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Dmitrii V. Semenok (dmitrii.semenok@skoltech.ru)
Skolkovo Institute of Science and Technology

Ivan A. Troyan
Shubnikov Institute of Crystallography, Federal Scientific Research Center Crystallography and Photonics, Russian Academy of Sciences

Andrey V. Sadakov
P. N. Lebedev Physical Institute, Russian Academy of Sciences

Di Zhou (D.Zhou@skoltech.ru)
Skolkovo Institute of Science and Technology

Michele Galasso
Skolkovo Institute of Science and Technology

Alexander G. Kvashnin
Skolkovo Institute of Science and Technology

Ivan A. Kruglov
Dukhov Research Institute of Automation (VNIIA), Institute of Physics and Technology

Alexey A. Bykov
NRC “Kurchatov Institute” PNPI

Konstantin Y. Terent’ev
Kirensky Institute of Physics

Alexander V. Cherepahin
Kirensky Institute of Physics

Oleg A. Sobolevskiy
P. N. Lebedev Physical Institute

Kirill S. Pervakov
P. N. Lebedev Physical Institute

Alexey Yu. Seregin
Shubnikov Institute of Crystallography, Federal Scientific Research Center Crystallography and Photonics, Russian Academy of Sciences, National Research Center “Kurchatov Institute”

Toni Helm
Hochfeld-Magnetlabor Dresden (HLD-EMFL), Helmholtz-Zentrum Dresden-Rossendorf (HZDR)

Tobias Förster
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Effect of paramagnetic impurities on superconductivity in polyhydrides:

*s*-wave order parameter in Nd-doped LaH$_{10}$

Dmitrii V. Semenok,¹,7 Ivan A. Troyan,² Andrey V. Sadakov,³ Di Zhou,¹,7 Michele Galasso,¹ Alexander G. Kvashnin,¹ Ivan A. Kruglov,⁸,⁹ Alexey A. Bykov,³ Konstantin Y. Terent’ev,⁴ Alexander V. Cherepahin,⁴ Oleg A. Sobolevskiy,⁵ Kirill S. Pervakov,⁵ Alexey Yu. Seregin,²,¹³ Toni Helm,¹² Tobias Förster,¹² Audrey D. Grockowiak,¹⁰,¹¹ Stanley W. Tozer,¹¹ Yuki Nakamoto,⁷ Katsuya Shimizu,⁷ Vladimir M. Pudalov,⁵,⁶ Igor S. Lyubutin,² and Artem R. Oganov¹,7

¹ Skolkovo Institute of Science and Technology, 121205, Bolshoy Boulevard 30, bld. 1, Moscow, Russia
² Shubnikov Institute of Crystallography, Federal Scientific Research Center Crystallography and Photonics, Russian Academy of Sciences, 59 Leninovsky Prospekt, Moscow 119333, Russia
³ NRC “Kurchatov Institute” PNPI, Gatchina, Russia
⁴ Kirensky Institute of Physics, Krasnoyarsk, Russia
⁵ P. N. Lebedev Physical Institute, Russian Academy of Sciences, Moscow 119991, Russia
⁶ National Research University Higher School of Economics, Moscow 101000, Russia
⁷ KYOKUGEN, Graduate School of Engineering Science, Osaka University, Machikaneyamacho 1-3, Toyonaka, Osaka, 560-8531, Japan
⁸ Dukhov Research Institute of Automatics (VNIIA), Moscow 127055, Russia
⁹ Moscow Institute of Physics and Technology, 9 Institutsky Lane, Dolgoprudny 141700, Russia
¹⁰ Brazilian Synchrotron Light Laboratory (LNLS/Sirius), Brazilian Center for Research in Energy and Materials (CNPEM), Campinas, Brazil
¹¹ National High Magnetic Field Laboratory, Florida State University, Tallahassee, Florida, 32310, USA
¹² Hochfeld-Magnetlabor Dresden (HLD-EMFL), Helmholtz-Zentrum Dresden-Rossendorf (HZDR), 01328 Dresden, Germany
¹³ National Research Center “Kurchatov Institute”, Moscow 123182, Russia

*Corresponding authors: Artem R. Oganov (a.oganov@skoltech.ru), Di Zhou (D.Zhou@skoltech.ru), Dmitrii Semenok (dmitrii.semenok@skoltech.ru)

Abstract

Polyhydrides are a novel class of superconducting materials with extremely high critical parameters, which is very promising for applications. On the other hand, complete experimental study of the magnetic phase diagram for the best so far known superconductor, lanthanum decahydride LaH$_{10}$, encounters a serious complication because of the large upper critical magnetic field $H_{C2}(0)$, exceeding 120–160 T. Partial replacement of La atoms by magnetic Nd atoms results in a decrease of the upper critical field, which makes it attainable for existing pulse magnets. We found that addition of neodymium leads to significant suppression of superconductivity in LaH$_{10}$: each atomic % of Nd causes decrease in $T_C$ by 10–11 K. Using strong pulsed magnetic fields up to 68 T, we constructed the magnetic phase diagram of the ternary (La,Nd)H$_{10}$ superhydride, which appears to be surprisingly linear with $H_{C2} \propto |T – T_C|$. The pronounced suppression of superconductivity in LaH$_{10}$ by magnetic Nd atoms and the robustness of $T_C$ with respect to nonmagnetic impurities (e.g., Y, Al, C) under Anderson’s theorem indicate the isotropic ($s$-wave) character of conventional electron-phonon pairing in the synthesized superhydrides.

