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Synthesis and superconductivity in yttriumcerium hydrides at high pressures

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Liu-Cheng Chen 1,2 , Tao Luo^{1,2}, Zi-Yu Cao 2,3 , Philip Dalladay-Simpson 2,3 , Ge Huang², Di Peng 2,2 , Li-Li Zhang⁴, Federico Aiace Gorelli^{2,5}, Guo-Hua Zhong 6,7 , Hai-Qing Lin⁸ & Xiao-Jia Chen 9

Further increasing the critical temperature and/or decreasing the stabilized pressure are the general hopes for the hydride superconductors. Inspired by the low stabilized pressure associated with Ce 4f electrons in superconducting cerium superhydride and the high critical temperature in yttrium superhydride, we carry out seven independent runs to synthesize yttrium-cerium alloy hydrides. The synthetic process is examined by the Raman scattering and X-ray diffraction measurements. The superconductivity is obtained from the observed zero-resistance state with the detected onset critical temperatures in the range of 97-141 K. The upper critical field towards 0 K at pressure of 124 GPa is determined to be between 56 and 78 T by extrapolation of the results of the electrical transport measurements at applied magnetic fields. The analysis of the structural data and theoretical calculations suggest that the phase of $Y_{0.5}Ce_{0.5}H_9$ in hexagonal structure with the space group of $P6_3/mmc$ is stable in the studied pressure range. These results indicate that alloying superhydrides indeed can maintain relatively high critical temperature at relatively modest pressures accessible by laboratory conditions.

Rare earth hydrides have been found to exhibit near room temperature superconductivity benefiting from the chemical pre-compression induced by the interaction between hydrogen and tetragen atoms^{1,2}. A typical example among them is the experimental discoveries of superconductivity at the critical temperature T_c as high as 250–260 K at pressure of -170 GPa^{3,4} for clathrate LaH₁₀ with *Fm*3*m* structure^{4,5} based on the early theoretical predictions^{6,7}. In parallel, yttrium hydrides as another attractive rare earth hydride system have drawn a lot of attentions from the predictions^{6–8} and experimental realization^{9–12} of superconductivity. The experiments reported the *I*4/*mmm*-YH₄ phase with the maximum T_c of 88 K at 155 GPa⁹, the *Im*3*m*-YH₆ phase with a similar T_c near 220 K in the pressure range of 166–180 GPa^{10,11}, and the

*P*63/*mmc*-YH₉ phase with the maximum $T_c \sim 243$ K at 201 GPa¹⁰ or with the significantly high maximum $T_c \sim 262$ K at 182 GPa¹². It is clear that the experimentally obtained YH_x superconductors are only stable at extreme pressures (above ~155 GPa)^{9–12}. Meanwhile, theoretical^{6,13–15} and experimental^{13,14} efforts established that cerium superhydrides can be stabilized at low pressures below 100 GPa but can be superconductive as well. The enhanced chemical pre-compression in CeH₉ was suggested to be associated with the delocalized nature of Ce 4*f* electrons¹⁶. Recently, cerium superhydrides were found to exhibit superconductivity with the maximum T_c of 115 K for $Fm\bar{3}m$ -CeH₁₀ at 95 GPa and T_c of 57 K for $P6_3/mmc$ -CeH₉ at 88 GPa¹⁷. This finding was confirmed for CeH₁₀ with even higher T_c values at the corresponding modest

¹School of Science, Harbin Institute of Technology, Shenzhen 518055, China. ²Center for High Pressure Science and Technology Advanced Research, Shanghai 201203, China. ³Center for Quantum Materials and Superconductivity (CQMS) and Department of Physics, Sungkyunkwan University, Suwon 16419, Republic of Korea. ⁴Shanghai Synchrotron Radiation Facility, Shanghai Advanced Research Institute, Chinese Academy of Sciences, Shanghai 201204, China. ⁵National Institute of Optics (INO-CNR) and European Laboratory for Non-Linear Spectroscopy (LENS), Via N. Carrara 1, 50019 Sesto Fiorentino (Florence), Italy. ⁶Shenzhen Institute of Advanced Technology, Chinese Academy of Sciences, Shenzhen 518055, China. ⁷University of Chinese Academy of Sciences, Beijing 100049, China. ⁸School of Physics, Zhejiang University, Hangzhou 310058, China. ⁹Department of Physics and Texas Center for Superconductivity, University of Houston, Houston, TX 77204, USA. Second and the second sec

pressures¹⁸. Meanwhile, the superconductivity in this superconductor has been found to follow the Bardeen-Cooper-Schrieffer theory with a moderate coupling strength from the determination of its superconducting gap¹⁸. Unlike La-H and Y-H systems, the stabilized pressure is dramatically reduced in the Ce-H compounds.

