

## Room-temperature Superconductivity – What More Needs to be Further Studied!

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The recent report [1] of room-temperature superconductivity in a carbonaceous sulfur hydride with a transition temperature  $T_c$  of 288 K achieved at 267 GPa by Ranga Dias *et al.*, represents one of the most exciting advancements in superconductivity science. The search for room-temperature superconductivity has been the main driving force for the century-long effort in superconductivity science and technology research and development. With a  $T_c$  near our ambient temperature ( $\sim 300$  K), we can operate a superconducting device without the burden of refrigeration. The serious temperature barrier that has prevented the ubiquitous application of superconductivity has thus been brought down. Unfortunately, to achieve the room-temperature superconductivity, ultrahigh pressure in hundreds of GPa is required. To make the ubiquitous applications of superconductivity practical, we have to overcome the ultrahigh pressure obstacle, *e.g.* to retain the ultrahigh-pressure-induced room-temperature superconducting phase without pressure. Such a possibility has recently been demonstrated in other superconductors [2].

Room-temperature as a relative term in superconductivity. In the early 1970s, the then-record  $T_c$  was 21 K. When Bernd Matthias, an undisputable leader in raising the  $T_c$  prior to high-temperature superconductivity, was asked “where to find room-temperature superconductivity”, his response was simply, “go to the edge of the universe” where the temperature is 3 K due to the residual background radiation remaining after the Big Bang. As a result, ever since its discovery in 1911, we have witnessed classes of superconductivity that refer to different operating temperatures of superconducting devices and that evolve with time, such as liquid-helium (4.2 K) [3] superconductivity for elements and alloys; liquid-hydrogen (20.3 K) [4] superconductivity for A15 compounds; liquid-nitrogen (77 K or high-temperature) [5] superconductivity for cuprates; liquid-Freon (115 K)[6] superconductivity for cuprates under high pressures; and now the room-temperature (293 K)[1] superconductivity for hydrogen-rich hydrides under ultrahigh pressures. This is also reflected in Vitaly Ginzburg’s selection of two topics of special importance and interest for problems in physics and astrophysics as controlled fusion and “high temperature superconductivity” in 1971[7], and then as controlled fusion and “high temperature/room temperature superconductivity” in 1999 [8].

Metallic hydrogen under high pressure. Being the simplest and most abundant element in the universe, hydrogen has fascinated many for practical and scientific reasons since its discovery in 1671 by Boyle and its later recognition as a discrete substance in 1776 by Cavendish. The understanding of its energy spectrum played a crucial role in the development of quantum mechanics. In 1928, Bernal envisioned that all substances would become metallic under adequately high pressure. In an attempt to test Bernal’s conjecture, to examine the then-developed free electron model, and to find the possible existence of a demarcation line between metals and nonmetals in the periodic table, Wigner and Huntington [9] found the possible metallization of hydrogen above a critical pressure  $P_c = 25$  GPa in 1935. With the invention of the diamond anvil cell, an extension of the Bridgman tungsten-carbide anvil, by van Valkenburg in 1956 and the subsequent crucial developments of the diamond anvil cell and the ruby pressure scale in the 1970s by Mao and Bell [10], the static pressure achieved in several research labs soon exceeded the suggested critical pressure  $P_c$  of 25 GPa for metallic hydrogen. The theoretical  $P_c$  has subsequently

continued to be pushed up to the TPa-range, but metallic hydrogen has yet to be unambiguously established up to  $\sim 500$  GPa [11].

*Metallic hydrogen as a room-temperature superconductor.* In 1968, Ashcroft [12] pointed out that, in addition to its astrophysical consequences, metallic hydrogen could be a superconductor and have a  $T_c$  of a few hundred Kelvin degrees based on the standard BCS formula  $T_c \propto \Theta_D \exp[-1/N(0)V]$  due to the high Debye temperature  $\Theta_D$  associated with its low ionic mass, a strong electron-phonon coupling  $V$  in the absence of an inner core structure, and a relatively large electron density of states  $N(0)$  due to its high electron density. In 1983, Carlsson and Ashcroft [13] suggested two possible techniques for reducing the external pressure required to induce the insulator-metal transition in solid hydrogen: to use impurities to lower the energy of the metallic phase relative to that of the insulating phase and to utilize a negative pressure induced in the insulating phase by electron-hole pairs. In 1997, Richardson and Ashcroft [14] showed that the correlated fluctuations between electrons and holes in the metallic modification of diatomic hydrogen through band-overlap under high pressure would reduce the Coulomb pseudopotential  $\mu^*$  and result in an enhanced  $T_c$  higher than its monatomic counterpart. Unfortunately, the metallic state in hydrogen was still elusive within the pressure range available in the labs, let alone high  $T_c$ . In 2004, Ashcroft [15] proposed a novel way to pre-compress hydrogen chemically by examining IVa hydrides, *e.g.*  $\text{SiH}_4$  should have a lower achievable critical pressure for the metallic state. Following this concept, J. Feng/N. W. Ashcroft *et al.* [16] estimated the crystal structure, the band structure, the electron-phonon coupling constant, and the Coulomb pseudopotential  $\mu^*$ , and, using the BCS/McMillan formula,  $T_c = (\Theta_D/1.45)\exp\{-1.04(1 + \lambda)/[\lambda - \mu^*(1 + 0.62\lambda)]\}$ , obtained a  $P_c \sim 91$  GPa and a  $T_c$  up to 166 K at 202 GPa. For the last 50 years, Neil Ashcroft has been considered the undisputed leader and motivator on the long, quiet, dark road to room-temperature superconductivity through hydrogen-rich molecular solids that has culminated in the discovery of Dias *et al.* [1].