Keywords: hydrides, high pressure, superconductivity, Anderson’s theorem
Introduction

The search for high-temperature superconductivity is one of the most challenging tasks of condensed matter physics and materials science. An appealing idea dating back to the early 1960s was that metallic hydrogen should be a high-temperature superconductor with a critical temperature \( T_C \) of 300–400 K. However, high pressures above 3–4 Mbar are needed for the transition of hydrogen from the molecular insulating phase to the metallic state. For this reason, researchers have recently started to explore the possibility of inducing a superconducting state by combining other elements to hydrogen, which led to the formation of a novel class of chemical compounds — polyhydrides — with a significant reduction of the metallization pressure while maintaining high \( T_C \).

Several hydrogen-rich superconductors, such as H\(_3\)S (\( T_C \) reaches 191–204 K), YH\(_6\) (\( T_C = 224–227 \) K) and lanthanum superhydride LaH\(_{10}\) (\( T_C \) is about 250 K), have been successfully synthesized by several groups. Because of its extremely high critical temperature, the lanthanum–hydrogen system has become of particular importance and been intensively studied. Key to a better understanding of polyhydrides is an exploration of their magnetic phase diagrams, which show the boundary between the superconducting and normal state of polyhydrides in the “temperature–magnetic field” coordinates, and enable to verify the type of the superconducting state. However, such comprehensive experimental studies of LaH\(_{10}\) encounter a great technical challenge due to very high values of the upper critical magnetic field \( \mu_0 H_{c2}(0) \) exceeding 120–160 T, whereas modern pulsed magnets can only generate fields of up to 70–100 T in a volume of a high-pressure diamond anvil cell (DAC).

As we show in this work, \( \mu_0 H_{c2}(0) \) can be significantly reduced by the partial replacement of the La atoms without disrupting the cubic crystal structure of lanthanum superhydride LaH\(_{10}\). This enables investigations in existing pulsed magnets. Moreover, the study of the effect of nonmagnetic (e.g., Y) and magnetic (such as Nd) impurities or dopants on superconductors makes it possible to distinguish between conventional and unconventional mechanisms of superconductivity.

Anderson’s theorem states that nonmagnetic (scalar) impurities do not affect the isotropic singlet \( s \)-wave order parameter in the conventional Bardeen–Cooper–Schrieffer (BCS) theory, whereas scattering on paramagnetic centers is very efficient in destroying \( s \)-wave electron–electron pairing. Nonmagnetic and magnetic impurities are equally detrimental to the critical temperature \( T_C \) of unconventional superconducting states. The introduction of such impurities can provide important information on the structure of the magnetic phase diagram and the mechanism of pairing in polyhydrides under pressure.

In this work, we successfully synthesized a series of ternary polyhydrides (La,Nd)H\(_{10}\) containing 8–20 at% of Nd at a pressure of 170–180 GPa. Using pulsed magnetic fields up to 68 T, we constructed the magnetic phase diagram of (La,Nd)H\(_{10}\), which showed a surprisingly linear \( \mu_0 H_{c2}(T) \propto (T_C - T) \). The results of the transport measurements indicate that the lanthanum superhydride follows the conventional Bardeen–Cooper–Schrieffer (BCS) model of superconductivity with the \( s \)-wave pairing.

Results

High-pressure synthesis and stability of La–Nd hydrides

Several La–Nd alloys with approximately 8, 9, 20, 25, and 50 at% of Nd were prepared using melting of La and Nd in an inert atmosphere. The samples were carefully characterized by the scanning electron microscopy and X-ray powder diffraction (Supporting Information Figures S1–S4). The La–Nd samples (pre-compressed 1-\( \mu \)m thick pieces) mixed with ammonia borane NH\(_3\)BH\(_3\) were heated by a pulsed laser light for
several hundred microseconds to 1500–2000 K at pressures of 170–180 GPa (determined via the Raman signal of diamond) in DACs M1–E3 (Supporting Information Table S1). We most fully investigated the properties of lanthanum-neodymium superhydrides prepared from the La–Nd (9 ± 0.5 %) alloy by powder X-ray diffraction (XRD) of synchrotron radiation with a wavelength of 0.413 Å (Figure 1).

