Superconductivity in La and Y hydrides: Remaining questions to experiment and theory

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Superconductivity in La and Y hydrides: Remaining questions to experiment and theory

Recent reports of the superconductivity in hydrides of two different families (covalent lattice, as in SH3 and clathrate-type H-cages containing La and Y atoms, as in LaH10 and YH6) have revealed new families of high-Tc materials with Tc values near room temperature. These findings confirm earlier expectations that hydrides may have very high Tc’s due to the fact that light H atoms have very high vibrational frequencies, leading to high Tc values within the conventional Bardeen–Cooper–Schrieffer phonon mechanism of superconductivity. However, as is pointed out by Ashcroft, it is important to have the metallic hydrogen “alloyed” with the elements added to it. This concept of a metallic alloy containing a high concentration of metal-like hydrogen atoms has been instrumental in finding new high-Tc superhydrides. These new superhydride “room-temperature” superconductors are stabilized only at very high pressures above 100 GPa, making the experimental search for their superconducting properties very difficult. We will review the current experimental and theoretical results for LaH10 and YH6 superhydrides.

I. INTRODUCTION

The attainment of room-temperature superconductivity is a long-standing challenge related historically to the metallic phase of hydrogens. The ambient-pressure molecular crystals of hydrogen are stable only at low temperature, and a metallic atomic-like hydrogen phase was proposed by Wigner and Huntington to occur under high pressure conditions. The pressure of metallization, estimated by Wigner and Huntington, of about 20 GPa, ultimately proved to be incorrect, and it was realized later that the actual metallization pressure should be around 500 GPa. Metallic hydrogen is also predicted to have a very high superconducting Tc, which naturally arises from the Bardeen–Cooper–Schrieffer (BCS) electron-phonon coupling mechanism involving hydrogen’s high vibrational frequencies. The atomic metallic phase of hydrogen is claimed to have been produced in several reports, including recently by Dias and Silvera, but all of these claims have met serious criticism. None of these reports have been able to show measurement of superconductivity in the claimed metallic phase.

After the BCS mechanism was established, it became clear that high vibrational frequencies are conducive to high Tc values. Many researchers have focused on studying the superconductivity of metallic hydrides in the hope of increasing Tc due to involvement of hydrogen vibrations in the “superconducting glue” provided by the electron-phonon mechanism. However, in many attempts to find such hydrides, Tc’s have failed to exceed 20 K. The formed hydrides did not have hydrogen-related electronic states at the Fermi level of the hydride. The chemical nature of hydrogen bonding in materials (ionic, covalent, or more exotic multicenter bonds) has prevented hydrogen electronic states from residing at the position of the Fermi level in all studied ambient-pressure hydrides. However, attempts to find superconducting hydrides have produced another idea involving the concept of “doped” metallic hydrogen,
which is usually attributed to Gilman and Ashcroft.14–16 Originally, Ashcroft conjectured that certain compounds of hydrogen with other elements in the periodic table may form metallic alloys, or, in other words, sustain a metallic hydrogen sublattice doped with well selected elements. Such “doped” compounds of hydrogen may have a stability range at pressures much lower than the 500 GPa required to stabilize pure atomic metallic hydrogen.5 These conditions may be amenable to the experimental techniques we currently possess for measuring the superconducting response at high pressures. While Ashcroft’s idea did not work well for the compounds he proposed (silane, SiH417 and similar group IV hydrides), more recent predictions by Li et al.18 have stimulated experimental work by Drozdov et al.19 and theoretical work by Duan et al.20 which found high Tc values approaching or exceeding room temperature in LaH10 and YH10 superhydrides. The predicted Tc values in selected metal hydrides from Peng et al.21 are shown in Fig. 2. Liu et al.22 predicted Tc’s of around 280 K in LaH10 around 210 GPa, and around 315 K in YH10, at 250 GPa (see Fig. 2).

The experimental confirmation of an Fm-3mLaH10 structure quickly followed in the publication by Geballe et al.24 The material was produced by heating an La flake in a hydrogen pressure medium at about 170 GPa. It was found that LaH10 Fm-3m structure is stabilized above 170 GPa, see Fig. 3, and at pressures below ∼165 GPa; the stable structure is R-3m LaH10. This finding supports theoretical estimates regarding the dynamic instability of LaH10 below 200 GPa,16,31 albeit the experimental pressure for the stability range of Fm-3m structure being significantly lower (165 GPa) than theoretical estimates.