The exploration of high- T_c superconductivity in superhydrides at low pressures or even ambient pressure is highly demanded. Compared with binary hydrides, ternary alloy hydrides possess diverse chemical compositions and thus provide more abundant structures for the operation with the advantages of different elements¹⁹⁻²². Interestingly, a series of lanthanum-yttrium ternary hydrides was reported to possess superconductivity with the maximum T_c of 253 K at pressures of 170-196 GPa²³, indicating that hydrides can be stabilized in solid solutions at relatively low pressures. Superconductivity in lanthanumcerium ternary superhydrides with $T_c \sim 176$ K was strikingly preserved to ~100 GPa^{24,25}. Theoretical calculations demonstrated that a series of ternary superhydrides can hold high T'_c s at relatively low stabilized pressures (~50 GPa)^{19,21}. Therefore, exploring superconductivity in ternary hydrides towards high T_c at modest pressures from the experimental side is an interesting direction. For such a purpose, we choose Y_{0.5}Ce_{0.5} alloy and synthesize such alloy hydrides with the combined advantages of the low synthesized pressure in Ce-H and high T_c in Y-H compounds.

Results and discussions

Synthetic process of Y_{0.5}Ce_{0.5} hydrides

The $Y_{0.5}Ce_{0.5}$ pieces with a thickness of about $1-2 \mu m$ were loaded into the sample chamber with ammonia borane NH₃BH₃ (AB) as the pressure transmitting medium and hydrogen source (Fig. 1a). The samples were heated around 2000 K from the direction of AB side by an YAG laser after compressing to a desirable pressure value (Supplementary Table 1). After laser heating, all the samples (Cell-1 to Cell-7) change dramatically in their shapes and colors (Fig. 1b and Supplementary Fig. 1). Raman spectroscopy is used to detect the vibrational properties of various phases and to identify the formation of phase after the chemical reaction. To eliminate the scattering signals from AB, *c*-BN, and PtH, the direction of the exciting laser for the Raman scattering is from the alloy side (Top panel of Fig. 1a). The Raman vibron (v_1) of H₂ is detected with large intensity for the sample after heating, indicating the presence of hydrogen in favor of the formation of Y_{0.5}Ce_{0.5} hydrides (Fig. 1c and Supplementary Fig. 2). The scaled Raman spectra at low frequencies (-300–1000 cm⁻¹) display a hump, which consists of the rotational bands of H₂, named S(0) and S(1), and the optical phonon associated to the transverse optical E_{2g} mode^{26,27}. The Raman bands marked by asterisks at lower frequencies should be the phonon modes from the synthesized alloy hydrides, because of the absence of any Raman active modes for H₂ in this region at the studied pressure^{26,27}. Thus, we can conclude that the chemical reaction takes place in the sample chamber judging from the micrographs and Raman spectra (Fig. 1 and Supplementary Figs. 1 and 2).

Superconductivity under pressure

To probe superconductivity in the synthesized $Y_{0.5}Ce_{0.5}$ alloy hydrides, we performed the electrical transport measurements under pressure. Representative temperature-dependent resistance data at high pressures are shown in Fig. 2a. Superconducting transitions can be clearly seen, as evidenced by the sharp drop of the resistance occurring at 129, 113, and 136 K at pressure of about 114, 195, 124 GPa for Cell-3, Cell-6, and Cell-7, respectively. In the resistance measurements, the zero-resistance state was realized for the samples in Cell-3, Cell-6, and Cell-7. In Supplementary Fig. 3, we showed the results of the temperature-dependent resistance measurements on Y_{0.5}Ce_{0.5} alloy before heating and Y_{0.5}Ce_{0.5} hydrides after heating in Cell-6. The large T_c differences before and after heating also indicate the high- T_c superconductivity is from the synthesized alloy hydrides. By putting the determined T_c values together from the temperature-dependent resistance measurements (Fig. 2a and Supplementary Figs. 3-8), we obtain the systematic evolution of T_c with pressure of synthesized Y_{0.5}Ce_{0.5} hydrides (Fig. 2b). With the increase of pressure, T_c is found to increase with pressure and then slightly decrease after passing the maximum of 141 K at an optimal pressure of around 130 GPa.





red lines, and the blue arrows in the right photos indicate the parts with apparent changes after heating. **c** Raman spectra for the synthesized $Y_{0.5}Ce_{0.5}$ hydrides collected at the apparent-changing parts (blue arrows in the right Fig. 1b) in the sample chamber. The Raman bands of the diamond and H₂ after laser heating are presented. The low-frequency Raman spectra are scaled for the clarity.



