Journal of Superconductivity and Novel Magnetism (2024) 37:1779–1781
https://doi.org/10.1007/s10948-024-06792-9

MATTERS ARISING

Evidence of Superconductivity in Electrical Resistance Measurements of Hydrides Under High Pressure

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Received: 7 May 2024 / Accepted: 24 June 2024 / Published online: 16 October 2024

1 Matter Arising

In the standard van der Pauw four-probe configuration commonly used for electrical resistance measurements [1], including those of hydrogen-rich samples at high pressures [2–4], the application of electrical current through one pair of leads while measuring voltage difference across another pair is a fundamental practice (see the inset in Fig. 1c). In the scenario described in Ref. [5], where electrical resistance vanishes due to disconnection between current and voltage probes or due to an onset of giant magnetoresistance in parts of the sample, it is crucial to consider the implications across different probe orientations: while one orientation may indeed result in vanishing electrical resistance, a divergence of the electrical resistance towards infinity will be observed in an alternative orientation where the applied current pattern is rotated a quarter turn (see the scheme on the inset in Fig. 1c). It is imperative to acknowledge that experimental data encompassing all possible probe orientations are essential for eliminating artifacts and assessing sample homogeneity.

Our experimental investigations into the electrical resistance of hydrogen-rich compounds at high pressures consistently demonstrate the vanishing resistance across all probe orientations, as illustrated in Figs. 1 and 2 (data sourced from Refs. [2–4]). Hydrogen-rich high-temperature superconductors are synthesized under pressure of the order of 1 to 4 Mbar. For electrical transport measurements, the samples are typically synthesized on to top of metallic leads, as pictured in Fig. 1c insert. In our extended studies of the sulfur-hydrogen, lanthanum-hydrogen, and yttrium-hydrogen systems (total about 300 trials), the overall success rate to synthesize the final high-temperature phases—namely, *Im-3m*-H_3S/D_3S, *Fm-3m*-LaH_{10}/LaD_{10}, *Im-3m*-YH_6/YD_6, and *P6₃/mmc*-YH_9/YD_9 compounds—is about 25% [2, 6, 7]. This relatively small success rate is caused by the complexity of the experiment, including diamond anvils' failure during pressurizing and laser heating treatment, lack of hydrogen for the complete hydrogenation reaction, burnout of some deposited electrical leads during laser heating, shift of the sample away from the tips of electrical leads, or rapture of the isolating layer between the metallic gasket and electrical leads during pressurizing.

If the above problems are overcome and the presence of the superconducting phase is confirmed by X-ray diffraction analysis, all samples demonstrate superconducting transitions [2–4, 6–8]. We find that while there could be a significant drop in T_c upon decompression, as hydrides show a dome-shaped pressure dependence of T_c, the superconducting state persists in the same sample upon cycling of pressure within the stability range for a given phase as well as upon cycling of a magnetic field [2–4, 6–8]. For samples synthesized in deficiency of hydrogen, the formed metal hydrides, if not superconducting, usually exhibit metallic behavior. There are few examples where samples showed insulating/semiconducting behavior: (1) low-pressure (50–130 GPa) *Cccm*/*I4/mcm* phases of $(H_2S)_2H_2$ [4] and (2) low-pressure (up to 50 GPa) phases of YH_3 hydrides [7, 9].

Notably, the data derived from several different hydrogen-rich compounds including H_3S, D_3S, and LaH_{10} contradict the central claim of Ref. [5] while underlining the emergence of superconductivity within these compounds. For the sake of brevity, our previously published reports often depict the

This article comments on a paper published in the Journal of Superconductivity and Novel Magnetism available at https://doi.org/10.1007/s10948-023-06594-5.
A reply to the comment above has been published available at https://doi.org/10.1007/s10948-024-06833-3.

