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Evidence for superconductivity above 260 K in lanthanum superhydride at megabar pressures

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Recent predictions and experimental observations of high $T_c$ superconductivity in hydrogen-rich materials at very high pressures are driving the search for superconductivity in the vicinity of room temperature. We have developed a novel preparation technique that is optimally suited for megabar pressure syntheses of superhydrides using modulated laser heating while maintaining the integrity of sample-probe contacts for electrical transport measurements to 200 GPa. We detail the synthesis and characterization of lanthanum superhydride samples, including four-probe electrical transport measurements that display significant drops in resistivity on cooling up to 260 K and 180-200 GPa, and resistivity transitions at both lower and higher temperatures in other experiments. Additional current-voltage measurements, critical current estimates, and low-temperature x-ray diffraction are also obtained. We suggest that the transitions represent signatures of superconductivity to near room temperature in phases of lanthanum superhydride, in good agreement with density functional structure search and BCS theory calculations.

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The search for superconducting metallic hydrogen at very high pressures has long been viewed as a key problem in physics [1,2]. The prediction of very high (e.g., room temperature) $T_c$ superconductivity in hydrogen-rich materials [3] has opened new possibilities for realizing high critical temperatures but at experimentally accessible pressures (i.e., below 300 GPa) where samples can be characterized with currently available tools. Following the discovery of novel compound formation in the S-H system at modest pressures [4], theoretical calculations predicted that hydrogen sulfide would transform on further compression to a superconductor with a $T_c$ up to the 200 K range [5,6]. The high $T_c$ of 203 K at 150 GPa in samples formed by compression of H$_2$S was subsequently confirmed [7,8], with x-ray measurements consistent with cubic H$_3$S as the superconducting phase [9].

Given that the higher hydrogen content in many simple hydride materials is predicted to give still higher $T_c$ values [3], we have extended our studies to higher hydrides, the so-called superhydrides, XH$_n$ with $n > 6$. Systematic theoretical structure searching in the La-H and Y-H systems reveals numerous hydrogen-rich compounds with strong electron-phonon coupling and $T_c$ in the neighborhood of room temperature (above 270 K) [10] (see also Ref. [11]). Of these superhydrides, LaH$_{10}$ and YH$_{10}$ have a novel clathrate-type structure with 32 hydrogen atoms surrounding each La or Y atom, and $T_c$ near 270 K at 210 GPa for LaH$_{10}$ and 300 K at 250 GPa in YH$_{10}$; see Ref. [12] for a recent review. Notably, in these phases the H-H distances are $\sim$1.1 Å, which are close to those predicted for solid atomic metallic hydrogen at these pressures [13].

Recently, our group successfully synthesized a series of superhydrides in the La-H system up to 200 GPa pressures. Specifically, we reported x-ray diffraction and optical studies demonstrating that the lanthanum superhydrides can be synthesized. The diffraction reveals that La atoms have a face centered cubic (fcc) lattice at 170 GPa upon heating to $\sim$1000 K [14], and a structure with compressibility close that for predicted cubic metallic phase of LaH$_{10}$. Experimental and theoretical constraints on the hydrogen content give a stoichiometry of LaH$_{10\pm x}$, where $x$ is between +2 and -1 [14]. On decompression, the fcc-based structure undergoes a rhombohedral distortion of the La sublattice to form a structure that has subsequently been predicted to also have a high $T_c$ [13]. Here we report the use of a novel synthesis route for megabar pressure syntheses of such superhydrides using pulsed laser heating and ammonia borane (NH$_3$BH$_3$, AB) as the hydrogen source. We detail the synthesis and characterization of several samples of the material using x-ray diffraction and electrical
resistance measurements at 180-200 GPa. The transport measurements reveal a clear resistance drops at 260 K using four-probe measurements and in other experiments. Consistent with low-temperature x-ray diffraction, we infer that the transition is a signature of near room-temperature superconductivity in LaH$_{10}$ and not related to any structural transition.

Samples were prepared using a variety of diamond-anvil cells (DACs), depending on the measurement [16]. For the electrical conductivity measurements, we used a multi-step process with piston-cylinder DACs [15] (Fig. 1). Composite gaskets consisting of a tungsten outer annulus and a cubic boron nitride epoxy mixture (cBN) insert were employed to contain the sample at megabar pressures while isolating the platinum electrical leads [16]. We synthesized the superhydride from a mixture of La and H$_2$ loaded in the gasket assembly as in our previous work [14], and found that maintaining good contact between the synthesized material and electrodes is not guaranteed. To overcome this problem, we used AB as the hydrogen source [17-19]. When completely dehydrogenated, one mole of AB yields three moles of H$_2$ plus insulating cBN, the latter serving both as a solid pressure medium and support holding the hydride sample firmly against the electrical contacts (Fig. 1).