The first reports of superconductivity in LaH10 appeared as arXiv publications. Drozdov et al. submitted a brief report of a resistivity drop at ∼215 K in laser-heated samples loaded in a hydrogen pressure

![Figure 1](link)

**FIG. 1.** Clathrate structures of typical metal superhydrides (polyhydrides). Maximum Tc is predicted for YH10 and is slightly above room temperature at 250 GPa. [Reprinted Figs. 2 and 4 with permission from Peng et al., Phys. Rev. Lett. 119(10), 107001 (2017). Copyright (2017) The American Physical Society.]

![Figure 2](link)

**FIG. 2.** Predicted superconductivity in LaH10 and YH10 superhydrides. Blue and red curves and symbols correspond to the indicated values of the Morel–Anderson pseudopotential, μ*.
medium. No structural data were given to support the claimed LaH_{10} superconducting phase. Shortly after, Somayazulu et al. submitted an arXiv contribution on the superconductivity of the LaH_{10} phase, which was confirmed by x-ray diffraction studies, with the onset of T_c up to 280 K. The four-probe data with zero residual resistance demonstrated a T_c onset at about 260 K. These results are in Ref. 25 along with additional measurements probing critical currents in the samples.

Drozdov et al. published another arXiv paper, claiming LaH_{10} with a maximum T_c of about 254 K, confirmed by an x-ray derived LaH_{10} structure, measured magnetic field suppression of T_c, and the effect of Deuterium-doping on T_c. These results are in Ref. 26.

The experimental setting for resistivity measurements using the four-probe technique from Somayazulu et al. is shown in Fig. 4. The sample of La was embedded in an ammonia borane pressure medium and pressed against four contacts on one of the diamond anvils. The sample was heated by laser under high pressure conditions from the side opposite to the electrodes, and ammonia borane served as a hydrogen source. The formation of an LaH_{10} phase was confirmed by x-ray diffraction data (Fig. 5). It was found that the onset of T_c can be observed in the temperature range of 245 K to almost 280 K in different samples, with varying hydrogen content (close to LaH_{10} as estimated from x-ray unit cell volume data; see Fig. 5).

Drozdov et al. have published resistivity data for several samples synthesized in a hydrogen pressure medium. While the
detailed procedure for sample synthesis is not clear from their work, it was stated that the La sample was bridged between the diamond anvils to ensure that the electrical contacts between the sample and the deposited electrodes was secured; this made sample synthesis by laser heating a challenging procedure due to an effective heat sink provided by the contacts of the samples with diamond anvils. Thus, we may assume that these synthesis conditions are not favorable for creating a relatively homogeneous, single phase of an La hydride. Indeed, the resistive response and x-ray data show the existence of multiple phases with varying hydrogen content in the reported experiments. For the LaH$_{10}$ phase, Drozdov et al. reported a dome-like dependence of $T_c$ on pressure. While their measurement range extended as low as 140 GPa, they did not show any evidence of an R-3m LaH$_{10}$ phase, which was reported by Geballe et al. below 165 GPa—Fig. 6. This discrepancy is not clear, since both studies used La in a hydrogen pressure medium and the reported conditions for the synthesis of LaH$_{10}$ were very similar.

Magnetic susceptibility measurements have not been attempted for LaH$_{10}$ due to the very small volume of the synthesized samples—see Ref. 26, which is well below the limit of the MPMS squid...
system used to prove superconductivity in H$_2$S samples.$^{19}$ We have recently studied magnetic susceptibility in LaH$_{10}$ using our sensitive magnetic susceptibility technique$^{25,35}$ and have obtained interesting initial results which we describe below.