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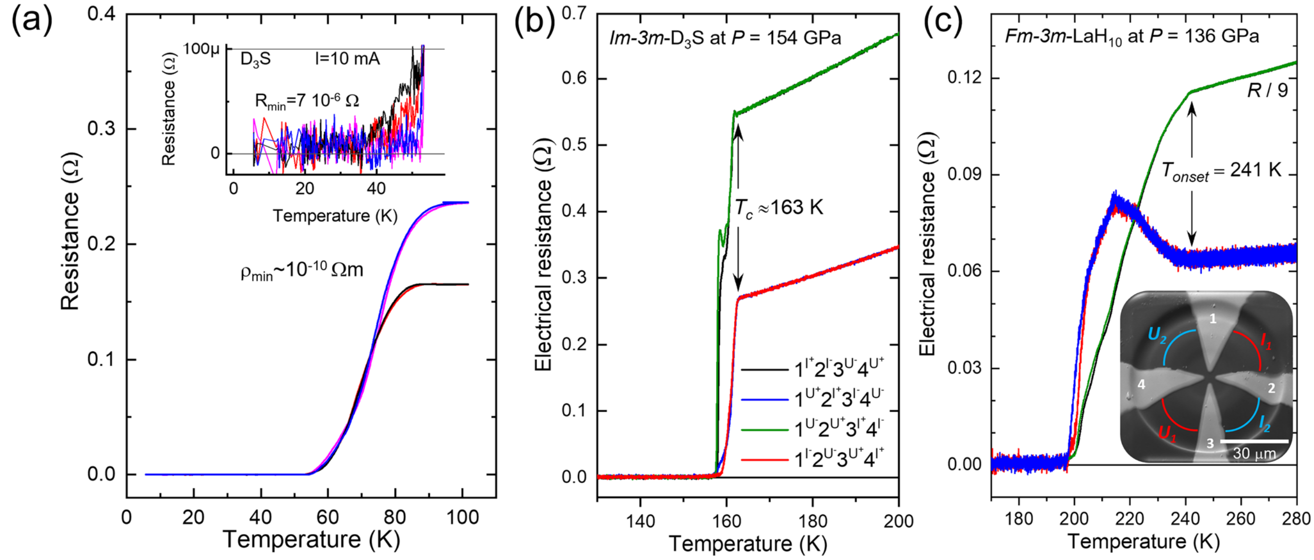

Fig. 1 Four-probe electrical transport measurements of hydrogen-rich compounds. **a**, **b** Temperature dependence of the electrical resistance of D_3S samples under high pressures. The samples were synthesized via disproportionation reaction from compressed D_2S and from elemental D_2 and S by laser heating treatment, respectively. Measurements were conducted in four different orientations of the current–voltage probes (marked by different colors). The inset provides a zoomed-in view highlighting the vanishing electrical resistance in D_3S. **c** Resistance versus temperature traces obtained for the LaH_{10} sample using the same measurement technique. The insert illustrates two measurement patterns, pattern 1 in red and pattern 2 in blue, rotated a quarter turn in relation to each other. The orientation of the current–voltage probes was rotated a quarter turn every second during slow warming (1–2 K/min), allowing the recording of resistance values from four different orientations within the same warming cycle. Any partial mismatch in $R(T)$ values between channels rotated half a turn could be attributed to non-linear resistive responses in superconducting transition regions due to sample inhomogeneity. Panel **a** is reproduced from Ref. [2]; panels **b** and **c** are reproduced from Ref. [3]

