Synthesis and Superconductivity of Yttrium Hexahydride

Im$\bar{3}$m-YH$_6$

I. A. Troyan$^1$, D. V. Semenok$^{2,*}$, A. G. Kvashnin$^{2,3}$, A. G. Ivanova$^1$, V. B. Prakapenka$^4$, E. Greenberg$^4$, A. G. Gavriliuk$^{1,8}$, I. S. Lyubutin$^1$, V. V. Struzhkin$^{5,6}$ and A. R. Oganov$^{2,3,7}$

$^1$Shubnikov Institute of Crystallography, Federal Scientific Research Center Crystallography and Photonics, Russian Academy of Sciences, Moscow, 119333, 59 Leninskii pr-t, Russia
$^2$Skolkovo Institute of Science and Technology, Skolkovo Innovation Center 121205, 3 Nobel Street, Moscow, Russia
$^3$Moscow Institute of Physics and Technology, 141700, 9 Institutsky lane, Dolgoprudny, Russia
$^4$Center for Advanced Radiation Sources, The University of Chicago, 5640 South Ellis Avenue, Chicago, Illinois 60637, USA
$^5$Center for High Pressure Science and Technology Advanced Research, Shanghai 201203, China
$^6$Geophysical Laboratory, Carnegie Institution of Washington, Washington, DC 20015, USA
$^7$International Center for Materials Discovery, Northwestern Polytechnical University, Xi'an, 710072, China
$^8$IC RAS Institute for Nuclear Research, Russian Academy of Sciences, Moscow, 117312 Russia

Abstract

Here we report a targeted high-pressure synthesis of high-$T_C$ hydride superconductor Im$\bar{3}$m-YH$_6$ with predicted $T_C \sim$ 270-280 K and upper critical magnetic field ($\mu H_C$) up to 75 tesla. According to our experiments cubic YH$_6$ can be prepared in a mixture with I4/mmm-YH$_4$ and Imm2-YH$_7$ at pressures of 160-180 GPa via laser heating of metallic yttrium in ammonia borane medium over 2400 K. Compressed Im$\bar{3}$m-YH$_6$ demonstrates clear superconducting transition with $T_C \sim$ 224 K at 166 GPa, unexpectedly lower than it was theoretically predicted (> 273 K). Currently, this is the second highest critical temperature that has been experimentally achieved in compressed hydrides. New phases were examined theoretically, and their electronic, phonon, mechanical and superconducting properties were studied.

Keywords: yttrium hydrides, USPEX, high pressure, superconductivity, X-ray diffraction
Introduction

Room-temperature superconductivity has been an unattainable dream and subject of speculative discussions for a long time, but times change. Theoretical prediction of record high-temperature superconductor LaH$_{10}$\cite{1} with $T_C \sim 260$ K followed by the experimental confirmation\cite{2,3} has opened a new field in high-pressure physics devoted to investigation of superconducting metal hydrides. Recently successful synthesis of previously predicted $hcp$-CeH$_6$\cite{4}, $hcp$- and $c$-PrH$_6$\cite{5}, UH$_7$ and UH$_8$\cite{6} motivated us to perform an experimental investigation of the Y-H system in order to synthesize previously predicted another potential room-temperature superconductor Im$3$m-YH$_6$, stable at pressures 100-300 GPa.

Interest to the study of yttrium hexahydride YH$_6$ is due to not only its outstanding superconducting properties, but also because of the very low stabilization pressure of this compound – just about 70-120 GPa. In other words, it is due to the highest pressure application efficiency – $\max(T_C/P) \approx 2.5$ K/GPa\cite{1,7,8}, among all known superconducting hydrides.

Stability and conditions of existence of yttrium hexahydride with sodalite-like crystal structure, similar to famous Im$3$m-CaH$_6$ denoting as $c1l4$-CaH$_6$, were studied in a series of papers starting from 2015\cite{1,7,8}. So, in the work by Li et al. (2015)\cite{7} the Im$3$m-YH$_6$ with hexagonal H$_6$ units was predicted for the first time to be stable at pressures over 110 GPa. The superconducting transition temperature was found via numerical solution of the Eliashberg equations to be in range 251–264 K at 120 GPa ($\mu^* = 0.1–0.13$), and the EPC parameter $\lambda$ of reached 2.93 at the same pressure. The authors were also investigated the band structure and the density of electronic states at the Fermi level of YH$_6$ at 120 GPa ($N(E_F) = 0.6$ states/eV/f.u.). The $\omega_{\text{log}}$ was not given in Ref.\cite{7}, but according to the provided Eliashberg function $\alpha^2F(\omega)$ we calculated it as $\sim 1080$ K at 120 GPa.

In the work by Liu et al.\cite{1} published in 2017, yttrium and lanthanum hydrides were theoretically studied in details, with an emphasis on decahydrides $fcc$-LaH$_{10}$ and YH$_{10}$. Superconductivity in YH$_6$ has not been studied independently. However, some new information was given for the hexahydride: H-H distance in YH$_6$ was found to be 1.19 Å at 300 GPa, stability range of the sodalite-like structure was determined from to be in range 150-300 GPa. In addition, the average $\omega_{\text{log}} = 1124$ K for YH$_6$ at 120 GPa was calculated, which agrees with the value of 1080 K calculated above from the data of Ref.\cite{7}.

![Yttrium hexahydride (YH$_6$) properties](image)

**Figure 1.** Yttrium hexahydride (YH$_6$) properties, previously studied by various authors: a) YH$_6$ unit cell parameters at 150 GPa; b) different orbital contributions to the density of electronic states $N(E_F)$\cite{8}; c) Dependence of superconducting parameters ($T_C, \lambda, \omega_{\text{log}}$) on pressure\cite{8}

The study of YH$_6$ was continued in Grishakov et al. in 2018\cite{8}, where, using the original method\cite{6}, significantly lower superconducting transition parameters were obtained: $\lambda = 3.0$, $T_C = 165$ K at 125 GPa. It was found in Ref.\cite{8} that the major contribution to the density of electronic states at Fermi level $N(E_F)$ comes from $s,d$-orbitals, while the contribution from $p$-orbitals was insufficient. The calculation of the pressure dependence of $N(E_F)$ in the range 100-
400 GPa shows that it decreases linearly with increasing pressure from about 0.9 to 0.75 eV-electron (Figure 1b). At the same time, contribution from $d, p$-orbitals decrease, while contribution from $s$-orbital increases. Calculated $N(E_F)$ values in Ref. 8 and Ref. 7 are different, which leads to contradiction.

The most detailed study of physical properties and superconductivity of $Im\bar{3}m$-YH$_6$ was made by Heil et al. in 2019. Calculations were carried out using fully anisotropic Migdal-Eliashberg theory (as implemented in EPW code) with Coulomb corrections. Almost isotropic superconducting gap resulting from a uniform distribution of the coupling over states of both Y and H sublattices was found. According to provided calculations the Coulomb screening is relatively weak, resulting in a Morel-Anderson pseudopotential $\mu^* = 0.11$. Predicted critical temperature for YH$_6$ was estimated to be 290 K at 300 GPa (Figure 1c), which exceeds “room temperature” limit (273 K). The H-H distances at the pressure of 300 GPa is 1.19 Å, EPC coefficient $\lambda = 1.73$, average SC gap is 50-55 meV. The dependence of $T_C$ on pressure was found to be very weak ($dT_C/dP = +0.2$ K/GPa) because of the compensation between the average phonon energy $\omega_{\text{log}}$, which increases with pressure, and the electron-phonon coupling constant $\lambda$, which, decreases instead. Authors were also able to estimate anharmonic effects (via frozen-phonon calculations) in YH$_6$ and found that anharmonic contrivution to the $T_C$ is weak in this compound.