We found that a series of the strongest reflections in the XRD pattern (Figure 1a) corresponds to a cubic crystal structure, which is typical for decahydrides such as LaH\textsubscript{10} at pressures above 150 GPa. The experimental equation of state \( V(P) \) of the main phase (Figure 1b) allowed us to determine the metal-to-hydrogen ratio in the obtained compound to be 1:10. Considering good agreement with theoretical calculations and the fact that the initial composition of the La–Nd alloy is close to LaNd, we further assign to this phase the chemical formula \( Fm\overline{3}m-(\text{La}_{0.91}\text{Nd}_{0.09})\text{H}_{10} \) with a unit cell volume of \( \sim30.3 \text{ Å}^3 \) per metal atom at 200 GPa (Supporting Information Table S2), slightly lower than the cell volume of \( Fm\overline{3}m-\text{LaH}_{10} \) (Figure 1b).\textsuperscript{6,16} It is worth pointing out that cubic NdH\textsubscript{10}, which was not obtained during the direct synthesis below 130 GPa,\textsuperscript{17} can be stabilized in the LaH\textsubscript{10} sublattice at higher pressure. We have previously observed the same behavior for YH\textsubscript{10}, stabilized in the lattice of ternary polyhydrides (La,Y)H\textsubscript{10}.\textsuperscript{18} The impurity phase in the sample (asterisks in the Figure 1a) has a low hydrogen content and can be explained by the composition \( \text{Immm-}(\text{La,Nd})_{3}\text{H}_8 \), which is \( \sim4.4 \text{ meV/atom} \) above the convex hull of the La-Nd–H system at 200 GPa.

Figure 1. (a) Experimental XRD pattern and the Le Bail refinement of the (La,Nd)H\textsubscript{10}, obtained from La\textsubscript{0.91}Nd\textsubscript{0.09} alloy, and \( \text{Immm-} \text{La}_3\text{H}_8 \) unit cell parameters at 159 GPa. (b) Experimental and theoretical equations of state of LaH\textsubscript{10} \textsuperscript{6,16,19,20} and newly synthesized (La,Nd)H\textsubscript{10}. (c) XRD patterns obtained during decompression of DAC M1 from 202 to 143 GPa. (d) Calculated convex hull of the ternary La–Nd–H system at 200 GPa considering the spin–orbit coupling (SOC) and zero-point energy (ZPE) at 0 K. Stable and metastable phases are shown in blue and red, respectively. (e, f) Crystal structures of the model compounds — ternary La–Nd hydrides with pseudocubic (fcc) metal sublattices \( I4/m-\text{La}_4\text{NdH}_{50} \) and \( C2/m-\text{La}_4\text{NdH}_{50} \).

In the context of the joint experimental and theoretical approach, we analyzed the stability of various
La–Nd–H phases using the USPEX code. The most thermodynamically stable ternary polyhydride is $C2/m$-La$_4$NdH$_{50}$, which has a high-symmetry modification $I4/m$-La$_4$NdH$_{50}$ lying 5 meV above the convex hull. Comparing the stability of different (La,Nd)H$_{10}$ phases toward their decomposition to LaH$_{10} +$ NdH$_{10}$, we see that pseudocubic $P1$-La$_9$NdH$_{100}$ and $C2/m$-La$_4$NdH$_{50}$ are stable at 200 GPa (Supporting Information Figure S8a-c). The situation does not change when the zero-point energy (ZPE) is considered (Supporting Information Figures S8d-f), but the results essentially depend on the spin–orbit coupling (SOC) and the type of La pseudopotential used. The calculations that include SOC give the Debye temperature $\Delta T_C \approx 30$ K, with the onset superconducting critical temperature $T_C = 148$ K (Figure 2a), about 100 K lower than in pure parent LaH$_{10}$ at the same pressure. The residual resistance at 100 K does not exceed 0.5 m$\Omega$ as compared to 0.8 $\Omega$ in the normal state at 160 K. In this case, we can consider the resistance $R(T)$ above $T_C$ also comes from the synthesized hydride and use the Bloch–Gruneisen formula which gives the Debye temperature $\theta_D \approx 750$ K and the corresponding electron–phonon coupling strength $\lambda_{BG} \approx 2.87$ ($\mu^* = 0.1$).

The superconducting transition was initially studied in steady magnetic fields up to 16 T (Figure 2b). In this range of fields the upper critical magnetic field of (La$_{0.91}$Nd$_{0.09}$)H$_{10}$ shows linear dependence $\mu_0H_{C2} \sim |T - T_C|$ with the slope $d\mu_0H_{C2}(T_C)/dT = -0.71$ T/K. The superconducting transitions reveal no significant broadening in the range of fields (0–16 T): $(T_C^{99\%} - T_C^{10\%}) \sim 23$–25 K, $\Delta T_C/T_C \sim 0.2$. Different models were used to extrapolate the upper critical magnetic field to 0 K. The Werthamer–Helfand–Hohenberg (WHH) model yields $\mu_0H_{C2}(0) = 73$ T (Figure 2d). A (1-t$^2$)-model, also called the Ginzburg-Landau (GL) model, gives much lower $\mu_0H_{C2}(0) \sim 54$ T. As we show below, a linear extrapolation yields a result that is much closer to the experiment.