The synthesis and structural characterization of samples were carried out \textit{in situ} at high pressure using a versatile diode-pumped ytterbium fiber laser heating system [20] that was adapted for our experiments. In all runs, we observed very good coupling of the laser with the sample and subsequent sharpening of La diffraction peaks; above 175 GPa and temperatures of 1000-1500 K, this resulted in the formation cubic LaH$_{10\pm x}$ [14]. To complete the transformation but avoid the formation of additional phases [16], we needed to maintain sample temperatures below 1800 K. This was achieved by varying the combination of laser power and pulse width of the heating laser. Typically, we found that a 300-ms pulse and a 30-μm laser spot size resulted in optimal transformation and sample coverage, and minimal anvil or electrode damage. Repeated checks on the two-probe resistance between the four electrodes between heating cycles were performed to ensure that electrodes in a given run were not damaged.

Representative diffraction patterns of three samples are shown in Fig. 2, including a LaH$_{10\pm x}$ sample that was verified to have the four-probe geometry intact at 188 GPa (Fig. 1 and Fig. 2c). The characteristic diffraction peaks of LaH$_{10\pm x}$ identified previously in synthesis from La and H$_2$ are observed, demonstrating that the superhydride phase can indeed be synthesized
from La and AB using the techniques described above. Further, the results show that synthesis can be carried out while maintaining the electrical leads intact. All the samples for which conductivity data are presented were also characterized by x-ray diffraction.

Figure 3 shows electrical resistance measurements as a function of temperature for the sample presented in Fig. 1 at an initial 300 K pressure of 188 GPa. On cooling, the resistance was observed to decrease around 275 K and to drop appreciably at 260 K, as first reported in Ref. [21]. The resistance abruptly dropped by $>10^3$ and remained constant from 253 K to 150 K. Upon warming, the resistance increased steeply at 245 K, indicating the change was reversible but shifted to lower temperature. Upon subsequent warming to 300 K, the pressure was measured to be 196 GPa. Since the pressure was not measured as a function of temperature during thermal cycling, the pressure at which the resistance change occurred was not directly determined. No dependence of the applied current on the transition temperature could be detected in this run, but the sample appeared to degrade with repeated thermal cycling at higher current levels (i.e., 10 mA) [16].

In a second four-probe conductivity measurement, we performed the laser heating synthesis, x-ray diffraction, and electrical conductivity in-situ on samples in the cryostat on the x-ray beamline. Although these samples were considerably smaller and mixed phase [16], we also more carefully monitored possible changes in transition temperature with applied current and measured the I-V characteristics of the material. We observed a 10 K decrease in transition temperature upon increasing the current from 0.1 mA to 1 mA and appeared to decrease further at higher currents (Fig. 4). Distinct I-V characteristics were observed above and below the transition temperature. The measurements suggest that well into the superconducting phase, LaH$_{10-\delta}$ could exhibit very large critical current densities [16,22]. In this experiment, part of the sample had spread outside the central culet, giving rise to the formation of lower pressure phases with their own transition temperatures. The high transition temperature observed here in several thermal cycles was also obtained from measurements on other samples using pseudo-four probe geometries (Fig. S4). Further discussion of these experiments is provided in Ref. [16].

An alternative explanation for the resistivity change is a temperature-induced, isostructural electronic transition with a dramatic increase in conductivity in the low-temperature phase. Consistent with our previous optical observations [14], the resistance measurements
indicate that the high-temperature phase (i.e., normal state) is a metal, so this would be a metal-metal transition. However, calculations reported to date do not predict such a transition within metallic LaH$_{10}$ [10,13]. Nevertheless, low-temperature x-ray diffraction was measured to determine whether the resistance change is due to a temperature-induced structural transition. The x-ray diffraction patterns reveal no change in volume and the available data show no indication of structural changes over the range of temperatures explored [16].

The complexity of the experiments prevents us from accurately determining the pressure dependence of the possible superconducting $T_c$. We did not attempt to determine the intrinsic resistivity of the superhydride samples because of their complex geometries, and some of the samples are clearly mixed phase, possibly with varying hydrogen stoichiometry [10,16]. In the three-point (pseudo-four probe) geometry, the contact resistance plays a major role and such low resistance values cannot be measured [23,24]. The samples could consist of layers of LaH$_x$ starting from $x = 10$ on the laser heated side and $x < 10$ toward the electrodes. The samples could also have a complicated toroidal geometry consisting of a central region of LaH$_{10-x}$ with a peripheral ring of untransformed La arising from the need to preserve the electrodes during laser heating and differential thermal heat transport for the sample in contact with the diamond versus the electrodes. Thus, variability in the observed transition temperatures may also arise from a changing network of the superconducting component on thermal cycling as well as the presence of multiple phases produced within a sample as a result of $P$-$T$ gradients during synthesis [16]. Indeed, after our work was completed, a report appeared of possible $T_c$ in lanthanum hydride at 215 K at slightly lower pressure [25], which we suggest may be a signature of another phase [13,16].