It should be mentioned a short communication of A. Bergara at the 56th EHPG Meeting, where the results of calculations of $T_C$ of YH$_6$ with anharmonic Eliashberg function at 120 GPa were reported. Anharmonicity leads to decrease in $\lambda$ and increase in $\omega_{\text{log}}$. As a result, critical temperature decreased by 34 K.

Previous results of theoretical studies of $Im\bar{3}m$-YH$_6$ are shown in Table 1 and partly agree with each other and partly contradict to each other. To establish the real state of affairs, in this work we carried out an experimental synthesis and study of the superconducting properties of yttrium hydrides $Im\bar{3}m$-YH$_6$, $Imm2$-YH$_7$ and $I4/mmm$-YH$_4$, obtained after laser heating of metallic yttrium in ammonium borane (NH$_3$BH$_3$) medium in diamond anvil cells (DACs) in the pressure range 165-180 GPa. Superconducting properties of the yttrium polyhydrides were investigated by measuring the electrical conductivity of samples in different current modes.

Table 1. Superconducting parameters of $Im\bar{3}m$-YH$_6$ calculated in previous theoretical studies sorted by pressure.

| Parameter | 100 GPa | 120 GPa | 125 GPa | 200 GPa | 300 GPa |
|-----------|---------|---------|---------|---------|---------|
| $N(E_F)$, states/eV/f.u. | 0.75 $^8$ | 0.6 $^7$ | 0.88 $^8$ | 0.63 $^8$ | 0.69 $^9$ |
| $\lambda$ | 3.44 $^*$ | 2.93 $^7$ | 3.0 $^8$ | 1.93 | 1.73 $^9$ |
| $\omega_{\text{log}}$, K | 851 $^*$ | 1080 $^7$, 1124 $^1$ | - | 1282 | 1612 $^9$ |
| $T_C$, K | 233 $^*$(A-D) | 251–264 $^7$ | 165 $^8$ | 285 $^9$ | 290 $^9$ |
| Anharmonic $\Delta T_C$, K | - | -34 $^{10}$ | - | ~0 $^9$ | ~0 $^9$ |

* Calculated from $\alpha^2F(\omega)$ given in Ref. 9 at $\mu^* = 0.1$.

Results and discussions

Having a number of theoretical studies of $c14$ yttrium hexahydride that have been already published, we focused on experimental verification of stability and superconducting properties of $Im\bar{3}m$-YH$_6$, as well as on the calculations of some physical properties that have not been analyzed before.

The high-pressure synthesis was carried out according to the previously developed scheme with ammonium borane (AB) as a source of hydrogen that showed excellent results. We prepared three diamond anvil cell (DAC) with 50 μm culets – K1, M1, M3, where pure
Yttrium metal was loaded into a sublimated ammonia borane and compressed to 166-180 GPa. The samples were heated up to 2400 K by $\sim 10^5$ pulses of 1 microsecond each, leading to formation of mixtures of $Im\bar{3}m$-YH$_6$ and YH$_6$, YH$_7$-YH$_{7.5}$ in all cells.

Results of the synthesis are highly depending on the pressure and temperature of the laser heating. In the cells K1 and M1 (XRD patterns are shown in Figure 2, S3), the laser heating of samples at 166 GPa yielded the complex mixture of products, with predominant formation of $Im\bar{3}m$-YH$_6$, $Imm2$-YH$_7$ with cubic Y-sublattice (M1, Figure S3), and, probably, low symmetry $P1$-YH$_{7+x}$ ($x = \pm 0.5$) hydride with non-stoichiometric composition. To estimate thermodynamic stability and possibility of these hydrides to be formed at experimental pressure and temperature conditions the evolutionary search of stable Y-H compounds was carried out using the USPEX algorithm. Computational predictions show that all these phases can be stabilized at high temperatures in a range 500-2000 K (see Figure 3).

![Image](https://example.com/image.png)

**Figure 2.** a) XRD pattern of K1 sample at 166 GPa recorded with wavelength of $\lambda = 0.2952$ Å; b) Le Bail refinements of $Im\bar{3}m$-YH$_6$, $Imm2$-YH$_7$ and $P1$-YH$_{7.5}$ at 180 GPa. Red circles are experimental data; black line is the fit; green line shows residues

![Image](https://example.com/image.png)

**Figure 3.** a) Calculated convex hull of Y-H system at 150 GPa at 0 K; b) at 500 K.

The examination of thermodynamic stability of predicted yttrium hydrides at 150 GPa shows that at 0 K (ZPE contribution was taken into account) the only stable phases are $I4/mmm$-YH$_4$ and $P1$-YH$_7$, see Figure 3a. YH$_6$ is metastable with the enthalpy of formation higher than convex hull by 17 meV/atom. The enthalpy difference between $Imm2$ and $P1$-YH$_7$ phases are about 38 meV/atom with more favorable polymorph with $P1$ space group. Temperature increase to 500 K (Figure 3b) leads to stabilization of YH$_6$, while the YH$_{10}$ still thermodynamically and
dynamically metastable. The difference in the enthalpies of YH\textsubscript{7} polymorphs vanishes as the temperature approaches \(~\sim\)2000 K. The more symmetrical \textit{Imm}2-YH\textsubscript{7} phase becomes thermodynamically more favorable at higher temperatures.

Calculated convex hull has complicated structure due to the presence of a large number of different phases located close to the convex hull and having similar compositions. The best candidates which explain observed XRD are slightly nonstoichiometric \textit{I4/mmm}-YH\textsubscript{4} and low-symmetry \textit{P1}-YH\textsubscript{7+}\textsubscript{x} (\(x= \pm 0.5\)) phase.

Higher pressures (172-180 GPa, M3 cell) lead to a much simpler XRD pattern (see **Figure 4**), showing XRD peaks only from \textit{Im\bar{3}m}-YH\textsubscript{6} and distorted \textit{I4/mmm}-YH\textsubscript{4} phases. Experimental lattice parameters and volumes of newly discovered Y-H phases are given in **Table 2**. All the phases were also theoretically examined for the dynamical and mechanical stability according to the Born’s criteria (\(C_{11} - C_{12} > 0, C_{11} + 2C_{12} > 0, C_{44} > 0\)), see Supporting Information.