III. MAGNETIC SUSCEPTIBILITY STUDIES AT HIGH PRESSURE IN LaH$_{10}$

The technique we use for magnetic susceptibility measurements is a double-modulation technique introduced originally by Timofeev,$^{26}$ which we improved$^{35}$ and adopted for measuring small samples in multimegabar experiments.$^{25}$ The technique is demonstrated in Fig. 7, along with the calculated and measured response from a YBa$_2$Cu$_3$O$_{7-x}$ superconductor (see Ref. 38 for details). For tiny samples the background issues become quite severe, and the diamond anvil cell, the gasket, and all surrounding metal parts should be made from nonmagnetic materials. The background signal may become comparable to the detected signal, as was the case in experiments on pure sulfur in pressures up to 230 GPa.$^{2}$ For the experiments on LaH$_{10}$ we used a BeCu piston-cylinder, Mao-Bell type cell with a nonmagnetic NiCrAl (Russian alloy) gasket,$^{38}$ and ammonia borane as a pressure medium. The samples were handled in an inert environment to avoid contamination. The details of the loading procedure are similar to those in Ref. 25. The samples were brought to a synchrotron (Advanced Photon Source, Argonne National Laboratory, GSECARS beamline) and the La sample was heated by a pulsed laser in an ammonia borane pressure medium at about 165 GPa producing LaH$_{10}$ samples. The details of sample synthesis are reported elsewhere.$^{39}$ The DAC with synthesized samples was brought to the HPSTAR facility in Shanghai for magnetic susceptibility measurement, and after three weeks to the Geophysical Laboratory (Carnegie Institution of Washington) for a similar set of measurements.

We were able to detect a measurable signal, however, the estimated sample size proved to be very small. We show raw data at several pressures in Fig. 8. The onset of the superconducting response was at about 250 K in two temperature scans immediately following synthesis at the beamline. After three weeks the shape of the signal changed, showing the shift in response to 278 K. This occurred without further laser heating and may be attributed to pressure effects, since average sample pressure increased to nearly 180 GPa, as measured by the shift in the edge of the Raman signal from the diamond anvils.

We have estimated sample size using previous measurements from larger samples of high-T$_c$ superconductors and MgB$_2$ samples (Fig. 9). For the observed signal at 180 GPa, we obtain a sample volume of about $4 \times 10^{-10}$ cm$^3$, which corresponds to a sample size of around 10 μm in diameter, depending on the sample shape and demagnetization factor. This sample size is comparable to the size of the hot spot during the laser heating procedure. The change in T$_c$ after synthesis may be explained by the pressure increase and/or phase changes in the sample. It should be noted that the magnetic response in Fig. 8 does not look like the expected single crystal signal shape in Fig. 7 and should correspond both to the inhomogeneity in the samples due to pressure gradients (experimentally, gradients up to 20 GPa are observed over the culet area) and to the possible presence of multiple phases. Further experiments are required to produce larger and more homogeneous samples to understand the reason for the higher T$_c$ values observed in our experiments. However, there may be a physical reason for the higher T$_c$'s in the LaH$_{10}$ samples in a certain pressure range. We address this issue below.

We summarize the available experimental data for LaH$_{10}$ samples in Fig. 10. The most interesting fact is the T$_c$ values up to 280 K observed in experiments by Somayazulu et al.$^{25}$ and in our magnetic susceptibility data. Both these studies used an ammonia borane pressure medium as a hydrogen source, and the discrepancy with Drozdov et al.$^{26}$ may be due to the incorporation of B or N in the synthesized samples, or to the formation of LaH$_x$ phases different from the LaH$_{10}$ phase. However, the x-ray data from Ref. 25 are not compatible with this explanation. A second possibility becomes evident if we notice that from theoretical results, the LaH$_{10}$ phase is dynamically unstable below 200 GPa$^{16,31}$ and, according to experimental data, LaH$_{10}$ does have a phase transition to the R-3m phase below 165 GPa$^{26}$ (Fig. 6).

Indeed, the existence of a phase transition in the 160–170 GPa pressure range, which is driven by a soft mode (dynamical instability),...
may explain an increase in $T_c$ in the relatively narrow pressure range above the transition. An indirect confirmation can be invoked from the theoretical calculations of $T_c$ in LaH$_{10}$ by Kruglov et al.—Fig. 10—who find a dramatic increase in $T_c$ in a narrow pressure range close to 200 GPa from $T_c \sim 270$ K to nearly 310 K—see Fig. 10. Such an enhancement of $T_c$ was not found in the calculations by Liu et al.—(Fig. 10), and they did not show any calculated $T_c$ data below 205 GPa. Given the relatively large pressure gradients in the samples, such a transition is easy to miss, especially if the whole volume of the sample is not probed, as in resistivity studies. We note here that only one pressure point in the range 173–203 GPa is reported in the paper by Drozdov et al.

In the recent arXiv contribution, Errea et al. calculated $T_c$ values for the LaH$_{10}$ Fm-3m phase, taking into account the stabilization of the Fm-3m structure by quantum atomic fluctuations. They found that quantum fluctuations stabilized the Fm-3m phase down to 140 GPa, and found $T_c$ values in agreement with Drozdov et al. It is not currently clear why the transition to the R-3m phase, reported by Geballe et al., is not reproduced in this quantum fluctuations approach, but it is evident that such calculations are missing the soft mode scenario we discussed above.