Fig. 2 | Temperature-dependent resistance measurements on the synthesized $Y_{0.5}Ce_{0.5}$ hydrides in the representative cells. a Superconducting transitions at various pressures were indicated by the arrows. b Pressure dependence of T_c for $Y_{0.5}Ce_{0.5}H_9$ with the comparison of YH₉ and CeH₉ with the same crystal structure in

the literature^{10,12,17,18}. The sold symbols with different colours represent the results from six runs in the current work. The open symbols denote the data points for YH₉ from the works^{10,12} and for CeH₉ from the works^{17,18}, respectively.





To further obtain the other superconducting properties, we performed the electrical transport measurements at external magnetic fields for sample 7 (Cell-7) at pressure of 124 GPa (Fig. 3). Supplementary Fig. 8a shows the Raman spectra of diamond used for the pressure calibration²⁸. As shown in Fig. 3a, with the application of magnetic field up to 9 T, the resistance gradually shifts to lower temperatures with the widening of the superconducting transition. The suppression of superconductivity by the applied fields can be clearly observed. These phenomena are the character of superconductivity. The temperature dependence of the upper critical field μ_0H_{c2} can be obtained from such measurements (Fig. 3b). The upper critical field is usually described by the Ginzburg-Landau (GL) equation²⁹ and the Werthamer-Helfand-Hohenberg (WHH) equation³⁰.

The upper critical field $\mu_0 H_{c2}(T)$ at temperature of 0 K is extrapolated to be 56 and 78 Tesla from the GL and WHH model fitting, respectively. The achievement of the zero-resistance state together with the downward shift of T_c and gradual widening of the superconducting transition with applied magnetic fields demonstrate the superconductivity in synthesized samples of Y_{0.5}Ce_{0.5} hydrides. The coherence length ξ can be roughly calculated via the equation of $\mu_0H_{c2}(0) = \phi_0/2\pi\xi^2$, where ϕ_0 is the magnetic flux quantum. By using the obtained $\mu_0H_{c2}(0)$ of 56-78 Tesla, one can roughly estimate ξ to be 20-25 Å. The high upper critical fields and short coherence lengths suggest the type-II character of the synthesized superconductors.

Structure characterization

Synchrotron x-ray diffraction (XRD) measurements were carried out on sample 1 (Cell-1) and sample 5 (Cell-5) to determine their structural properties. For sample 1, the representative XRD patterns are given in Fig. 4a for Point-1 and Supplementary Fig. 9 for Point-2, respectively, at pressure of 128 GPa determined from the Raman spectrum of the used



Fig. 4 | **Synchrotron x-ray diffraction patterns of sample 1 at pressure of 128 GPa (Point-1). a** The experimental data along with the model fitting for the phases of $P6_3/mmc$ - $Y_{0.5}Ce_{0.5}H_9$ and $P6_3/mmc$ -PtH. The experimental data points, calculated values, and Bragg peak positions are shown by the small open circles, thin curves, and vertical sticks, respectively. Top panel shows the cake view of the raw

XRD patterns. The red point in the inset photograph displays the corresponding sample position of the collected patterns. The marked peak with asterisk represents the unidentified reflection. **b** Schematic diagram of the crystal structure of P_{6_3}/mmc ·Y_{0.5}Ce_{0.5}H₉.

diamond anvil (Supplementary Fig. 10). Comparing the structural features of the similar system in the literature^{9,10,12,17,25}, we have the phases with the structures in the space groups of *P*6₃/*mmc*, *I*4/*mmm*, and *C*2/*m* for the syntheized products. As shown in Fig. 4a, the observed peaks collected at the red dot in the inset can be indexed by the *P*6₃/*mmc* phase with the help of the Le Bail method³¹. The fitted lattice parameters are *a* = 3.653(5) Å, *c* = 5.476(5) Å, and *V* = 63.33(4) Å³ (Supplementary Table 2). The weak diffraction peaks at larger angles are from PtH-II (*P*6₃/*mmc*) as reported previously³², which is common in the chemical reaction of Pt leads with hydrogen^{32,33}. Other representative XRD patterns (Supplementary Fig. 9) correspond to three Y_{0.5}Ce_{0.5} hydride phases with the space groups of *P*6₃/*mmc*, *I*4/*mmm*, and *C*2/*m*, and the Pt hydride phase of *P*6₃/*mmc*.