![Figure 4](image.png)

**Figure 4.** a) The XRD pattern of M3 sample at 172 GPa recorded at \(\lambda=0.2952\) Å ; b) Le Bail refinements for \textit{Im\bar{3}m}-YH\textsubscript{6} and \textit{I4/mmm}-YH\textsubscript{4}. Red circles are experimental data; black line is the fit; green line shows residues.

| **Table 2.** The experimental and predicted lattice parameters and volumes of \textit{Im\bar{3}m}-YH\textsubscript{6} (Z = 2), \textit{I4/mmm}-YH\textsubscript{4} (Z = 2) и \textit{Imm}2-YH\textsubscript{7} (Z=2). |
|---|---|---|---|---|
| \textit{Im\bar{3}m}-YH\textsubscript{6} | | | |
| DAC | Pressure, GPa | \(a\), Å | \(V\), Å\(^3\) | \(a\text{DFT},\) Å | \(V\text{DFT},\) Å\(^3\) |
| M1 | 165 | 3.578(3) | 45.8(2) | 3.573 | 45.62 |
| K1 | 168 | 3.582(3) | 45.9(1) | 3.565 | 45.31 |
| M3 | 172 | 3.571(2) | 45.5(3) | 3.557 | 45.02 |

| \textit{I4/mmm}-YH\textsubscript{4} | | | |
| DAC | Pressure, GPa | \(a\), Å | \(c\), Å | \(V\), Å\(^3\) | \(V\text{DFT},\) Å\(^3\) |
| K1 | 168 | 2.751(4) | 5.15(8) | 39.01 | 39.63 |

| \textit{Imm}2-YH\textsubscript{7} | | | |
| DAC | Pressure, GPa | \(a\), Å | \(b\), Å | \(c\), Å | \(V\), Å\(^3\) | \(V\text{DFT},\) Å\(^3\) |
| M1 | 165 | 3.29(4) | 3.33(6) | 4.68(7) | 51.50 | 50.85 |
Measurements of superconducting properties of Im$\bar{3}$m-YH$_6$

To measure the superconducting transition temperature of the yttrium hexahydride, all cells were equipped with four Ta/Au electrodes. We used the DACs with a 50 $\mu$m culet beveled to 300 $\mu$m at 8.5$^\circ$. Four Ta electrodes (~200 nm) with gold plating (~80 nm) were sputtered onto the piston diamond. Composite gaskets consisting of a tungsten ring and a CaF$_2$/epoxy mixture were used to isolate the electrical leads.

An ~1-2 $\mu$m-thick yttrium sample was sandwiched between the electrodes and AB in the gasket hole with the diameter of 20 $\mu$m. The pressure in the cell was increased to 166 GPa (K1 cell), 165 GPa (M1 cell) and 172 GPa (M3 cell). The electrodes in the M3 cell were found to be connected with the W-gasket, therefore, no resistivity measurements were made for this sample.

The heating of the sample was performed by ~$10^5$ pulses of a Nd:YAG infrared laser with the wavelength $\lambda = 1.064$ $\mu$m, and duration of 1 $\mu$s each pulse. The temperature dependence of the resistance is shown in Figure 5.

Figure 5. Superconducting transitions in Im$\bar{3}$m-YH$_6$: a) The dependence of electrical resistance on temperature in sample K1. Inset: the resistance drops to zero after the cooling below $T_C$; b) the jump in $R(T)$ dependence of resistance (9 times increase) on temperature for M1 sample.

Two slightly different superconducting transitions with $T_C$ of 224 K (Figure 5a) and 218 K (Figure 5b) were observed in K1 and M1 samples, respectively. In the K1 cell the electrical resistance dropped sharply to zero (from 50 m$\Omega$ to 5 $\mu$Ω, width $\Delta T_C \sim$1-2 K), while in M1 cell the complete disappearance of the electrical resistance was not observed due to presence of the side phases (Imm2-YH$_7$, see Supporting Information, Figure S3).

Analysis of the electronic and superconducting properties of tetragonal YH$_4$ and orthorhombic YH$_7$ indicates (see Table 3) that YH$_4$ is a metal with significantly lower critical temperature ($\leq$ 107 K) compared to YH$_6$. Another product of the synthesis (in the M1 cell), the Imm2-YH$_7$ phase, has pronounced “pseudogap” in the electronic density of states, quite low $N(E_F)$ and, as a result, even lower $T_C$ of 31-43 K. Thus, as will be shown below, these impurities have practically no effect on the superconducting transition in yttrium hexahydride.

It is interesting to compare the experimentally obtained $T_C$ with theoretical calculations based on the Bardeen–Cooper–Schrieffer and Migdal–Eliashberg theories. As we mentioned in the introduction, the results of the analysis of superconducting properties in different works are different: the estimates range from 250 to 285 K which is quite far from the experimental value (224 K). Given this, as well as the fact that studied pressure range of 160-180 GPa was not covered by previous calculations, we carried out a series of independent harmonic calculations of SC properties in Im$\bar{3}$m-YH$_6$, I4/mmm-YH$_4$ and Imm2-YH$_7$ at fixed pressure of 165 GPa (see Table 3). The critical temperature for YH$_6$ obtained by numerical solution of the isotropic
Eliashberg equation\(^\text{15}\) turned out to be significantly lower (220-240 K, at \(\mu^* = 0.15-0.1\)) than the estimates given by Heil et. al\(^\text{9}\) and close to the results from Ref.\(^\text{1}\) after the anharmonic correction\(^\text{10}\). This allows us to estimate the expected upper critical magnetic field \(\mu_0H_C(0) \sim 68\) Tesla, the superconducting gap in \(\text{YH}_6\) as 57 meV, and the coherence length \(\xi_{\text{BCS}} = 0.5\sqrt{\hbar/\pi eH_C^2} = 22\) Å.

**Table 3.** Parameters of the superconducting state of \(\text{Im}\overline{3}m\)-\(\text{YH}_6\), \(I4/mmm\)-\(\text{YH}_4\), and \(\text{Im}m2\)-\(\text{YH}_7\) at 165 GPa calculated by isotropic Eliashberg equations (E)\(^\text{15}\) and Allen-Dynes (A-D)\(^\text{16}\) formula. Here \(\gamma\) is Sommerfeld constant, \(\mu^*\) is 0.15-0.1, \(\beta\) – Allen-Dynes isotopic coefficient.

| Parameter | \(\text{Im}\overline{3}m\)-\(\text{YH}_6\) | \(I4/mmm\)-\(\text{YH}_4\) | \(\text{Im}m2\)-\(\text{YH}_7\) |
|-----------|-----------------|-----------------|-----------------|
| \(\lambda\) | 2.24            | 1.30            | 0.89            |
| \(\omega_{\text{bg}}\), K | 929             | 970             | 695             |
| \(\alpha_2\), K | 1449            | 1545            | 1287            |
| \(\beta\) | 0.465 - 0.484   | 0.45-0.48       | 0.42-0.47       |
| \(T_c\) (A-D), K | 158-184         | 87-107          | 31-43           |
| \(T_c\) (E), K | 220-240         | 111-131         | 36-46           |
| \(N(E_F)\), states/eV/f.u. | 0.71            | 0.475           | 0.07            |
| \(\Delta(0)\), meV | 51-57           | 15.5-20         | 5-7.2           |
| \(\mu_0H_C(0)\), T | 64-68           | 16.3-20.5       | 1.9-2.7         |
| \(\Delta C/T_c\), mJ/mol·K\(^2\) | 31-29.4         | 11.5-12.6       | 1.1-1.2         |
| \(\gamma\), mJ/mol·K\(^2\) | 10.81           | 5.14            | 0.62            |
| \(R_{\lambda} = 2\Delta(0)/k_B T_c\) | 5.38-5.47       | 4.14-4.34       | 3.74-3.88       |

A good agreement between experiment and theoretical calculations of superconducting properties, both approximate and more accurate, performed with anharmonic correction, is a tangible argument in favor of the classical electron-phonon pairing mechanism in \(\text{Im}\overline{3}m\)-\(\text{YH}_6\), which is also characteristic to other polyhydride materials.