To extend the magnetic phase diagram of (La$_{0.91}$Nd$_{0.09}$)H$_{10}$ and verify its extrapolated value $\mu_0H_{C2}(0)$, we used pulsed fields up to 68 T (Figure 2c,e) and a special DAC E1 made of Ni–Cr–Al alloy. The dependence of the upper critical magnetic field on temperature is linear up to fields of about 68 T, which is 64–70 % of...
the extrapolated value $\mu_0 H_{C2}(0) = 100–110$ T (Figure 2d,e). It has recently been shown that some other hydrides (e.g., low-pressure modification of LaH_{10}, SnH_{4}, YH_{4}) exhibit the linear dependence $\mu_0 H_{C2}(T) = a \times (T_C - T)$ in the overall temperature range (see also Discussion). This contradicts the conventional WHH model widely used for hydrides, which predicts saturation of $\mu_0 H_{C2}(T)$ in such strong magnetic fields and, hence, underestimates $\mu_0 H_{C2}(0)$.

Another important result is the observation of the broadening of superconducting transitions in the first series of experiments with pulsed magnetic fields. The broadening of resistive transitions in magnetic fields is a distinctive feature of most superconductors, but it is not always observed in compressed polyhydrides. The absence of broadening of resistive transitions is also seen in our (La,Nd)H_{10} sample in weak magnetic fields up to 16 T (<16% of $\mu_0 H_{C2}(0)$, Figure 2b). However, as seen in Figure 2c, such broadening appears in much stronger magnetic fields $H > 0.5 H_{C2}(0)$, which are available only with pulse magnets. More specifically, in the range from 7 to 53 T, the superconducting transitions of (La_{0.91}Nd_{0.09})H_{10} demonstrate a notable broadening expected in conventional superconductors, with $T_{C0.25} - T_{C0.025}$ changing from 14 to 28 K and $\Delta T_C/T_C$ increasing from 12% to 41% (Figure 2c). In the second series (run 2) of high-field measurements, it was not possible to observe the broadening of the superconducting transitions because of their large width for this sample even in the absence of a magnetic field (Supporting Information Figure S19b).

Figure 2. High-pressure electrical measurements in La–Nd hydrides in DACs E0 and E1 (180 and 170 GPa). (a) Temperature dependence of the electrical resistance of the synthesized sample at 180 GPa (DAC E0, the current frequency ($f$) is 4 Hz). Inset: residual resistance in the superconducting state. (b) Superconducting transitions in magnetic fields of 0–16 T. (c) Electrical resistance of the sample in strong pulsed magnetic fields ($f = 3.33$ kHz) at different temperatures (run 1). (d) Extrapolation of the upper critical magnetic field $H_{C2}(0)$ using the Werthamer–Helfand–Hohenberg model (WHH), linear fit, and $(1 - t^2)$ Ginzburg-Landau model. (e) Magnetic phase diagram of (La_{0.91}Nd_{0.09})H_{10} synthesized in DAC E1 (170 GPa). Inset: photo of the diamond anvil culet with the sample and electrodes. (f) Dependence of the critical current $I_C(T, B)$ on the applied magnetic field and temperature (DAC E1).

The linear dependence of the upper critical magnetic field on the temperature requires a brief discussion. For BCS superconductors, the generally accepted model of the $\mu_0 H_{C2}(T)$ dependence is WHH, which predicts saturation (flattening) of $\mu_0 H_{C2}(T)$ at low temperatures.$^{33}$ However, for many compressed polyhydrides (YH_{4},...
LaH\textsubscript{10} at low pressures\textsuperscript{39}, completely linear $\mu_0H_{C2}(T)$ dependence was observed down to $\sim$1–2 K. This behavior may indicate the presence of two superconducting gaps,\textsuperscript{40-44} which is confirmed for a number of polyhydrides ($Fm\overline{3}m$-LaH\textsubscript{10}, $Fm\overline{3}m$-YH\textsubscript{10}, $P6_3/mmc$-YH\textsubscript{6}, etc.) by solving the anisotropic Migdal–Eliashberg equations.\textsuperscript{45,46} However, this explanation seems unsatisfactory, since the two-gap superconductivity is not universal in hydrides (e.g., $Im\overline{3}m$-CaH\textsubscript{6}\textsuperscript{47}), nevertheless, ascending segments of the $\mu_0H_{C2}(T)$ dependence have never been observed, and polyhydrides following the WHH model are unknown at the moment. Almost all superconducting polyhydrides known today exhibit a close to linear $\mu_0H_{C2}(T)$ dependence (Supporting Information Figure S18).