To draw more structural information, Cell-5 with large diffraction angles was prepared for synchrotron XRD measurements. Typical results are given in Supplementary Figs. 11-13. By indexing the observed peaks of Sample 5 at 140 GPa, we can identify the appearance of three phases with the P63/mmc, I4/mmm, and C2/m structure for Y_{0.5}Ce_{0.5} alloy hydrides and the P6₃/mmc PtH-II. The XRD patterns of pure Y_{0.5}Ce_{0.5} phase and pure PtH-II phase in Cell-5 at 140 GPa were also collected (Supplementary Fig. 13). Although the accurate occupancy of the hydrogen atoms can not be determined experimentally owing to the weak x-ray scattering cross sections, the hydrogen concentration (x) can be obtained through the cell unit volume ($V_{lattice}$) by using the formula $x = (V_{lattice} - V_Y - V_{Ce})/V_H/2$, where V_Y , V_{Ce} , V_H are the volumes of Y, Ce, and H atoms, respectively. The V_Y , V_{Ce} , and V_H can be estimated from their elemental phases (Supplementary Fig. 14)³⁴⁻³⁶ because of the absence of the atomic volume at high pressures. As a result, the hydrogen content of the synthesized P63/mmc phase of Y_{0.5}Ce_{0.5} alloy hydrides is in the range of 8.3-8.8 (Supplementary Table 2). The calculated hydrogen concentration has a small deviation with the ideal value of 9, because of the direct use of the elemental phases for the evaluation of volumes.

Superconducting phase from theoretical calculations

To identify the obtained superconducting phase, we calculated the electronic, phononic, and superconducting properties of $P6_3/mmc$ - $Y_{0.5}Ce_{0.5}H_9$ in terms of the density functional theory (Fig. 5). The minimum pressure that can theoretically stabilize the structure $P6_3/mmc$ - $Y_{0.5}Ce_{0.5}H_9$ is 180 GPa. As shown in Fig. 5a, the $P6_3/mmc$ structure

is metallic and shows a large density of states (DOS) near the Fermi level E_{F} . The large DOS is in favor of high- T_c superconductivity. The contributions of the electronic states of H near E_F usually plays an important role for the hydride superconductor. From the partial DOS shown in Fig. 5a, we can see that the large DOS near E_F is dominant from the contribution of the H-*s*, Ce-*f*, and Y-*d* orbitals. In detail, the contributions of the electronic states of H-*s*, Ce-*f* near E_F are both 0.37 states/eV. The large contribution of Ce-*f* indicates that the delocalized nature of Ce-*f* electrons is responsible for the enhanced chemical precompression in Y_{0.5}Ce_{0.5} hydrides.

The calculated phonon dispersion, phonon density of states (PHDOS), Eliashberg function $\alpha^2 F(\omega)$, and electron-phonon coupling (EPC) constant $\lambda(\omega)$ of $P6_3/mmc$ -Y_{0.5}Ce_{0.5}H₉ are given in Fig. 5b. The optical phonon modes at high frequencies (above ~390 cm⁻¹) are derived from the H atoms. The acoustic phonon modes with frequencies lower than ~180 cm⁻¹ and the low-frequency optical modes between 145 and 320 cm⁻¹ are mainly attributed to the contributions of Y and Ce atoms. From the phonon frequency dependence of $\alpha^2 F(\omega)$ and integrated EPC constant $\lambda(\omega)$, we find that the contributions to $\alpha^2 F(\omega)$ and $\lambda(\omega)$ arise from all three phonon modes including the Y-derived acoustic, the Ce-derived acoustic, and the H-derived optical modes. In addition, the wide high-frequency $\alpha^2 F(\omega)$ is significantly higher than those on the narrow low-frequency side. Obviously, the high-energy H-derived vibrations dominate the total $\lambda(\omega)$ value. The calculations give a high EPC constant λ of 2.23 at 180 GPa. By numerically solving the Allen-Dynes-modified McMillan formula with the Coulomb pseudopotential parameter μ^* of 0.1–0.15^{37,38}, we obtained T_c of 104–119 K (Supplementary Table 3), in fair agreement with our experiments. The absence of the imaginary parts of the phonon spectrum (Fig. 5b) along with the comparable T_c values with experiments supports the obtained superconducting phase of P63/ mmc-Y0.5Ce0.5H9.