From the results of this study, one can make some predictions regarding the superconducting properties of other hydride materials. So, for instance, predicted \(T_c\) values for \(\text{Im}\overline{3}m\)-CaH\(_6\) of 220-235 K\(^\text{17}\), isostuctural to yttrium hexahydride, are much lower than similar parameters of the \(\text{YH}_6\). If the same difference remains valid for the experimental data, the critical temperature of CaH\(_6\) may be lower than stated, in accordance with our experimental results for \(\text{YH}_6\).

**Conclusions**

Currently, the second highest (after \(\text{fcc-LaH}_{10}\)) critical temperature superconductor \(\text{Im}\overline{3}m\)-\(\text{YH}_6\), together with the novel phases of \(I4/mmm\)-\(\text{YH}_4\) and \(\text{Im}m2\)-\(\text{YH}_7\), were synthesized at 160-180 GPa confirming earlier theoretical predictions\(^\text{7}\). The measured critical temperature of the \(\text{Im}\overline{3}m\)-\(\text{YH}_6\) was found to be 224 K, which is unexpectedly lower than theoretically predicted values (> 273 K\(^\text{9}\)). Low symmetry (\(P1\) or \(\text{Im}m2\)) molecular yttrium hydrides \(\text{YH}_{7+x}\) (\(x \pm 0.5\)) were found to be responsible for complex XRD patterns at 166 GPa. The experiment shows the great potential of Y-H system in terms of the increasing of hydrogen content and transformation of \(\text{Im}\overline{3}m\)-\(\text{YH}_6\) to \(\text{fcc-YH}_{10}\) which has the best chances to be a room-temperature superconductor.
Methods

Experiment

To perform this experimental study, three (K1, M1, M3) diamond anvil cells (DACs) were loaded. The diameter of the working surface of the diamond anvils was 280 μm beveled at angle of 8.5° to culet of 50 μm. X-ray diffraction patterns of all samples in diamond anvil cells were recorded at the GSECARS synchrotron beamline at the Advanced Photon Source (Argonne, US) with beam focused down to $2.5 \times 3.5$ μm X-ray beam at 42 KeV and 37 KeV and ($\lambda = 0.295$ Å and 0.334 Å) and a Pilatus 1M CdTe detector placed at a distance of ~200 mm from the sample. The exposure time was 20–60 s. LaB6 standard was used for the detector geometry calibration. The X-ray diffraction data were analyzed and integrated using the Dioptas software package (version 0.4) 19. The full profile analysis of the diffraction patterns, as well as the calculation of the unit cell parameters, was performed in the JANA2006 program 20 by the Le Bail method 21.

Online laser heating of the samples was done at GSECARS by pulses of infrared laser with the wavelength of $\lambda = 1.064$ μm and pulse duration of 1 μs 36. Temperature measurements were carried out using the grey body radiation fit within the Planck function at the laser heating system of the GSECARS beamline of APS. Applied pressure was measured by the edge position of the Raman signal of diamond 22 using the Acton SP2500 spectrometer with PIXIS:100 spectroscopic-format CCD 37.

Theory

The equations of state of the discovered YH4, YH6, YH7 phases were calculated using density functional theory (DFT) 23,24 within the generalized gradient approximation (Perdew-Burke-Ernzerhof functional) 25, and the projector-augmented wave method 26,27 as implemented in the VASP code 28-30. Plane wave kinetic energy cutoff was set to 500 eV and the Brillouin zone was sampled using $\Gamma$-centered $k$-points meshes with resolution $2\pi\times0.05$ Å$^{-1}$. We also calculated phonon densities of states of studied materials using the finite-displacements method (VASP and PHONOPY31,32).

Calculations of superconducting $T_c$ were carried out using QUANTUM ESPRESSO (QE) package 33. Phonon frequencies and electron-phonon coupling (EPC) coefficients were computed using density-functional perturbation theory 34, employing plane-wave pseudopotential method and Perdew-Burke-Ernzerhof exchange-correlation functional 25. In our ab initio calculations of the electron-phonon coupling (EPC) parameter $\lambda$, the first Brillouin zone was sampled using $4\times4\times4$ $q$-points mesh, and a denser $16\times16\times16$ $k$-points mesh (with Gaussian smearing and $\sigma = 0.005$ Ry, which approximates the zero-width limits in the calculation of $\lambda$). $T_c$ was calculated from the Eliashberg equations 15 which were solved by iterative self-consistent method for the imaginary part of the order parameter $\Delta(T, \omega)$ (superconducting gap) and the renormalization wave function $Z(T, \omega)$ 35 (see Supporting Information). More approximate estimates of $T_c$ were made using the Allen-Dynes formula 16.

Author Contributions:

These authors contributed equally to this work. I.A.T., V.V.S. V.B.P, E.G., A.G.G., I.S.L. performed the experiment, D.V.S., A.G.I., A.G.K. and A.R.O. prepared theoretical analysis.
Acknowledgments

The work was performed on APS (Argonne, USA), station GSECARS. The work on high-pressure experiments was supported by the Ministry of Science and Higher Education of the Russian Federation within the State assignment of the FSRC “Crystallography and Photonics” of RAS and by the Russian Science Foundation (Project No.19-12-00414). Authors thank RFBR foundation № 19-03-00100, A.G.K. thanks facie foundation, grant UMK1 №13408GU/2018 for financial support of this work. A.R.O. thanks Russian Science Foundation (grant 19-72-30043). Portions of this work were performed at GeoSoilEnviroCARS (The University of Chicago, Sector 13), Advanced Photon Source (APS), Argonne National Laboratory. GeoSoilEnviroCARS is supported by the National Science Foundation – Earth Sciences (EAR – 1634415) and Department of Energy- GeoSciences (DE-FG02-94ER14466). This research used resources of the Advanced Photon Source, a U.S. Department of Energy (DOE) Office of Science User Facility operated for the DOE Office of Science by Argonne National Laboratory under Contract No. DE-AC02-06CH11357.