We believe that the linear behavior of $\mu_0H_{C2}(T)$ is associated with the mesoscopic inhomogeneity of the sample, that is the presence of regions with different composition and hydrogen content and, consequently, different values of $T_C$ and $\mu_0H_{C2}$.\textsuperscript{48,49} Such inhomogeneous state may be considered as a granular superconductor. As long as “islands” of superconductivity are bound via the Josephson effect, we can detect superconductivity in the sample. The proposed explanation agrees with the current absence of observations of the Meissner effect in hydride superconductors, which is very difficult to detect, whereas the diamagnetic screening (at zero-field cooling) has been found.\textsuperscript{16,50}

### Critical current, impurity concentration and magnetoresistance of La–Nd hydrides

Superconductivity can be suppressed by reaching the critical temperature, the upper critical magnetic field or the critical current density. The critical currents and voltage–current ($U$–$I$) characteristics for the (La\textsubscript{0.91}Nd\textsubscript{0.09})H\textsubscript{10} sample were investigated in the range from $10^{-5}$ to $10^{-2}$ A in external magnetic fields at pressure of 170 GPa (DAC E1, Figure 2f). The critical current density was estimated from the size and thickness of the sample, which were 25–30 µm and $\sim$1 µm (the thickness of the loaded piece of the La–Nd alloy), respectively. It was established that the critical current density in (La,Nd)H\textsubscript{10} exceeds 400 A/mm\textsuperscript{2} at 105 K. The extrapolation to $T$=0 K using the conservative Ginzburg–Landau model\textsuperscript{34} $J_C = J_C(0) \times (1 - T/T_C)^{3/2}$ gives the critical current $I_C(0) = 0.22$ A and the critical current density $J_C(0)$ that may exceed 8.8 kA/mm\textsuperscript{2}. The single vortex model\textsuperscript{51} $J_C = J_C(0) \times (1 - T/T_C)^{5/2}(1 + T/T_C)^{-1/2}$ yields much higher values: the critical current $I_C$ may reach 3 A at 0 K and the critical current density $J_C(0)$ may exceed 120 kA/mm\textsuperscript{2}. However, the heterogeneous structure of the sample can significantly reduce the real values of the current density that can be achieved.

The critical current measurements can be used to estimate the superconducting gap (Figure 3d). Talantsev and Tallon proposed the following model for s-wave superconductors:\textsuperscript{52}

$$I_C(T)^{2/3} = I_C(0)^{2/3} \left(1 - 2 \frac{\pi\Delta(0)}{k_B T} \exp \left( - \frac{\Delta(0)}{k_B T} \right) \right),$$  \hspace{1cm} (1)

where $I_C(T)$ is the critical current at temperature $T$ in the absence of the magnetic field and $\Delta(0)$ is the superconducting gap at 0 K. Interpolation of the $I_C(T)$ data using both Ginzburg–Landau and Dew-Hughes models for $I_C(0)$ and Eq. (1) yields similar $\Delta(0) = 14.5$–15 meV, which, however, is lower than expected from $2\Delta(0)/k_BT_c = 3.52$.

According to the Gor’kov theory, the $T_C$-dependence for (La,Nd)H\textsubscript{10} on the Nd concentration ($x << 1$) can be expressed as a linear function:\textsuperscript{11,53}

$$k_B(T_C^{\text{max}} - T_C(x)) = \frac{\pi h}{4\tau} x,$$  \hspace{1cm} (2)

where $\tau$ is the collision time resulting from the impurity potential in the Born approximation. In our case,
\[ \tau = 5.4 \times 10^{-15} \text{ s}, \ x = 0.09 \] (atomic fraction of Nd), which corresponds to a “dirty” metal. Thus, each atomic percent of neodymium suppresses \( T_c(\text{LaH}_{10}) \) by about 10–11 K or, in relative values, \( \Delta T_c(1\% \text{ Nd})/T(\text{LaH}_{10}) = 0.044 \) (Figure 3e), which is much less than the degree of suppression of superconductivity in pure lanthanum,\(^{54} \) where \( \Delta T_c(1\% \text{ Nd})/T(\text{La}) = 0.144 \). This must be considered when synthesizing \( \text{LaH}_{10} \), where impurities of \( \sim 0.1\% \) of other lanthanides can negatively affect the attainable critical temperature.