Compared the synthetic conditions, stable pressures, and T_c values of P_{6_3}/mmc ·Y_{0.5}Ce_{0.5}H₉ with YH₉ and CeH₉ (Fig. 2b and Supplementary Table 4), we found that the synthetic pressure and minimum stable pressure of Y_{0.5}Ce_{0.5}H₉ are much lower those of YH₉^{10,12}, and the T_c values are overall higher than those of CeH₉^{17,18} at around the same pressure level. Y_{0.5}Ce_{0.5}H₉ possesses the maximum of T_c of 141 K at the optimal pressure of about 130 GPa. This T_c value is apparently higher than all the reported values for CeH₉^{17,18}. The optimal pressure is



Fig. 5 | The electronic and phononic properties of $P6_3/mmc Y_{0.5}Ce_{0.5}H_9$ at 180 GPa. a Band structure, total and particial electron density of states (DOS). The horizontal line shows the location of the Fermi level. **b** Calculated phonon

dispersion, phonon density of states (PHDOS) projected onto the Y, Ce, and H atoms, Eliashberg function $\alpha^2 F(\omega)$, and integrated electron-phonon coupling (EPC) constant $\lambda(\omega)$.

much lower than those of $YH_9^{10,12}$ from two sets of data points in independent runs. $Y_{0.5}Ce_{0.5}H_9$ thus serves an excellent example for the successful realization of high- T_c -superconductivity in ternary alloy hydrides. The comparison of our results for $Y_{0.5}Ce_{0.5}H_9$ with those for (La,Ce)H₉ and La_{0.5}Ce_{0.5}H₁₀ taken from the recent works^{25,33} was given in Supplementary Fig. 15 as well. It is clear that the hydrides based on the alloy including Ce and other rare-earth element can substantially reduce the optimal pressure while keeping higher T_c compared to Cebased hydrides.

In summary, we have successfully synthesized $Y_{0.5}Ce_{0.5}$ hydrides by laser heating at high pressures in seven runs (Cell-1 to Cell-7). The synthesized $Y_{0.5}Ce_{0.5}H_9$ with high T_c of 97–141 K in the pressure range of 98–200 GPa was comprehensively studied by the measurements of the electrical transport, x-ray diffraction, and Raman scattering, and theoretical calculations. Compared with the binary cerium hydrides (CeH₉)^{17,18}, this half-replaced ternary alloy superhydride ($Y_{0.5}Ce_{0.5}H_9$) possesses significantly high T_c values. Our findings point out the promising direction for exploring high T_c superconductivity under pressure in alloy hydrides. In addition, the detailed synthesis process of the yttrium-cerium hydrides hopes to help subsequent investigations for other hydrides.

Methods

Sample synthesis

The $Y_{0.5}Ce_{0.5}$ alloy ingots were prepared by melting pure Y (99.9%, Alfa Aesar) and Ce (99.8%, Alfa Aesar) in an arc-melting machine under a Tigettered high purity argon atmosphere. To ensure uniformity of alloy ingot, we flipped each ingot and melted it at least six times. $Y_{0.5}Ce_{0.5}$ hydrides were synthesized by using a series of symmetric diamond-anvil cells (DACs), which have diamonds bevelled at 8–8.5° to a

diameter of $300 \,\mu\text{m}$ with a culet size of $60\text{--}100 \,\mu\text{m}$. In detail, we employed the ammonia borane (NH₃BH₃ or AB) as the hydrogen source, which has been successfully used in several recent studies^{3,17}. When being completely dehydrogenated, one mole of AB can yield three moles of H₂ along with the insulating c-BN by the decomposing reaction $NH_3BH_3 \rightarrow 3H_2 + c \cdot BN^{39,40}$. The $Y_{0.5}Ce_{0.5}$ pieces were cut into 15-30 μ m in diameter and 1-2 μ m thick. The sheared pieces and AB were then layered into the sample chamber. To isolate the surrounding atmosphere, we handled all the samples in a glove box with the residual contents of O₂ and H₂O of <0.1 p.p.m. One-side laser-heating experiments were performed using a yttrium-aluminum-garnet (YAG) laser, after the pressure was increased to the desirable value. The heating temperature is between 1500-2000 K (Supplementary Table 1) and the exposure time is 10-20 min. In our experiments, we used the laser power of 5 mW to focus on the sample with the laser spot around 1.5 micrometer.

