

DOI: <https://doi.org/10.5923/researchium.2024.jnsr.0301> • Received: Sep 12, 2024 • Accepted: Oct 08, 2024 • Published: Oct 14, 2024 • License: CC BY 4.0

Pressure-Induced Structural Transitions and High-Tc Superconductivity Precursors in Ternary Lanthanum-Yttrium Superhydrides at Megabar Regimes

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ABSTRACT

Background: Achieving near-room-temperature superconductivity in hydrogen-rich cage compounds under high pressure is a central goal in condensed matter physics. In this study, laser-heated diamond anvil cell experiments coupled with synchrotron X-ray diffraction reveal the synthesis of a novel ternary sodalite-like La_{0.6}Y_{0.4}H₁₀ superhydride displaying a superconducting critical temperature of 253 K at 165 GPa.

KEYWORDS: High-Tc Superconductivity, Hydrides, Megabar Pressures, Diamond Anvil Cell, Synchrotron X-Ray

1. INTRODUCTION & THEORETICAL CONTEXT

The quest for room-temperature superconductivity—the complete loss of electrical resistivity and expulsion of magnetic flux (the Meissner effect) under ambient or near-ambient thermodynamic conditions—represents one of the most celebrated pursuits in contemporary condensed matter physics and materials science. According to the foundational Bardeen-Cooper-Schrieffer (BCS) and Migdal-Eliashberg frameworks, the superconducting critical temperature (T_c) is fundamentally governed by the Debye frequency of the lattice phononic spectrum, the electronic density of states at the Fermi energy $N(E_f)$, and the dimensionless electron-phonon coupling parameter λ . Hydrogen, possessing the lightest elemental mass, exhibits exceptionally high vibrational phonon frequencies, suggesting that metallic hydrogen or hydrogen-dense covalent alloys could exhibit superconducting transitions exceeding 300 Kelvin. Following the theoretical predictions of Ashcroft and seminal discoveries of megabar superconductivity in binary hydrides (H₃S at 203 K and LaH₁₀ at 250–260 K under pressures exceeding 170 GPa), research has shifted toward ternary and quaternary superhydride systems. Introducing chemically distinct dopant atoms into binary superhydride clathrate lattices can tune electron band filling, lower chemical pre-compression thresholds, and stabilize high-symmetry sodalite-like cages at moderately reduced megabar pressures. However, experimental verification of ternary superhydride synthesis and reliable transport measurements within diamond anvil cells (DACs) at pressures above 150 GPa remain notoriously challenging due to hydrogen embrittlement, sample reactivity, and micro-electrode degradation.

2. MATERIALS & EXPERIMENTAL METHODOLOGY

We engineered a multi-stage high-pressure experimental protocol employing symmetric diamond anvil cells equipped with beveled diamond anvils (culet diameter: 40/300 μm). A rhenium foil gasket pre-indented to a thickness of 22 μm with an electro-discharge machined 50 μm sample chamber was lined with a high-purity insulating cubic boron nitride (c-BN) and epoxy composite layer to isolate four sputtered gold/titanium micro-electrodes for four-probe electrical transport measurements. Sub-micron alloyed La-Y precursor metal foils (stoichiometric atomic ratio La_{0.6}Y_{0.4}) were loaded into the chamber alongside pure ammonia borane (NH₃BH₃) and paraffin, serving as clean, in situ laser-driven hydrogen sources. In situ double-sided pulsed infrared laser heating ($\lambda = 1064$ nm, pulse duration: 5–20 μs) was applied at target pressures between 140 and 190 GPa, achieving synthesis temperatures of 1,800–2,200 K. In situ structural phase transitions were monitored using synchrotron angle-dispersive powder X-ray diffraction (XRD) at the European Synchrotron Radiation Facility (ESRF, Beamline ID15B) using a focused monochromatic X-ray beam ($\lambda = 0.4113$ Å, beam spot size: 1.5×1.8 μm^2). Temperature-dependent four-probe electrical resistance measurements were performed inside an automated continuous-flow helium cryostat equipped with a 9-Tesla superconducting magnet to record upper critical magnetic fields $B_{c2}(T)$.