References

1. Liu, H., Naumov, I. I., Hoffmann, R., Ashcroft, N. W. & Hemley, R. J. Potential high-Tc superconducting lanthanum and yttrium hydrides at high pressure. PNAS 114, 6990–6995 (2017).
2. Drozdov, A. P. et al. Superconductivity at 250 K in lanthanum hydride under high pressures. Nature 569, 528 (2019).
3. Somayazulu, M. et al. Evidence for Superconductivity above 260 K in Lanthanum Superhydride at Megabar Pressures. Phys. Rev. Lett. 122, 027001 (2019).
4. Salke, N. P. et al. Synthesis of clathrate cerium superhydride CeH9 at 80 GPa with anomalously short H–H distance. arXiv:1805.02060 https://arxiv.org/ftp/arxiv/papers/1805/1805.02060.pdf (2018).
5. Zhou, D. et al. Superconducting Praseodymium Superhydrides. arXiv:1904.06643 [cond-mat] (2019).
6. Kruglov, I. A. et al. Uranium polyhydrides at moderate pressures: prediction, synthesis, and expected superconductivity. Sci. Adv. 4, eaat9776 (2018).
7. Li, Y. et al. Pressure-stabilized superconductive yttrium hydrides. Scientific Reports 5, 09948 (2015).
8. Grishakov, K. S., Degtyarenko, N. N. & Mazur, E. A. Electron, Phonon, and Superconducting Properties of Yttrium and Sulfur Hydrides under High Pressures. J. Exp. Theor. Phys. 128, 105–114 (2019).
9. Heil, C., di Cataldo, S., Bachelet, G. B. & Boeri, L. Superconductivity in sodalite-like yttrium hydride clathrates. Phys. Rev. B 99, 220502 (2019).
10. Bergara, A. Anharmonic effects and optical spectra in superconducting hydrogen. in *Proceedings of 56th EHPRG Meeting* (2018).
11. Semenok, D. V. *et al.* Synthesis of ThH4, ThH6, ThH9 and ThH10: a route to room-temperature superconductivity. arXiv:1902.10206 [cond-mat] (2019).
12. Oganov, A. R. & Glass, C. W. Crystal structure prediction using ab initio evolutionary techniques: Principles and applications. *J. Chem. Phys.* **124**, 244704 (2006).
13. Oganov, A. R., Ma, Y., Lyakhov, A. O., Valle, M. & Gatti, C. Evolutionary crystal structure prediction as a method for the discovery of minerals and materials. *Rev. Mineral Geochem* **271–298** (2010).
14. Oganov, A. R., Lyakhov, A. O. & Valle, M. How Evolutionary Crystal Structure Prediction Works—and Why. *Acc. Chem. Res.* **44**, 227–237 (2011).
15. Eliashberg, G. M. Interactions between Electrons and Lattice Vibrations in a Superconductor. *JETP* **11**, 696–702 (1959).
16. Allen, P. B. & Dynes, R. C. Transition temperature of strong-coupled superconductors reanalyzed. *Phys. Rev. B* **12**, 905–922 (1975).
17. Wang, H., Tse, J. S., Tanaka, K., Itaka, T. & Ma, Y. Superconductive sodalite-like clathrate calcium hydride at high pressures. *PNAS* **109**, 6463–6466 (2012).
18. Kvashnin, A. G., Semenok, D. V., Kruglov, I. A. & Oganov, A. R. High-Temperature Superconductivity in Th–H System at Pressure Conditions. *ACS Applied Materials & Interfaces* **10**, 43809–43816 (2018).
19. Prescher, C. & Prakapenka, V. B. DIOPTAS: a program for reduction of two-dimensional X-ray diffraction data and data exploration. *High Pressure Research* **35**, 223–230 (2015).
20. Petříček, V., Dušek, M. & Palatinus, L. Crystallographic Computing System JANA2006: General features. *Zeitschrift für Kristallographie - Crystalline Materials* **229**, 345–352 (2014).
21. Bail, A. L. Whole powder pattern decomposition methods and applications: A retrospection. *Powder Diffraction* **20**, 316–326 (2005).
22. Akahama, Y. & Kawamura, H. Pressure calibration of diamond anvil Raman gauge to 410 GPa. *J. Phys.: Conf. Ser.* **215**, 012195 (2010).
23. Hohenberg, P. & Kohn, W. Inhomogeneous electron gas. *Phys. Rev* **136**, B864–B871 (1964).
24. Kohn, W. & Sham, L. J. Self-consistent equations including exchange and correlation effects. *Phys Rev* **140**, A1133–A1138 (1965).
25. Perdew, J. P., Burke, K. & Ernzerhof, M. Generalized gradient approximation made simple. *Physical review letters* **77**, 3865–3868 (1996).
26. Blöchl, P. E. Projector augmented-wave method. *Phys. Rev. B* **50**, 17953–17979 (1994).
27. Kresse, G. & Joubert, D. From ultrasoft pseudopotentials to the projector augmented-wave method. *Phys. Rev. B* **59**, 1758–1775 (1999).
28. Kresse, G. & Furthmüller, J. Efficient iterative schemes for ab initio total-energy calculations using a plane-wave basis set. *Phys. Rev. B* **54**, 11169–11186 (1996).
29. Kresse, G. & Hafner, J. Ab initio molecular dynamics for liquid metals. *Phys. Rev. B* **47**, 558–561 (1993).

30. Kresse, G. & Hafner, J. Ab initio molecular-dynamics simulation of the liquid-metal amorphous-semiconductor transition in germanium. *Phys. Rev. B* **49**, 14251–14269 (1994).

31. Togo, A. & Tanaka, I. First principles phonon calculations in materials science. *Scripta Materialia* **108**, 1–5 (2015).

32. Togo, A., Oba, F. & Tanaka, I. First-principles calculations of the ferroelastic transition between rutile-type and CaCl2-type SiO2 at high pressures. *Phys. Rev. B* **78**, 134106 (2008).

33. Giannozzi, P. *et al.* QUANTUM ESPRESSO: a Modular and Open-Source Software Project for Quantum Simulations of Materials. *Journal of Physics: Condensed Matter* **21**, 395502 (2009).

34. Baroni, S., de Gironcoli, S., Dal Corso, A. & Giannozzi, P. Phonons and Related Crystal Properties from Density-Functional Perturbation Theory. *Reviews of modern Physics* **73**, 515–562 (2001).

35. Allen, P. B. & Dynes, R. C. A computer program for numerical solution of the Eliashberg equation to find Tc. *Technical Report #7 TCM/4/1974* (1974).

36. Prakapenka, V. B., Kubo, A., Kuznetsov, A., Laskin, A., Shkurikhin, O., Dera, P., Rivers, M. L. and Sutton, S. R. Advanced flat top laser heating system for high pressure research at GSECARS: application to the melting behavior of germanium. *High Pressure Research* **28**, 3, p. 225-235 (2008).

37. Nicholas Holtgrewe, Eran Greenberg, Clemens Prescher, Vitali B. Prakapenka & Alexander F. Goncharov. Advanced integrated optical spectroscopy system for diamond anvil cell studies at GSECARS, *High Pressure Research* (2019, in press), DOI: 10.1080/08957959.2019.1647536.
SUPPORTING INFORMATION

Synthesis and Superconductivity of Yttrium Hexahydride

Im\(\bar{3}\)m-YH\(_6\)

I. A. Troyan\(^1\), D. V. Semenok\(^2,*\), A. G. Kvashnin\(^2,3\), A. G. Ivanova\(^1\), V. B. Prakapenka\(^4\), E.
Greenberg\(^4\), A. G. Gavriliuk\(^1,8\), I. S. Lyubutin\(^1\), V. V. Struzhkin\(^5,6\) and A. R. Oganov\(^2,3,7\)

\(^1\)Shubnikov Institute of Crystallography, Federal Scientific Research Center Crystallography and
Photonics, Russian Academy of Sciences, Moscow, 119333, 59 Leninskii pr-t, Russia

\(^2\)Skolkovo Institute of Science and Technology, Skolkovo Innovation Center 121205, 3 Nobel Street,
Moscow, Russia

\(^3\)Moscow Institute of Physics and Technology, 141700, 9 Institutsky lane, Dolgoprudny, Russia

\(^4\)Center for Advanced Radiation Sources, The University of Chicago, 5640 South Ellis Avenue,
Chicago, Illinois 60637, USA

\(^5\)Center for High Pressure Science and Technology Advanced Research,
Shanghai 201203, China

\(^6\)Geophysical Laboratory, Carnegie Institution of Washington,
Washington, DC 20015, USA

\(^7\)International Center for Materials Discovery, Northwestern Polytechnical University, Xi’an, 710072,
China