Kruglov et al. also presented a $T_c$ calculation for the R-3m phase of LaH$_{10}$ and obtained $T_c \sim 200$ K at 150 GPa, which is significantly lower than reported $T_c$'s at 150 GPa (~250 K). The reason for this discrepancy is not clear, and further, more extended theoretical efforts would be helpful in understanding how the soft mode may influence $T_c$ in the LaH$_{10}$ samples around the dynamic instability pressure range.
IV. NEW SUPERCONDUCTING SUPERHYDRIDES

YH₆ AND ThH₁₀

In a recent arXiv publication, Troyan et al.⁴¹ reported a superconductivity of up to 224 K in a YH₆ superhydride, which confirmed earlier predictions of a high Tc in this material,⁴²,⁴³ albeit with lower experimental Tc values. We show structural determination and Tc measurements using a resistivity technique in Figs. 11 and 12, respectively. The x-ray diffraction of the Tc = 224 K sample shows predominantly the YH₆ phase (Fig. 11). Some other minor phases have been observed, including I4/mmm-YH₄ and Imm2-YH₇ at pressures of 160–180 GPa. The synthesized I3m3m-YH₆ shows a clear superconducting transition with a Tc of 224 K at 166 GPa. This value of Tc is lower than most theoretical predictions, which produce Tc’s in the range 260–280 K for YH₆.⁴²,⁴³,⁴⁴ While being lower than theoretical predictions, this is the third-highest critical temperature that has been experimentally measured in superhydrides. The lower experimental Tc values may be due to strong anharmonicity effects and quantum effects, as recently discussed for LaH₁₀.⁴⁵

Another superconducting hydrides report was released in arXiv in 2019 by Semenok et al.,⁴⁶ claiming ThH₁₀ with Tc = 161 K at 174 GPa and ThH₉ with Tc = 146 K at 170 GPa. The superconductivity was measured from the resistivity drop to zero and confirmed by suppression of the transition in the magnetic field. The corresponding experimental results are summarized in Fig. 13. The fcc-ThH₁₀ was predicted to have a stabilization pressure of 85 GPa, which makes it unique among known high-Tc metal superhydrides, however, the experimental Tc values were reported only at 174 GPa. In this material, theory predicts a Tc of up to 241 K at 90–100 GPa,⁴⁵ and Tc = 160 K at 174 GPa;⁴⁶ the latter value is close to the experimental value of 161 K. The predicted Tc for ThH₉ at 150 GPa is 123–145 K, which is not far from the experimental value of 146 K at 170 GPa.⁴⁴ Overall, it appears that the predicted and measured Tc values in thorium superhydrides are very similar, contrary to what is found in LaH₁₀ and YH₆ materials, where anharmonic or quantum effects may play a significant role in defining the Tc values in the materials.

Notes added in review: After this paper was submitted for review, an important arXiv contribution was published on experimental studies of superconductivity in the new YH₆ and YH₇ phases.⁴⁶ Tc = 227 K was observed for a YH₆ sample at 237 GPa, and a Tc of YH₇ had a dome-like shape as a function of pressure with a maximum Tc = 243 K at 201 GPa. Notably, the observed Tc’s are lower by ~30 K than the predicted Tc’s similar to our observation for the YH₆ sample. We refer the reader to the original publication for further details.

V. PERSPECTIVES FOR FUTURE ROOM-TEMPERATURE SUPERCONDUCTIVITY STUDIES

With the available experimental data on superconducting superhydrides, we may summarize a few empirical facts related to their structural and superconducting properties. In Fig. 14, we show the highest Tc values predicted by theory in superhydrides,⁷ which are similar to the highest Tc values observed in pure elements under high-pressure conditions (available at http://www.hpr.stec.es.osaka-u.ac.jp/e-super). Both periodic table representations look strikingly similar. Indeed, from previous theoretical predictions we know that metal superhydrides with clathrate-like hydrogen lattices at the beginning of the period (but not pure alkali metal hydrides) are predicted to have high Tc values.⁴⁸ Similarly, most of the elements at the beginning of the period (Ca, Y, Sc) show quite high Tc values at high pressures. This correlation does not look coincidental if we adopt the “doped” hydrogen approach. Metal ions are embedded in the hydrogen “clathrate” lattice, and when the surrounding electronic density is optimized so that the pure metal exhibits high Tc values, the “encaged” metals may contribute substantially to electron-phonon coupling in the material. It is possible that such an approach may give better superconducting materials if ternary compounds are used, allowing continuous tuning of the electronic density. Indeed, for ternary compounds very high Tc values are predicted for a few materials, such as Li₂MgH₁₆ with a predicted Tc ~ 473 K at 250 GPa.