*Hydrogen-rich molecular solids under ultrahigh pressure as room-temperature superconductors.* The metallization of the hydrogen-rich molecular solids under ultrahigh pressure is known to be the prerequisite for superconductivity. The rapid development of the high-pressure diamond anvil cell by David Mao and Peter Bell since the 1970s has provided the means to attain the required pressure and the calibrated ultrahigh pressure scale. As a result, Mikhail Erements' announcement in 2015 of superconductivity up to 203 K in  $\text{H}_3\text{S}$  at  $\sim 155$  GPa [17] heralded in the new era of very-high-temperature superconductivity in hydrogen-rich molecular solids. In 2017, Hemley and colleagues [18] examined and predicted the structural stability of high-pressure phases of lanthanum hydrides with strong electron-phonon interaction for a  $T_c$  of 274-286 K for  $\text{LaH}_{10}$  at 210 GPa. In 2019, Hemley and colleagues successfully synthesized  $\text{LaH}_{10}$  inside the diamond anvil cell through laser heating and achieved a  $T_c$  of 260 K under 180-200 GPa [19], very close to room temperature, opening up the era of room-temperature superconductivity. By continued thermal annealing, they reported a trace of superconductivity up to 556 K in the lanthanide "super hydrides" at 160 GPa, heralding in a new era of "hot hydride superconductivity" as coined in their recent 2020 article [20]. Later, Dias *et al.* reported the 288-K superconductivity at 267 GPa in a C-S-H compound formed under pressure by laser heating  $\text{CH}_4$  and  $\text{H}_2\text{S}$  in a diamond anvil cell [1]. The era of the room-temperature superconductor is thus born and it holds immense promise for technologists while offering great challenges for scientists.

*Interesting work ahead.* To fully realize the immense technological potential and to meet the great scientific challenges of the room-temperature superconductivity of the hydrogen-rich molecular

compounds, the exact nature of the ultrahigh-pressure-induced phase transitions must be understood and possible ways to retain this ultrahigh-pressure phase at ambient need to be explored. The primary challenges of the present experiments, in sample synthesis, handling, and characterization, are extraordinary, mainly arising from the very small sample size on the  $\mu\text{m}$  scale and the ultrahigh-pressure environment above 100 GPa. As a result, there are only a few teams in laboratories around the world who can carry out these types of experiments with rigor. Reports of superconductivity in hydrogen-rich molecular solids to date have been based on the agreement of experiment with theory and the detection of all or most of the following: a drastic drop in resistance to zero, suppression of the transition by magnetic field, and a diamagnetic shift in magnetic susceptibility and the isotope effect, all of which are expected of a standard superconductor. Given the significance of the reports and the constraints in the experimental conditions, the following points may be worthy of further study:

1. The existence of a pressure-induced insulator-metal (I-M) transition at a temperature  $T_m$  above  $T_c$  – this is important for exacting information about the normal-state behavior of the superconductor [21].
2. The achievement of a real “zero-resistance” state – it is extremely challenging to determine the “zero-resistance” or more so the “zero-resistivity” state due to the small size of the sample and the large change of resistance at the transition.
3. The field effect on the transition – a similar effect has been observed for the not-superconducting I-M transition [22].
4. The isotope effect on the transition – a similar effect has been observed for the not-superconducting I-M transition [23].
5. The diamagnetic shift in ac magnetic susceptibility – a similar effect has been detected in a temperature region where resistance changes greatly due to the eddy current.
6. Only the detection of the true Meissner effect (in the field-cooled mode) can clarify the above confusion, although the possible inherent defects in the sample under pressure may make such a test difficult. However, the sharpness of the transition may imply that the defects in the sample are small.
7. The very sharp transition and the almost downward shift of the transition in the presence of field suggest that the absence of flux flow is very puzzling for a type-II superconductor with such high a  $T_c$  [24].
8. The absence of a systematic experiment on the same individual sample for different types of measurements makes difficult the judgement of the reproducibility of the experiment – this is especially critical in determining the isotope effect [25].
9. The exact role of hydrogen in the samples investigated appears not to be clear – for instance, the role of B in the B-rich superconducting  $\text{ZrB}_{12}$  is rather limited [26].
10. The retention at ambient without pressure of the ultrahigh-pressure-induced room-temperature superconducting phase in these hydrogen-rich molecular solids should be the most exciting and rewarding endeavor in superconductivity science and technology research and development. Recent preliminary work on several superconducting elements and compounds has demonstrated such a possibility [2].

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