\(^8\)IC RAS Institute for Nuclear Research, Russian Academy of Sciences, Moscow, 117312 Russia

Content

Cell parameters..............................................................................................................................13
Raman spectra ............................................................................................................................14
Elastic properties .........................................................................................................................15
Electronic and phonon properties...............................................................................................17
Eliashberg functions and equations for calculating T\(\text{C}\) and related parameters .................19
References ..................................................................................................................................23
## Cell parameters

Table S4. Crystal data of predicted yttrium hydrides and pure elements at 150 GPa.

| Phase       | Volume Å³/atom | Lattice parameters | Coordinates |
|-------------|----------------|-------------------|-------------|
|             |                |                   | atom        | x    | y    | z    |
| **Fdd-Y**   | 12.88          | a = 16.867 Å      | Y1          | -0.187 | 0.125 | 0.125 |
|             |                | b = 4.564 Å       |             |       |      |      |
|             |                | c = 2.677 Å       | H1          | 0.261  | 0.083 | 0.254 |
| **C2/c-H**  | 1.86           | a = 5.332 Å       | H2          | 0.148  | 0.197 | 0.271 |
|             |                | b = 3.052 Å       | H3          | 0.000  | -0.159 | 0.250 |
|             |                | c = 4.484 Å       | H4          | 0.000  | -0.403 | 0.250 |
|             |                | β = 142.25 °      |             |       |      |      |
| **Fm3m-YH** | 7.643          | a = 3.939 Å       | Y1          | 0.500  | 0.500 | 0.500 |
|             |                |                   | H1          | 0.000  | 0.000 | 0.000 |
| **I4/mmm-YH** | 4.927      | a = 2.995 Å       | Y1          | 0.000  | 0.000 | 0.000 |
|             |                | b = 4.392 Å       | H1          | 0.000  | 0.500 | 0.000 |
| **I4/mmm-YH** | 4.072      | a = 2.799 Å       | H2          | 0.000  | 0.000 | 0.000 |
|             |                | c = 5.278 Å       | H3          | 0.000  | 0.000 | -0.371 |
| **Im3m-YH** | 3.339          | a = 3.602 Å       | H4          | 0.000  | 0.500 | 0.250 |
|             |                |                   |             |       |      |      |
| **Imm2-YH** | 3.261          | a = 3.281 Å       | Y1          | 0.000  | 0.000 | 0.018 |
|             |                | b = 3.402 Å       | H1          | 0.000  | -0.268 | -0.333 |
|             |                | c = 4.676 Å       | H2          | 0.203  | 0.000 | 0.447 |
|             |                |                   | H3          | -0.263 | 0.000 | 0.384 |
| **P1-YH**  | 3.242          | a = 3.266 Å       | Y1          | 0.123  | 0.334 | -0.351 |
|             |                | b = 5.758 Å       | H1          | 0.087  | 0.343 | 0.262 |
|             |                | c = 5.517 Å       | H2          | 0.111  | 0.003 | -0.338 |
|             |                | α = β = γ = 90 °  | H3          | 0.122  | 0.169 | -0.047 |
|             |                |                   | H4          | -0.169 | 0.094 | 0.465 |
|             |                |                   | H5          | -0.316 | 0.386 | -0.128 |
|             |                |                   | H6          | 0.388  | 0.085 | 0.454 |
| **P1-YH**  | 3.051          | a = 3.119 Å       | Y1          | -0.147 | 0.294 | -0.332 |
|             |                | b = 3.356 Å       | H1          | -0.481 | 0.490 | 0.316 |
|             |                | c = 3.304 Å       | H2          | 0.172  | 0.192 | 0.030 |
|             |                | α = 93.95 °       | H3          | 0.073  | -0.236 | 0.349 |
|             |                | β = 62.19 °       | H4          | -0.481 | -0.223 | -0.059 |
|             |                | γ = 91.91 °       | H5          | -0.045 | -0.198 | -0.104 |
| **Fm3m-YH** | 2.726          | a = 4.931 Å       | Y1          | 0.000  | 0.000 | 0.000 |
|             |                |                   | H1          | 0.378  | 0.378 | 0.378 |
|             |                |                   | H2          | 0.250  | 0.250 | 0.250 |
Raman spectra

Figure S6. Electrode system of the cell K1 at 166 GPa: a) before laser heating; b) after laser heating. The heating area in the center of the culet became black.

Figure S7. Raman spectra of samples of a) M1 cell and b) M3 cell, before and after heating. Raman signals in M3 cell may come from impurities of higher molecular yttrium hydrides.

Figure S8. Diffraction patterns (low intensity) of M1 cell at 165 GPa: a) Le Bail refinement by Im3m-YH₆ and Imm2-YH₇; b) qualitative interpretation of smoothed XRD pattern with subtracted background.
### Elastic properties

**Table S5.** Elastic and thermodynamic parameters of *Im3m*-YH₆ (Z = 2) in the investigated range of pressure.

| Parameter                  | 150 GPa | 165 GPa | 180 GPa |
|----------------------------|---------|---------|---------|
| a, Å                       | 3.602   | 3.573   | 3.546   |
| V_{DFT}, Å³                | 46.73   | 45.61   | 44.59   |
| C₁₁, GPa                   | 637     | 870     | 980     |
| C₁₂, GPa                   | 435     | 460     | 482     |
| C₄₄, GPa                   | 109     | 196     | 283     |
| B, GPa                     | 569     | 597     | 648     |
| G, GPa                     | 109     | 196     | 283     |
| E, GPa                     | 307     | 593     | 705     |
| B/G                        | 5.22    | 3.04    | 2.28    |
| Poisson ratio, η           | 0.410   | 0.352   | 0.308   |
| Density ρ, kg/m³            | 6744    | 6910    | 7069    |
| Transverse sound velocity (vₜ), m/s | 4019   | 5326   | 6354    |
| Longitudinal sound velocity (vₗ), m/s | 10291  | 11145  | 12062   |
| Debye temperature, θₐ, K   | 908     | 1204    | 1438    |
| ω_{log}=0.827θₐ , K        | 751     | 995     | 1189    |

**Table S6.** Elastic and thermodynamic parameters of *I4/mmm*-YH₄ (Z = 2) in the investigated range of pressure.

| Parameter                  | 150 GPa | 165 GPa | 180 GPa |
|----------------------------|---------|---------|---------|
| a, Å                       | 2.779   | 2.752   | 2.724   |
| c, Å                       | 5.277   | 5.259   | 5.236   |
| V_{DFT}, Å³                | 40.758  | 39.829  | 38.866  |
| C₁₁, GPa                   | 800     | 813     | 856     |
| C₁₂, GPa                   | 425     | 406     | 361     |
| C₁₃, GPa                   | 480     | 508     | 607     |
| C₃₃, GPa                   | 1014    | 1082    | 1035    |
| C₄₄, GPa                   | 229     | 234     | 167     |
| C₆₆, GPa                   | 207     | 233     | 218     |
| B, GPa                     | 592     | 601     | 631     |
| G, GPa                     | 214     | 209     | 183     |
| E, GPa                     | 573     | 562     | 501     |
| B/G                        | 2.768   | 2.876   | 3.449   |
| Poisson ratio, η           | 0.338   | 0.344   | 0.367   |
| Density ρ, kg/m³            | 7570    | 7747    | 7939    |
| Transverse sound velocity (vₜ), m/s | 5330   | 5197   | 4816    |
| Longitudinal sound velocity (vₗ), m/s | 10774  | 10658  | 10508   |
| Debye temperature, θₐ, K   | 1115    | 1097    | 1028    |
| ω_{log}=0.827θₐ , K        | 923     | 907     | 850     |
Table S7. Elastic and thermodynamic parameters of \textit{Imm}2-YH$_7$ $(Z = 2)$ in the investigated range of pressure.