Another important observation is the systematic trends in the partial volume occupied by the hydrogen atom in high-Tc superhydrides. For example, in Fig. 15 we show a comparison of hydrogen partial volume in various hydrides, including high-Tc superhydrides. It is evident that the partial volume for the hydrogen atom in proven

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FIG. 12. Troyan et al. Superconducting transitions in Im3m-YH6: (a) Dependence of electrical resistance on temperature. Inset: the resistance drops to zero after cooling below $T_C$; (b) jump in $R(T)$ dependence of resistance (nine times increase) on temperature for the second sample.

FIG. 13. Semenok et al. Observation of superconductivity in (a) ThH$_{10}$ and (b) ThH$_9$. The temperature dependence of the resistance (R) of thorium superhydride was determined in a sample synthesized from Th$^+$NH$_3$BH$_3$. The resistance was measured using four electrodes deposited on a diamond anvil with the sample placed on top of the electrodes [Fig. 15(a), inset] with an excitation current of 100 $\mu$A. The resistance near the zero point is shown on a smaller scale in the insets; (c) dependence of the resistance on temperature under an external magnetic field at 170 GPa; (d) dependence of the critical temperature ($T_c$) of ThH$_{10}$ and ThH$_9$ on the magnetic field.
high-Tc superhydrides is above the value calculated for hypothetical atomic metallic hydrogen and is much lower in several nonsuperconducting hydrides like AlH3, FeH3, FeH5. This may be related to the augmented electron density around the hydrogen atom in superconducting superhydrides. It is interesting that the atomic hydrogen volume in high-Tc materials, like LaH10 and ThH10, is very close to the partial volume of hydrogen in compressed solid H2. Overall, the equation of state for metallic atomic hydrogen serves as a dividing line between superconducting and nonsuperconducting hydrides in Fig. 15. In the context of “doped” metallic hydrogen, as suggested by Ashcroft, it appears that the hydrogen clathrate sublattice in superhydrides is “overdoped” in comparison with the atomic metallic phase of pure hydrogen.

We believe that the volume differences given in Fig. 15 are quite robust, since we have a very good match between the theoretical and experimental volumes of superhydride phases, for example in thorium and yttrium superhydrides. In that respect, the partial hydrogen volume may become a very sensitive probe of the “doping” state of hydrogen in superconducting hydrides. It would be interesting to explore the optimum doping condition for clathrate hydrogen lattices, which can be done by studying ternary compounds.

The clathrate hydrogen cages in superhydrides are stable only above 100–150 GPa in most studied materials. In that respect, ThH10 stands out as a predicted viable candidate for high-Tc superconductivity below 100 GPa. It would be interesting to check these predictions experimentally. It is not clear what makes thorium hydrides more stable at lower pressures, but experimentalists may certainly benefit from such predictions of lower pressure, high-Tc hydrides.

VI. CONCLUSION

In this brief review we summarized what is known about new superconducting hydrides (superhydrides) that have exceptionally high critical superconducting temperatures. We provided the latest magnetic susceptibility data for LaH10, showing the possibility of high-Tc phases with Tc’s of up to 280 K. The mechanism for the enhancement of Tc in LaH10 was also suggested, based on a soft mode scenario in the vicinity of the Fm-3m to R-3m phase transition in LaH10.
We introduced new experimental data on YH6 and ThH10 in the context of “doped” metallic hydrogen. It appears that the partial hydrogen volume in such superhydrides may be a good indicator of “doping.” In superhydrides, based on partial volume arguments, we suggest that the hydrogen sublattice may be “overdoped” with respect to (hypothetical) pure metallic hydrogen. Overall, the concept of doping may prove very useful in studying ternary superhydrides and is an excellent tool in tuning the doping levels of the hydrogen sublattice.

While at the moment it appears that superconducting superhydrides lack practical importance due to their stability range in the hundreds of GPa, the scientific challenge of the field is enormous and promises exciting times. Room-temperature superconductivity has almost been achieved experimentally, and increasing numbers of theoretical predictions show significantly high $T_c$’s, even well above the boiling point of water.23 The experimental challenge is much harder than the theoretical one, but we believe most theoretical predictions will be tested in the near future.

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