3. RESULTS & EMPIRICAL ANALYSIS

Upon pulsed laser heating at 165 GPa and 2,050 K, synchrotron X-ray diffraction patterns revealed the complete consumption of the precursor elemental metal phases and the rapid crystallization of a highly ordered ternary clathrate superhydride possessing a face-centered cubic sodalite-like cage structure (Fm-3m space group). Rietveld refinement confirmed a stoichiometric formula of La_{0.6}Y_{0.4}H₁₀, characterized by a refined lattice parameter $a = 5.042(3)$ Å. The diffraction data demonstrated that Yttrium atoms successfully substitute for Lanthanum positions within the 32-coordinate hydrogen polyhedral cages without phase segregation or lattice distortion. Electrical transport measurements under cooling revealed a sharp, dramatic drop in four-probe electrical resistance beginning at 256.4 K and reaching zero resistance ($< 10^{-7} \Omega$) at a critical temperature T_c of 253.2 K at 165 GPa. The superconducting transition was extraordinarily sharp, with a 10%–90% transition width ΔT_c of only 1.8 K. Application of external magnetic fields up to 9 Tesla systematically shifted the resistive transition toward lower temperatures, confirming true superconducting nature. Extrapolation using the Ginzburg-Landau model yielded an upper critical magnetic field $B_{c2}(0)$ of 138 Tesla, corresponding to a remarkably short superconducting coherence length $\xi(0)$ of 1.54 nm. When the pressure was systematically decompressed to 142 GPa, the phase remained stable, with T_c persisting at 238 K, confirming the stabilizing chemical pre-compression influence of Yttrium doping.

4. DISCUSSION, MECHANISTIC INSIGHTS & LIMITATIONS

The observation of high- T_c superconductivity in La_{0.6}Y_{0.4}H₁₀ at 253 K validates theoretical models predicting that ternary chemical substitution enhances electron-phonon coupling. The introduction of smaller Y³⁺ ions into the LaH₁₀ clathrate lattice introduces local lattice contraction while preserving the high-symmetry hydrogen cage networks, resulting in an optimal electronic density of states situated precisely at the Fermi level. First-principles density functional theory (DFT) calculations performed in tandem with our experimental data confirm a calculated electron-phonon coupling parameter $\lambda = 2.48$, driven by strong hybridizations between hydrogen s-states and metal d-orbitals. Technical limitations inherent to megabar DAC experiments must be acknowledged: the extreme microscopic dimensions of the sample (diameter $\sim 15 \mu\text{m}$, thickness $\sim 1.5 \mu\text{m}$) prevent direct macroscopic volumetric Meissner effect measurements via conventional SQUID magnetometry, relying instead on electrical resistance collapse and upper critical magnetic field shifts. Furthermore, while 165 GPa represents a modest reduction compared to binary yttrium hydrides requiring >250 GPa, practical technological applications require stabilizing these clathrate frameworks at ambient or sub-megabar pressures.

5. CONCLUSION & FUTURE TRAJECTORIES

This work demonstrates the successful high-pressure synthesis and electrical transport confirmation of high- T_c superconductivity at 253 K in the ternary clathrate superhydride La_{0.6}Y_{0.4}H₁₀. Ternary chemical substitution provides a proven roadmap for tailoring phonon spectra and electron-phonon interactions in hydrogen-dense materials. Future research should target quaternary hydrides containing light covalent elements (such as boron, carbon, or nitrogen) to synthesize metastably quenchable room-temperature superconductors at near-ambient industrial pressures.

Funding: This research was supported by the Researchium Global Scholarship Consortium and Institutional Strategic Grants.

Data Availability: Analytical datasets and computational models are archived under open repositories under DOI: 10.5923/researchium.2024.jnsr.0301-data.

Conflict of Interest: The authors declare no competing financial or non-financial interests.

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