| Parameter                  | 150 GPa | 165 GPa | 180 GPa |
|----------------------------|---------|---------|---------|
| a, Å                       | 3.281   | 3.258   | 3.235   |
| b, Å                       | 3.402   | 3.369   | 3.340   |
| c, Å                       | 4.676   | 4.632   | 4.590   |
| $V_{\text{DFT}}$, Å$^3$    | 52.19   | 50.85   | 49.59   |
| $C_{11}$, GPa              | 821     | 847     | 910     |
| $C_{12}$, GPa              | 369     | 374     | 436     |
| $C_{13}$, GPa              | 480     | 505     | 559     |
| $C_{22}$, GPa              | 896     | 923     | 958     |
| $C_{23}$, GPa              | 310     | 334     | 351     |
| $C_{33}$, GPa              | 840     | 881     | 926     |
| $C_{44}$, GPa              | 138     | 140     | 142     |
| $C_{55}$, GPa              | 122     | 136     | 143     |
| $C_{66}$, GPa              | 328     | 407     | 425     |
| $B$, GPa                   | 541     | 563     | 608     |
| $G$, GPa                   | 195     | 212     | 218     |
| $E$, GPa                   | 523     | 566     | 583     |
| $B/G$                      | 2.766   | 2.649   | 2.793   |
| Poisson ratio, $\eta$      | 0.338   | 0.332   | 0.340   |
| Density $\rho$, kg/m$^3$   | 5920    | 6075    | 6228    |
| Transverse sound velocity ($v_t$), m/s | 5760 | 5920 | 5113 |
| Longitudinal sound velocity ($v_l$), m/s | 11650 | 11810 | 12010 |
| Debye temperature, $\theta_D$, K | 1300 | 1345 | 1356 |
| $\omega_{\log} = 0.827\theta_D$, K | 1075 | 1110 | 1122 |
Electronic and phonon properties

Figure S9. Electronic properties of yttrium hydrides: a) density of electronic states of $I4/mmm$-$YH_4$ at $E_F$; b) density of electronic states of $Imm2$-$YH_7$ at $E_F$; c) band structure of $cI4$-$YH_6$ at 150 GPa.

Figure S10. Electronic density of states and band structure of $Imm2$-$YH_7$ at 150 GPa.
Figure S11. Electronic density of states and band structure of $I4/mmm$-YH$_4$ at 150 GPa.

Figure S12. Phonon density of states of a) YH$_4$ and b) YH$_6$ at 150 GPa.
Eliashberg functions and equations for calculating $T_C$ and related parameters

**Figure S13.** Series of Eliashberg functions of YH$_4$ at 165 GPa calculated with different $\sigma$-broadening from 0.005 to 0.05 Ry (marked by colors).

**Figure S14.** Series of Eliashberg functions of YH$_6$ at 165 GPa calculated with different $\sigma$-broadening from 0.005 to 0.05 Ry (marked by colors).
Figure S15. Series of Eliashberg functions of YH\textsubscript{7} at 165 GPa calculated with different σ-broadening from 0.005 to 0.05 Ry (marked by colors).

The critical temperature of superconducting transition was calculated using the Matsubara-type linearized Eliashberg equations: 1

\[
h\omega_j = \pi(2j + 1) \cdot k_B T, j = 0, \pm 1, \pm 2, ...
\]  
(S1)

\[
\lambda(\omega_i - \omega_j) = 2 \int_0^\infty \frac{\omega \cdot a^2F(\omega)}{\omega^2 + (\omega_i - \omega_j)^2} d\omega
\]  
(S2)

\[
\Delta(\omega = \omega_i, T) = \Delta_{\text{mc}}(T)
\]

\[
= \pi k_B T \sum_j \rho \left[ \lambda(\omega_i - \omega_j) - \mu^* \right] K_{ij} \Delta_{\text{mc}}(T)
\]

where \( T \) is the temperature in kelvins, \( \mu^* \) is the Coulomb pseudopotential, \( \omega \) is the frequency in Hz, \( \rho(T) \) is a pair-breaking parameter, the function \( \lambda(\omega_i - \omega_j) \) relates to an effective electron-electron interaction via the exchange of phonons. 2 The transition temperature can be found as the solution of the equation \( \rho(T_C) = 0 \), where \( \rho(T) \) is defined as max(\( \rho \)), provided than \( \Delta(\omega) \) is not a zero function of \( \omega \) at a fixed temperature.

These equations can be rewritten in a matrix form as 3

\[
\rho(T)\psi_n = \sum_{m=0}^{N} K_{mn} \psi_m \Rightarrow \rho(T) \left( \begin{array}{c} \psi_1 \\ \vdots \\ \psi_N \end{array} \right) = \left( \begin{array}{ccc} K_{11} & \ldots & K_{1N} \\ \ldots & K_{ii} & \ldots \\ K_{N1} & \ldots & K_{NN} \end{array} \right) \times \left( \begin{array}{c} \psi_1 \\ \vdots \\ \psi_N \end{array} \right),
\]

where \( \psi_n \) relates to \( \Delta(\omega, T) \), and

\[
K_{mn} = F(m - n) + F(m + n + 1) - 2\mu^* \sum_{l=1}^{m} F(l)
\]

\[
F(x) = F(x, T) = 2 \int_0^{\omega_{\text{max}}} \frac{a^2F(\omega)}{(h\omega)^2 + (2\pi k_B T x)^2} h\omega d\omega,
\]

where \( \delta_{mn} = 1 \) and \( \delta_{mn} = 0 \) (n ≠ m) is a unit matrix. Now we can replace the equation \( \rho(T_C) = 0 \) by the vanishing of the maximum eigenvalue of the matrix \( K_{nm} \): \( \rho = \text{max}_n \text{eigenvalue}(K_{nm}) = f(T) \)\
\( f(T_C) = 0 \).
\[ \beta_{MM} = \frac{d \ln T_C}{d \ln M} = \frac{1}{2} \left[ 1 - \frac{1.04(1 + \lambda)(1 + 0.62\lambda)}{[\lambda - \mu^*(1 + 0.62\lambda)]^2} \mu^* \right] \]  

\[ \beta_{AD} = \beta_{MM} - \frac{2.34\mu^2\lambda^{3/2}}{(2.46 + 9.25\mu^*) \cdot (2.46 + 9.25\mu^*)^{3/2}} \]  
\[ - \frac{130.4 \cdot \mu^2\lambda^2(1 + 6.3\mu^*)}{(8.28 + 104\mu^* + 329\mu^2 + 2.5 \cdot \lambda^2 \frac{\omega_{\text{log}}}{\omega_2})} \left( 8.28 + 104\mu^* + 329\mu^2 + 2.5 \cdot \lambda^2 \frac{\omega_{\text{log}}}{\omega_2} \right)^2 \]  

where the last two correction terms are usually small (~0.01).