According to theory\(^{11,53} \), the critical concentration \( x_{cr} \) of a magnetic impurity that completely suppresses superconductivity in \( \text{LaH}_{10} \) can be derived from the experiment:

\[ x_{cr} = \frac{\pi^2}{8 \gamma} \left( \frac{T_{c,\text{max}}}{T_c(x)} \right)^x, \]  

(3)

where \( \gamma \approx 1.78 \) is the Euler constant; then \( x_{cr} = 0.15 \). Indeed, \( \text{(La},\text{Nd})\text{H}_{10} \) synthesized from the \( \text{La}_{0.8}\text{Nd}_{0.2} \) alloy containing 20 at\% of neodymium does not demonstrate clear superconducting transitions (DAC E2, Supporting Information Figure S20).

Figure 3. Galvanomagnetic effects in \( \text{La}_{0.91}\text{Nd}_{0.09}\text{H}_{10} \) at \( \sim 170 \) GPa measured in steady and pulsed magnetic fields. (a) Dependence of the detected voltage on the applied magnetic field (from –16 to 16 T) at different temperatures. Between 90 and 150 K, the main contribution to the magnetoresistance (MR) is due to the superconducting transition. (b) Dependence of the sample resistance (arbitrary units, the curves are offset-shifted for clarity) on the applied magnetic field for \( T > T_c \). There is an obvious change in the sign of MR in 200-250 K region. (c) Detailed comparison of the dependence of the relative resistance on the magnetic field (\( \rho \sim H^2 \)) and its quadratic character at low fields. Dashed lines correspond to parabolic fit. (d) Dependence of the linear term in MR and the Hall coefficient divided by \( d(\text{RH}/d) \), where \( d \) is the poorly known thickness of the sample) at \( T > T_c \). (e) Experimental and calculated (using the local density approximation) dependence of \( T_c \) on the concentration of Nd in \( \text{(La},\text{Nd})\text{H}_{10} \) at 170–180 GPa. Inset: sample position in the DAC E1 for measurements of MR in pulsed magnetic fields. (f) Linear term in magnetoresistance in pulsed magnetic fields of 37–67 T at 105±5 K.

Low-field measurements in \( \text{(La},\text{Nd})\text{H}_{10} \) doped by 9 at\% of Nd (Figure 3a) indicate the presence of two terms in the field dependencies of the magnetoresistance (MR, Figure 3c): quadratic \( \rho = (\text{R-R}_0)/\text{R}_0 = \mu^2 H^2 \) (dominating in low fields), where \( \mu \) – is the electron mobility (Table 1), and linear \( \rho = aH \) (dominating in
fields above 7–9 T). In the experiment with a pulsed magnetic field, a trend of a linear resistance increase with the field is seen despite the noise (Figure 3f). The linear behavior of the magnetoresistance is typical for polycrystalline metals with an open Fermi surface (e.g., spherical with “necks”), for example, Li, Cu, Ag, Au, SrZnSb$_2$, MnBi$^5$, and others. This was explained by Lifshitz and Peschanskii in 1959 by considering the shape of the Fermi surface and the anisotropy of the MR. It leads to a linear dependence of the MR on the field when averaged over all directions ($\Delta \theta \sim H_0/H$, here $H_0$ - is the field for which the cyclotron time equals the electron mean free time) and open sections of the Fermi surface (where $\rho \sim H^2$) for polycrystals. For materials with a closed FS, linear MR is observed in disordered polycrystalline narrow-band semiconductors and semimetals. It is also intrinsic to topological insulators and semimetals with linear dispersing Dirac cones.

The configuration of the potential contacts didn't allow to fully separate the Hall and diagonal voltage drops, and, therefore, the measured voltage drop contained an admixture of the Hall voltage to the diagonal one. The Hall component was then extracted from the asymmetry of the quadratic dependence of the MR in low fields. We found an anomaly in the behavior of the sample (DAC E1) around 200-250 K. The Hall coefficient (Table 1) changes sign in this temperature interval (Figure 3b-d). At the same time, magnetoresistance becomes very small and even negative at 250 K (Figure 3b). Both anomalies are correlated and clearly indicate the band structure and, possibly, crystal structure changes in this temperature range.

The properties of the (La,Nd)H$_{10}$ sample in DAC E1 at low temperature are also unusual. Studying this sample, we found anomaly at 9 K (Supporting Information Figure S22). The electrical resistance begins to decrease sharply at 150 K and reaches a plateau at about 50 K, which corresponds to the superconducting state. Upon cooling below 9 K, the resistance begins to rise again, forming a low-temperature anomaly that does not depend on either the frequency or the excitation current, but can be suppressed by an external magnetic field of ~9 T. The most likely reason for this behavior is a change in the magnetic ordering in the Nd sublattice of (La,Nd)H$_{10}$. Namely, low-temperature jump in electrical resistance may be due to the antiferromagnetic ordering in (La,Nd)H$_{10}$ below the Néel temperature $T_N = 12$ K, which was predicted using the DFT calculations for simplified $P\bar{T}$-La$_9$NdH$^{100}$ model (see Supporting Information Figures S22-S24).

