. In fig. 8 we show on the left panel the susceptibility increments for the raw data (black points) and for the background signal obtained through eq. (2) (red points). The difference between these two sets of points obtained through what the papers say are separate measurements at different pressures gives rise to the data points shown on the right panel of fig. 8.

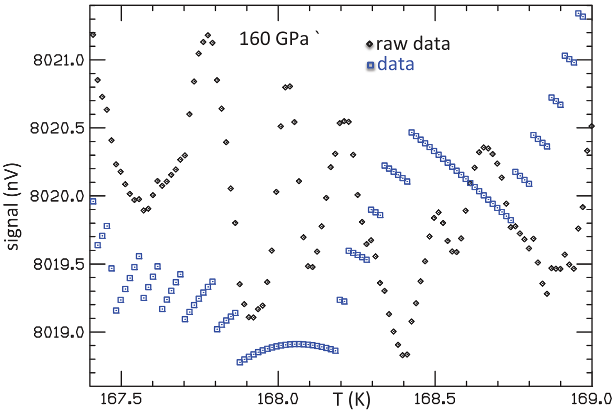

Fig. 9: Raw data (black diamonds) and data (blue squares) for a pressure of 160 GPa in the temperature range 167.4 K to 169 K, from table 5, lines 294 to 404, in ref. [2].

Finally, to highlight the anomalous features of the data for 160 GPa we show in fig. 9 the data and raw data for a limited range of temperatures that encompasses 112 points. The data show a complete disconnect with the raw data, and they follow a highly regular pattern. We cannot understand how such a regular pattern could result from a physical measurement *vs.* temperature, or from a combination of physical measurements *vs.* temperature.

Conclusion. – In this paper we have analyzed the underlying data for the ac susceptibility results reported in ref. [1] in support of the claim that carbonaceous sulfur hydride is a room temperature superconductor. These underlying data were supplied by two of the authors of ref. [1] in tables 1–10 of ref. [2]. To reiterate the nomenclature, in this paper we called "raw data" and "data" what ref. [2] calls "Measured voltage" and "Superconducting Signal" respectively. We have assumed that the "data" are related to the "raw data" through eq. (1), *i.e.*, subtraction of a "background signal" measured at a lower pressure, as reported by the authors of [1] in the figure caption of fig. 2(a). This is general practice in the field, the background signal is usually obtained for a pressure where no superconductivity is expected in the temperature range of interest [5]. Reference [1] informs that the background signal was obtained through measurements at a pressure of 108 GPa. The authors did not report the numerical values of the background signal in either of refs. [1,2], so assuming the validity of eq. (1) we obtained those numerical values using eq. (2) and the numerical values for the two terms on the right side of eq. (2) reported by the authors in ref. [2]. The numerical values for the background signal that we obtained from eq. (2) for 138 GPa appear to be identical to the background signal curve for that case shown in the upper panel of fig. 7 of ref. [2], the only case for which a background signal is given in refs. [1,2].

Our analysis has revealed several features that appear to contradict what is stated in the papers [1,2]. These features are:

1) The background signal that we obtained through eq. (2) shows anomalous temperature dependence and is double-valued in some temperature range, as shown in fig. 5.

2) The fine structure of the background signal obtained through eq. (2) closely tracks the fine structure of the raw data for all the pressure values as shown in fig. 6. This fine structure is presumably due to random noise and should not reproduce in independent measurements at different pressures. In refs. [3,4] we showed several examples of measurements in other materials, where the fine structure at any two different pressures is completely different.

3) The difference in the values of the data for neighboring temperatures $\Delta\chi$ shows substantially more scatter in the raw data than in the data, as shown in fig. 7. The opposite should be the case for data obtained from subtracting from the raw data an independently measured background signal. For pressure value 160 GPa the data show no scatter at all, as shown in fig. 8.

4) The highly regular data for 160 GPa given in table 5 of ref. [2], shown for a limited temperature interval in fig. 9, do not seem to have resulted from a physical measurement nor from a combination of physical measurements.

These data were a substantial part of the evidence presented in ref. [1] in favor of the claim that CSH is a room temperature superconductor. As a consequence, the results of this paper call that claim into question. Other reasons to question that claim were reported in refs. [6–8].

In addition, we do not have an explanation of the features 1), 2), 3) 4).

Data availability statement: The data that support the findings of this study are available upon reasonable request from the author, and they can be downloaded from `https://jorge.physics.ucsd.edu/cshdata.html`.

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