The Sommerfeld constant was found as
\[ \gamma = \frac{2}{3} \pi^2 k_B^2 N(0) (1 + \lambda), \]  
and was applied to estimate the upper critical magnetic field and the superconductive gap in yttrium hydrides by well-known semi-empirical equations of the BCS theory (Ref. 4, Equations 4.1 and 5.11), which works for \( T_C/\omega_{\text{log}} < 0.25 \):
\[ \frac{\gamma T_c^2}{H_c^2(0)} = 0.168 \left[ 1 - 12.2 \left( \frac{T_c}{\omega_{\text{log}}} \right)^2 \ln \left( \frac{\omega_{\text{log}}}{3T_c} \right) \right] \]  
\[ \frac{2\Delta(0)}{k_B T_C} = 3.53 \left[ 1 + 12.5 \left( \frac{T_c}{\omega_{\text{log}}} \right)^2 \ln \left( \frac{\omega_{\text{log}}}{2T_c} \right) \right] \]  

The lower critical magnetic field was calculated according to the Ginzburg-Landau theory:
\[ \frac{H_{c1}}{H_{c2}} = \frac{\ln k}{2\sqrt{2k^2}}, k = \frac{\lambda_L}{\xi} \]  

where \( \lambda_L \) is the London penetration depth, found as
\[ \lambda_L = 1.0541 \cdot 10^{-5} \sqrt{\frac{m_e c^2}{4\pi e n_e e^2}} \]  

here \( c \) - is the speed of light, \( e \) - is the electron charge, \( m_e \) - is the electron mass, and \( n_e \) - is an effective concentration of charge carriers, evaluated from the average Fermi velocity \( (V_F) \) in the Fermi-gas model:
\[ n_e = \frac{1}{e\pi^2} \left( \frac{m_e V_F}{\hbar} \right)^3 \]  

The coherence length \( \xi \) was found as \( \xi = \sqrt{\hbar/2e(\mu_0 H_{c2})} \) and was used to estimate the average Fermi velocity
\[ V_F = \frac{\pi \cdot \Delta(0)}{\hbar} \xi \]  

Average Fermi velocity was also calculated directly from the band structure of \( c14-YH_6 \) using
\[ \langle V_F \rangle = \sqrt{\frac{\sum_k \delta(E_k - E_F)V_k^2}{\sum_k \delta(E_k - E_F)}} = \frac{a}{\pi \hbar} \sqrt{\frac{\sum_k \delta(E_k - E_F)(dE_k/dt)^2}{\sum_k \delta(E_k - E_F)}} \]  

where \( dE/dk \) (k = \( -n\pi \) ... \( n\pi \)) were replaced by \( dE/dt \) (t = \( -1\) ... \( 1\)). In this case the first Brillouin zone was sampled by the very dense \( k \)-points mesh with resolution \( 2\pi \times 0.005 \text{ Å}^{-1} \). Obtained \( V_F \) of \( c14-YH_6 \) are 9.1 \( \times \) 10^3, 9.3 \( \times \) 10^3 and 9.4 \( \times \) 10^3 m/s at 150, 165 and 180 GPa, respectively.
Table S8. Additional electronic and superconducting parameters of yttrium hydrides at 165 GPa and $\mu^*=0.15-0.1$

| Parameter                           | $I4/mmm$-YH₄ | $Im\bar{3}m$-YH₆ | Imm2-YH₇ |
|-------------------------------------|---------------|------------------|---------|
| $N(E_F)$, states/eV/f.u.            | 0.475         | 0.7075           | 0.07    |
| Average Fermi velocity ($V_F$)*     | 3.3-3.8$\times10^5$ | 5.5-5.9$\times10^5$ | 3.1-3.8$\times10^5$ |
| London penetration depth ($\lambda_L$), nm | 188-152       | 87-79            | 208-155 |
| Coherence length, Å                  | 45-40         | 23-22            | 130-110 |
| Ginzburg-Landau parameter, $\kappa$ | 42-38         | 38-36            | 16-14.2 |
| Lower critical magnetic field ($\mu_0H_{C1}$), mT | 12-18         | 55-67            | 7.5-13  |
| Upper critical field ($\mu_0H_{C2}$), T | 16-20         | 64-68            | 2.7     |
| Clogston-Chandrasekhar paramagnetic limit, T | 189-244       | 622-691          | 61-88   |
| Critical current density ($J_c$), A/cm² | 1.1-1.86$\times10^8$ | 9.9-12.5$\times10^8$ | 3-6.5$\times10^7$ |

*According to formula (S15)

Calculations of electron-phonon interaction in cubic YH₆ at 174 GPa yield $\lambda = 2.24$, $\omega_{\text{log}} = 929$ K, $T_C \sim 220-240$ K, and $\mu_0H_{C2}(\theta) = 64-68$ T (Tables 3, S5). Estimation of the average Fermi velocity $V_F \sim 5.5-5.9\times10^5$ m/s makes it possible to calculate the London penetration depth $\lambda_L \sim 79$ nm, coherence length $\xi_{\text{BCS}} = 22$ nm, and lower critical magnetic field $\mu_0H_{C1} = 0.067$ T. The critical current density ($J_c = en_vV_L$), evaluated by the Landau criterion for superfluidity $^5$ $V_L = min \frac{\varepsilon(p)}{p} \equiv \frac{\Delta_0}{\hbar k_F}$, was calculated to be up to $10^9$ A/cm², much higher than in H₃S $^6$. The calculated Ginzburg-Landau parameter $^7$ is over 36 which is typical for II type superconductors.

It is interesting to note that recalculation of superconducting parameters of YH₆ using Eliashberg functions $\alpha^2F(\omega)$ from Ref. $^9$ (Ref. from the main text) leads to the $T_C$ (A-D) of 233 K ($\omega_2 = 1126$ K), 215 ($\omega_2 = 1640$ K) and 238 K ($\omega_2 = 1936$ K) at 100, 200 and 300 GPa, respectively. Thus, critical temperatures obtained using the simple Allen-Dynes formula $^16$ (Ref. from the main text) surprisingly turn out to be more successful approximation to the experiment. Systematic underestimation of $T_C$ by Allen-Dynes formula compared to the numerical solution of the Eliashberg equations turns out to be approximately equal to the $T_C$ correction due to anharmonic effects.
References for Supporting Information

1. Eliashberg, G. M. Interactions between Electrons and Lattice Vibrations in a Superconductor. *JETP* **11**, 696–702 (1959).
2. Bergmann, G. & Rainer, D. The sensitivity of the transition temperature to changes in $\alpha^2F(\omega)$. *Z. Für Phys.* **263**, 59–68 (1973).
3. Allen, P. B. & Dynes, R. C. A computer program for numerical solution of the Eliashberg equation to find $T_c$. *Tech. Rep. 7 TCM41974* (1974).
4. Carbotte, J. P. Properties of boson-exchange superconductors. *Rev. Mod. Phys.* **62**, 1027–1157 (1990).
5. Landau, L. Theory of the Superfluidity of Helium II. *Phys. Rev.* **60**, 356–358 (1941).
6. Drozdov, A. P., Eremets, M. I., Troyan, I. A., Ksenofontov, V. & Shylin, S. I. Conventional superconductivity at 203 kelvin at high pressures in the sulfur hydride system. *Nature* **525**, 73–76 (2015).
7. Ginzburg, V. L. & Landau, L. D. On the Theory of superconductivity. *Zh.Eksp.Teor.Fiz.* **20**, 1064–1082 (1950).