Table 1. Parameters of the normal state of the La–Nd polyhydride (9 at% of Nd) obtained in the study of magnetoresistance in steady and pulsed magnetic fields.

| Temperature, K | Electron mobility, $10^{-3}$ m$^2$/s×V | Electron relaxation time, 10$^{-14}$ s | $R_H/d$, 10$^{-6}$ m$^2$/C | Hall coefficient $R_H$ at $d = 1$ μm, 10$^{-12}$ m$^3$/C | Linear MR, (dp/dH)$_H$, 10$^{-5}$ T$^{-1}$ |
|---------------|----------------------------------------|-------------------------------------|-------------------------------|---------------------------------|----------------------------------|
| 105           | –                                      | –                                   | –                            | –                               | 200                              |
| 150           | –4.7                                   | –2.70                               | –1.79                        | –1.79                           | ~66.9                            |
| 175           | 1.9                                    | 1.07                                | 0.92                         | 0.92                            | 3.10                             |
| 200           | 3.5                                    | 1.99                                | 7.27                         | 0.73                            | 54.4                             |
| 210           | 0.55                                   | 0.32                                | –6.27                        | –6.25                           | ~0.63                            |
| 230           | 0.8                                    | 0.45                                | –0.21                        | –0.21                           | ~0.79                            |
| 250           | 2.8                                    | 1.58                                | –1.01                        | –1.015                          | ~9.64                            |
| 270           | 2.6                                    | 1.46                                | –0.82                        | –0.815                          | 20.8                             |

* C is Coulomb

The mobility of electrons and their relaxation time (Table 1) in (La,Nd)H$_{10}$, calculated from the quadratic part of the $\rho(H) \propto \mu^2H^2$ dependence, correspond to the parameters of typical metals. Measurements in low magnetic fields allow to estimate the Hall coefficient $R_H$ at ~10$^{-12}$ m$^3$/C (sample thickness ~1–2 μm). Within an order of magnitude, this value of $R_H$ corresponds to those of ordinary metals, which suggests that the concentration of charge carriers in La–Nd hydrides is ~10$^{30}$ electron/m$^3$. 

8
**Discussion**

The main goal of studying ternary hydrides is to find a way to control the superconducting properties of known polyhydrides (such as H$_3$S, LaH$_{10}$, YH$_6$, YH$_9$, and CeH$_9$) by doping. Nowadays, main hopes for increasing $T_C$ are pinned on ternary and higher order hydrides. However, their studies require complex and accurate calculations, presumably for this reason, many discrepancies between theoretical and experimental results and overestimations of the superconducting properties have been published in the past few years.\(^{24,60,64-66}\)

Studies of the effect of nonmagnetic and magnetic impurities on superconductors enabled the distinction between isotropic and anisotropic superconductivity.\(^{11,67}\) Nonmagnetic (scalar) impurities do not affect the isotropic singlet $s$-wave order parameter (for example, small carbon impurities in lanthanum do not affect the superconducting transition in (La, C)H$_{10}$ that is observed at about 245 K\(^ {68}\)). The absence of the influence of C and Al impurities\(^ {68}\) also casts doubt on attempts to explain the supposedly huge increase in $T_C$ by doping in experiments with C-doped H$_3$S\(^ {69}\) and LaH$_{10}$ doped with Ga, B/N.\(^ {63,66,70}\) One might think that the point here is the formation of true ternary and quaternary polyhydrides. But a detailed theoretical analysis shows\(^ {66,71}\) that this can hardly lead to high-$T_C$ superconductivity in the framework of the BCS theory. Experimental results\(^ {63}\) also do not allow us to draw unambiguous conclusions. Moreover, as we have shown previously, no significant increase in $T_C$ is observed in the La–Y system, which can be considered as the absence of the influence of nonmagnetic yttrium impurities on the superconducting properties of LaH$_{10}$.\(^ {18}\)

This corresponds to so-called Anderson’s theorem,\(^ {12,72}\) which states that BCS superconductors are practically insensitive to small amount of nonmagnetic impurities. If the leading mechanism in compressed polyhydrides is the electron–phonon interaction, as most experimental and theoretical studies suggest,\(^ {73,74}\) then it is impossible to expect a significant effect of small additives, such as C or CH$_4$ in H$_3$S,\(^ {75,76}\) on superconductivity. The opposite situation would be a strong argument in favor of an unconventional pairing mechanism in these compounds. As this study shows, La–Nd hydrides exhibit the properties of typical metals, have a very low Hall coefficient, a rather high electron mobility, and a linear term in the magnetoresistance, indicating an open topology of the Fermi surface, which is confirmed by the DFT calculations.\(^ {64,77}\) As we have shown, the observed superconducting and normal state properties are explained within conventional concepts, leaving little chances for unconventional superconductivity in polyhydrides.