In summary, we report four-probe, ac resistance measurements on LaH$_{10-x}$ synthesized at pressures of 180-200 GPa by a modulated, pulsed laser heating technique that preserves the integrity of multi-probe electrical contacts on the sample after synthesis. Our multiple measurements reveal the signature of superconductivity at temperatures above 260 K at pressures of 180-200 GPa. The transition temperature is close to that predicted for the superconducting $T_c$ based on BCS calculations for LaH$_{10}$ at comparable pressures. Whereas diamagnetic measurements are needed to confirm the present results, we note the magnitude of the resistance drops repeatedly observed in our experiments is comparable to that observed in previous high-pressure studies where the transition was subsequently found to be correctly identified as
signatures of superconductivity by magnetic susceptibility (e.g., Refs. [23,26]). Extending and applying the latter technique for the smaller and more complex superhydride samples is progress. Infrared, optical, and x-ray spectroscopy [27,28] would also provide useful characterization, including identifying different superconducting phases and establishing the pressure dependence of $T_c$ for each component. A recent report of similar measurements of high $T_c$ in lanthanum superhydride samples, including the observation of a decrease in transition temperature in an external magnetic field [29], further validates this discovery of very high $T_c$ superconductivity in this system. The results reported here thus provide the first experimental evidence of near room temperature, as exhibited in this new class of hydride materials.

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Figure 1. (a) Schematic of the assembly used for synthesis and subsequent conductivity measurements. The sample chamber consisted of a tungsten outer gasket (W) with an insulating cBN insert (B). The piston diamond (P) was coated with four 1-μm thick Pt electrodes which were pressure-bonded to 25-μm thick Pt electrodes (yellow). The 5-μm thick La sample (red) was placed on the Pt electrodes and packed in with ammonia borane (AB, green). Once the synthesis pressure was reached, single-sided laser heating (L) was used to initiate the dissociation of AB and synthesis of the superhydride. To achieve optimal packing of AB in the gasket hole, we loaded AB with the gasket fixed on the cylinder diamond (C). (b) Optical micrograph of a sample at 178 GPa after laser heating using the above procedure (sample A).
**Figure 2.** Synchrotron x-ray diffraction patterns obtained from three samples in which LaH$_{10\pm x}$ was synthesized by laser heating at pressures above 175 GPa. (a) X-ray diffraction following laser heating of a mixture of La and H$_2$ (Au pressure marker); (b) and (c) results obtained following laser heating of La and NH$_3$BH$_3$ (samples A and C; Pt pressure marker). The data were obtained from three separate runs with an x-ray spot size of 5 x 5 μm. In all three panels, the pattern in black is from unreacted La, and that in red is fcc-based LaH$_{10\pm x}$ obtained after laser heating. The sample corresponding to (c) was nominally 5-μm thick and exhibited a high degree of anisotropic stress as indicated by the relative intensities, broadening, and shift of the La peaks prior to the reaction. The diffraction peak marked by the green symbol in (a) is due to residual WH$_x$ which forms when H$_2$ is present. This hydride also gives rise to the tail above 10.5 degrees. WH$_x$ features are absent in the other two patterns because insulating cBN gaskets were used. Further details are provided in Ref. [16].
Figure 3. Normalized electrical resistance of the LaH$_{10\pm\epsilon}$ sample characterized by x-ray diffraction and radiography (Figs. 1 and 2) and measured with the four-probe technique (sample A). The initial pressure determined from Raman measurements of the diamond anvil edge was 188 GPa. The lowest resistance we could record was 20 $\mu$Ω whereas the 300 K value was 50 mΩ. The measurements were performed at 10 mA and 10 kHz; for further details see Ref. [16].
Figure 4: Electrical resistance measurements using the four-probe technique (sample F). At the lowest current of 0.1 mA, the sample resistance displays an abrupt drop (grey curve, first heating cycle), and the transition temperature decreases with increasing current, as shown for 1 mA (blue curve, third heating cycle) and 10 mA (red curve, fifth heating cycle). The inset shows I-V curves obtained from this experiment, and compares data measured at 300 K (mV scale, left axis) and at 180 K (μV scale, right axis), well below the transition temperature. These data were obtained in-situ on the synchrotron beamline with different instrumentation and data collection protocols compared to the data presented in Fig. 3 (sample A). The flat response for temperatures above the transition reflects the dynamic range setting of the instrument needed for these smaller samples, and there is a lower density of points on the R-T curves; see Ref. [16] for further details.