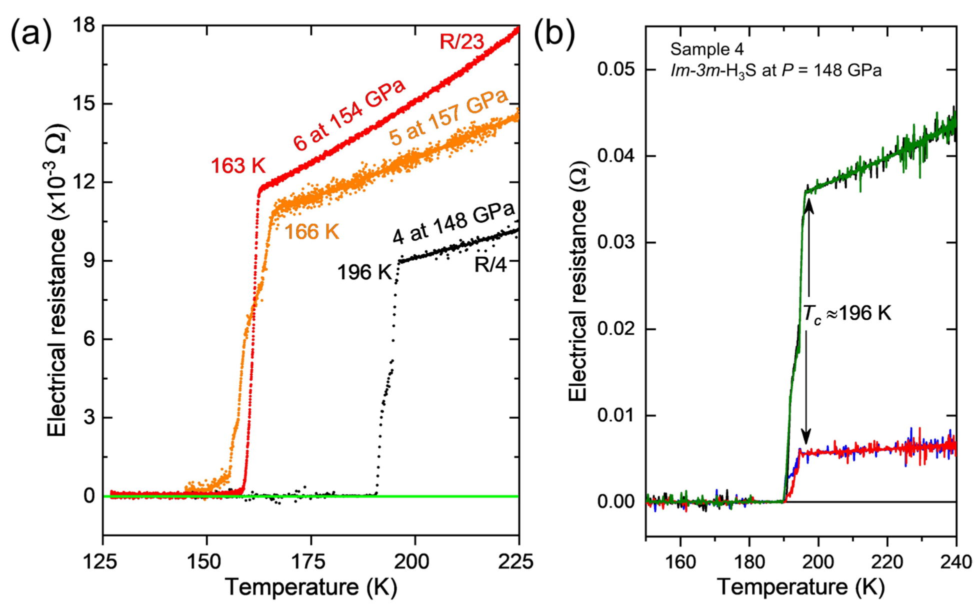

Fig. 2 Routine examination of the temperature dependence of electrical resistive response across all four orientations of the current–voltage probes. **a** $R(T)$ traces for *Im-3m*-H_3S (sample 4, black) and *Im-3m*-D_3S (sample 5, orange, and sample 6, red) as reported in Ref. [4]. The resistance for different samples was divided by factors between 1 and 23 as indicated on the plot. **b** Illustration of the routine examination of electrical resistance for sample 4 by alternating the orientation of the current–voltage probes during warming cycle. Each of the four channels is distinguished by a different color. The resistive response of sample 6 with varied current–voltage probe orientations is depicted in Fig. 1b. Panel **a** is reproduced from Ref. [3]

electrical resistance in a single probe orientation. However, it is essential to note that the temperature dependence of the samples' resistive response is routinely examined across all four orientations of the current–voltage probes, as exemplified by the H_3S sample in Fig. 2. This resistance measurement technique which includes rotation of the current flow pattern along four quadrants is explicitly described in Ref. [3] on Extended Data Fig. 3d and Ref. [4] on Fig. 2c, d and is reproduced here on Fig. 1, but was somehow overlooked in the comprehensive alternative explanation Ref. [5]. Consequently, we advocate for the authors of Ref. [5] to reassess their analytical approach and integrate all relevant data, including previously published findings that offer alternative perspectives.

In conclusion, our critique highlights the necessity for a thorough reevaluation of the assertions in Ref. [5]. Furthermore, we encourage the experimentalists investigating superconductivity to measure and provide data on electrical resistance for claimed superconducting samples utilizing different patterns for the current–voltage probes. Such an approach not only helps to eliminate potential artifacts but also provides more robust evidence supporting the observed phenomena. By adopting a comprehensive methodology, researchers can enhance the reliability and reproducibility of findings in the study of superconductivity.

Acknowledgements M.I.E. is thankful to the Max Planck community for valuable support and U. Pöschl for the encouragement. The National High Magnetic Field Laboratory is supported by the National Science Foundation through NSF/DMR-2128556, the State of Florida, and the U.S. Department of Energy.

Author Contribution F.F.B. and V.S.M. wrote the main manuscript text and F.F.B., V.S.M., A.P.D., and M.I.E. prepared Figs. 1 and 2. All authors reviewed the manuscript.

Funding Open Access funding enabled and organized by Projekt DEAL.

Data Availability The data that support the findings of this study are openly available in Open Science Framework at https://doi.org/10.17605/OSF.IO/J682T.

Declarations

Competing Interests The authors declare no competing interests.

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