In sharp contrast, magnetic scattering is very efficient in destroying $s$-wave superconductivity, as shown in this study. The main observation of this work is that Nd effectively suppresses superconductivity in LaH$_{10}$, whereas its $Fm\bar{3}m$ crystal structure remains almost unchanged because of the great similarity of the physical and chemical properties of La and Nd atoms. With available pulsed magnetic fields, we suppressed superconductivity in (La, Nd)H$_{10}$ much effectively on the $h = B/B_{C2}(0)$ scale than it is currently possible for pure LaH$_{10}$. Given such a strong suppression of superconductivity in (La, Nd)H$_{10}$, where the critical concentration of Nd is ~15 at%, the absence of the superconducting properties in NdH$_9$,\(^ {17}\) PrH$_9$,\(^ {78}\) and in EuH$_9$\(^ {79}\) becomes not surprising anymore. It is hard to believe that high-$T_C$ superconductivity above 200 K can be realized in binary polyhydrides of most other lanthanides (Tm, Yb, Lu), as was recently predicted.\(^ {80}\)

**Conclusions**

Superhydrides are a new class of hydrogen-rich materials whose research is a novel direction in materials science. What is the mobility of hydrogen in hydrides at high pressures and temperatures? Are there a photovoltaic effect and photoconductivity? How do photons affect the superconducting transition in hydrides?
What are the Hall coefficient and the magnetoresistance? Are they subjected to the topological Lifshitz transition? All these questions have yet to be answered.

In this research, we made a step forward in this direction and successfully synthesized novel ternary polyhydrides \((\text{La,Nd})\text{H}_{10}\) containing 8–20 at% of Nd atoms, randomly distributed in \(\text{LaH}_{10}\)-like metal sublattice. The electric transport measurements demonstrated that the addition of magnetic impurities (Nd) leads to a significant suppression of superconductivity in \(\text{LaH}_{10}\): each atomic % of Nd causes decrease in \(T_C\) by 10–11 K. Superconductivity disappears at a critical concentration of Nd of about 15–20 at%.

Using strong pulsed magnetic fields of up to 68 T, we constructed the magnetic phase diagram of synthesized \((\text{La,Nd})\text{H}_{10}\), studied the magnetoresistance and the Hall effect. Surprisingly, the dependence of the upper critical magnetic field on temperature is found to be completely linear, possibly due to the mesoscopic inhomogeneity of the hydride sample. The extrapolated upper critical magnetic field \(\mu_0H_C^2(0)\) for \(\text{La}_{0.91}\text{Nd}_{0.09}\text{H}_{10}\) is 105–110 T at 170 GPa. The current-voltage measurements showed that the critical current density \(J_C(0)\) in \((\text{La,Nd})\text{H}_{10}\) may exceed 8.8 kA/mm².

In this study, we showed that \(\text{La–Nd}\) hydrides exhibit the properties of normal metals: they have a rather small Hall coefficient \((R_H \sim 10^{-12} \text{m}^3/\text{C})\), high concentration of charge carriers of \(\sim 10^{30} \text{electron/m}^3\), and a linear part of the magnetoresistance, indicating an open topology of the Fermi surface and the presence of Dirac cones in the vicinity of the Fermi energy. The validity of Anderson’s theorem for the studied hydrides and the typical-metal galvanomagnetic properties provide strong evidence for the conventional electron-phonon mechanism of superconductivity in hydrogen-rich materials under pressure.

**Author contributions**

D.V.S., I.A.T., and A.V.S. contributed equally to this work. I.A.T, D.V.S., D.Z., K.S., Y.N., A.Yu.S., T.H., T.F., A.D.G., and S.W.T. performed the experiments. I.A.K. performed the T-USPEX and anharmonic phonon density of states calculations. A.A.B., K.Y.T., A.V.C., and K.S.P. prepared the \(\text{La–Nd}\) alloys. M.G. wrote the Python scripts for accelerated USPEX data processing and automatic interpretation of diffraction patterns, and performed the calculations of the magnetic properties. A.G.K. and D.Z. prepared the theoretical analysis and calculated the equation of states, electron and phonon band structures, and superconducting properties. D.V.S., D.Z., and A.R.O. analyzed and interpreted the experimental results and wrote the manuscript. I.A.T., A.V.S., and O.A.S. made the electric transport measurements in low magnetic fields. T.H., T.F., A.D.G., and S.W.T. carried out the measurements in pulsed magnetic fields at HZDR HLD. Y.N. and K.S. performed the X-ray diffraction studies at SPring-8 synchrotron source. I.S.L., A.R.O., and V.M.P. directed the research, analyzed the results and edited the manuscript. All the authors provided critical feedback and helped shape the research.

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Data availability

The raw and processed data required to reproduce these findings are available to download from GitHub: https://github.com/mark6871/SPring-8-February-2021-, and as Supporting Information for the manuscript.

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