Raman scattering measurements

Raman scattering measurements were used to characterize the synthesizing processes of $Y_{0.5}Ce_{0.5}$ hydrides. The measurements were performed with the exciting laser (488 nm) and the beam size of the laser of about 3μ m. The scattered light was focused on a 300 and 1800 g/mm grating and recorded with a 1300 pixel charge-coupled device designed by Princeton Instrument. In all the experiments, the pressure was determined using the Raman shift of the stressed edge of the diamond peak²⁸.

Electrical transport measurements

A typical diamond anvil cell made from Cu-Be alloy was used for the electronic transport measurements at external magnetic fields from

0 to 9 T. Four Pt wires were adhered between the $Y_{0.5}Ce_{0.5}$ alloy and AB in the sample hole with silver epoxy (Supplementary Fig. 1) by the Van Der Pauw method⁴¹ to measure the in-plane resistance at high pressures. This technique was successfully used to measure the resistivity and Hall resistivity in the study of superconductivity for materials in hydrostatic or quasi-hydrostatic pressure environments^{18,33,42–46}. In the present resistance measurements, the rhenium gaskets insulated by the mixture of cubic boron nitride (*c*-BN) and epoxy were used to contain the sample at megabar pressures while isolating the platinum (Pt) electrical leads. The whole progress was performed in the glove box. Since hydrogen produced from the decomposed AB serves as the pressure transmitting medium, the technique developed here and previously^{18,33} for hydrides ensures the resistance measurements in the excellent hydrostatic pressure environments at the studied pressures.

X-ray diffraction measurements

The structural characterization was carried out at Shanghai Synchrotron Radiation Facility with a wavelength 0.6199 Å and the size of the x-ray focus beam is less than 2μ m. Considering the possible inhomogeneity of the synthesized size, we collected the synchrotron XRD patterns with 5μ m step across the culet. The obtained twodimensional XRD patterns are converted to one dimensional diffraction data with the help of Dioptas⁴⁷. The data was analyzed by using the software of Jana based on the Le Bail method^{31,48}. The fitted lattice parameters are summarized in the Supplementary Information.

Theoretical calculations

For the theoretical calculations, the structure was fully relaxed before calculating the electronic band structures. The electronic band structures were calculated by using Vienna ab initio simulation package $(VASP)^{49,50}$ with the density functional theory $(DFT)^{51,52}$. The generalized gradient approximation (GGA) of Perdew-Burke-Ernzerhof (PBE) functional was used for the exchange-correlation functional⁵³. The cutoff energy of the plane-wave basis was set to 400 eV. The phonon spectrum, electron-phonon coupling (EPC) parameters, and T_c were carried out using the QUANTUM ESPRESSO (QE) package⁵⁴ by employing the plane wave pseudopotential method and PBE exchange-correlation functional⁵³. The selected *k*-point mesh in this part is $12 \times 12 \times 12$, and the *q*-point mesh is $3 \times 3 \times 3$.

Data availability

The data that support the findings of this study have been included in the plotted figures and listed tables in the main text and Supplementary Information. If any additional information or any extra data will be required in order to reproduce the results reported in this work, please contact the corresponding author.

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Author contributions

X.J.C. designed and supervised the whole study. D.P. synthesized the $Y_{0.5}Ce_{0.5}$ alloy. L.C.C., Z.Y.C., and G.H. synthesized Y-Ce-H samples. L.C.C. performed high-pressure resistance and Raman scattering measurements under the guidance of X.J.C. L.C.C. and L.L.Z. collected the XRD data. T.L., G.H.Z., and H.Q.L. carried out the theoretical calculations. P.D.S. and F.A.G. help to heat the Cell-1. L.C.C. and X.J.C. analyzed the data and wrote the paper with the inputs from other authors. All authors discussed the results. The manuscript reflects the contributions of all authors.

Competing interests

The authors declare no competing interests.

Additional information

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Correspondence and requests for materials should be addressed to Xiao-Jia Chen.

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