Effect of impurities on superconductivity in LaH_{10}

Near-room-temperature superconductors are certainly among the most attention-grabbing compounds in materials science. Theoretically, compressed hydrogen should be the best high-temperature superconductor, but it is hard to squeeze it above 450 GPa to turn hydrogen into a metal [1]. Thus instead, scientists are exploring compounds that contain additional elements, besides lots of hydrogen. In that way, some critical temperature (T_C) is sacrificed to reduce the pressures needed to stabilize the superconducting material to 100–200 GPa and into the realm of what is technologically possible. At this moment, lanthanum superhydride LaH_{10} with a well-reproducible critical temperature (T_C) of 250 K is the best-known superconductor [2]. This is very impressive, but to attain even higher critical temperatures, we first had to understand how superconductivity in this material works.

There are multiple microscopic mechanisms of superconductivity. The one best understood is called conventional phonon-mediated superconductivity [3]. The well-established theory of conventional superconductivity can be applied to improve the properties of LaH_{10}, perhaps by introducing some crucial third element to create a novel ternary compound. The problem was, up until now, that no general models of ternary superconducting systems existed determine how much we can improve the properties of polyhydrides by introducing a third element into the system. In this work, we have cleared the way by eliminating this uncertainty. We investigated the La–Nd–H system under pressure and found that the doping of polyhydrides obeys Anderson's theorem proposed for conventional superconductors back in 1959 [4].

Anderson's theorem [4] states that nonmagnetic impurities do not affect the order parameter in the conventional Bardeen–Cooper–Schrieffer (BCS) theory [3], whereas scattering on magnetic centers (e.g., Nd, Eu, Fe, Gd, etc.) is very efficient in destroying electron-electron pairing. Nonmagnetic and magnetic impurities are equally detrimental to the critical temperature T_C of unconventional superconducting states [5]. Therefore, the introduction of such impurities can provide important information on the mechanism of pairing in LaH_{10} under pressure.

The first step in our work [6] was the laser-assisted synthesis of La–Nd polyhydrides in diamond anvil cells (DACs). X-ray diffraction experiments (XRD) at high pressures were performed at SPring-8 **BL10XU**. The analysis of the XRD showed that we successfully synthesized ternary polyhydride (La, Nd)H_{10} containing about 9 at% of Nd atoms, randomly distributed in a LaH_{10}-like metal sublattice (Fig. 1). Transport measurements demonstrated that the addition of magnetic Nd leads to a significant suppression of superconductivity in LaH_{10}: each atomic % of Nd causes a decrease in T_C by 10–11 K (Fig. 2(b)). Superconductivity in the (La, Nd)H_{10} hydrides disappears at a critical concentration of Nd of about 15–20 at%. The pronounced suppression of superconductivity in LaH_{10} by magnetic Nd atoms and the robustness of T_C with respect to nonmagnetic impurities (such as B and N from ammonia borane, C and CH_4 from diamond anvils, Y [7], Al [8], etc.) within Anderson's theorem indicate the isotropic character of electron–phonon pairing in LaH_{10}.

The applicability of Anderson's theorem is limited by the following two conditions:

1. Introduced 3rd element does not change the lattice symmetry of the parent polyhydride and nor lead to the appearance of new phase transitions.
2. Concentration of the doping element remains low (5–15 at%).

For many metal alloys, these conditions are not met. However, they are satisfied in Nb–Ti alloys and T_C almost does not depend on Ti concentration (x) at $x < 0.3$ (Fig. 2(a)) [8]. Surprisingly, La–Y, La–Ce, and La–Nd polyhydrides are one of the best examples of the experimental realization of Anderson's theorem due to the similarity of the physical and chemical properties of La, Y, Ce, and Nd atoms.

From the obtained results, we can draw important conclusions concerning all ternary polyhydrides. As we can see, nonmagnetic impurities do not affect T_C so much. This leads to good reproducibility of the superconducting properties of polyhydrides synthesized in many laboratories around the world. The absence of the effects of C, B and N impurities from NH_3BH_3 also casts doubt on attempts to explain the supposedly huge increase in T_C in experiments with doped H_3S and LaH_{10}. If the leading

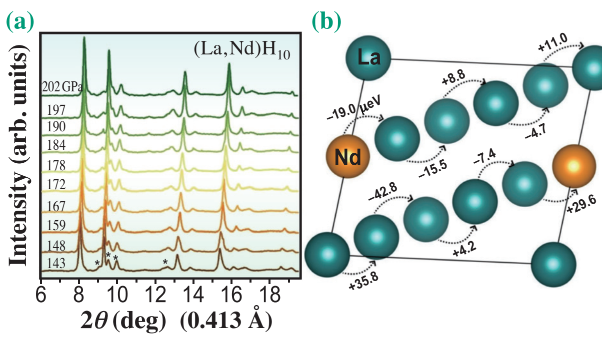

Fig. 1. **(a)** XRD patterns of (La, Nd)H_{10}, obtained from $La_{0.91}Nd_{0.09}$ alloy, recorded during decompression from 202 to 143 GPa. Asterisks mark the XRD peaks of an impurity phase. **(b)** Migration (indicated by arrows) of Nd atoms in the LaH_{10} lattice. The values given with the arrows are the formation energy differences ΔH in μeV/atom between the corresponding $P1$-La_9NdH_{100} modifications. For simplicity, hydrogen is not shown. Such low barriers between structures indicate that Nd atoms are randomly distributed in the La sublattice of LaH_{10}.

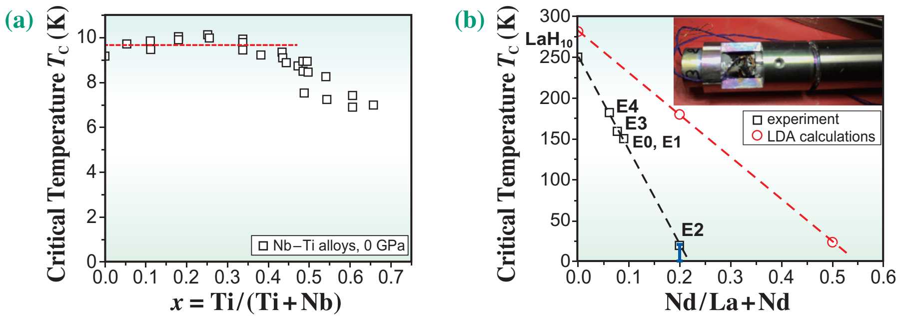

Fig. 2. **(a)** Transition temperature of Nb–Ti alloys *vs* their composition at 0 GPa [9]. Data points are the midpoints of transitions. **(b)** Experimental and calculated (using the local density approximation) dependences of T_C on the concentration of Nd in $(La,Nd)H_{10}$ at 170–180 GPa. Inset: DAC for electrical measurements in pulsed magnetic fields.

mechanism in compressed polyhydrides is the electron–phonon interaction, as most results of experimental and theoretical studies suggest, then it is impossible to expect a significant effect of a small additive, such as carbon or methane in H_3S.

The partial replacement of La atoms by magnetic Nd atoms results in a decrease in not only T_C but also the upper critical field $\mu_0 H_{C2}(0)$, which makes the upper critical field $H_{C2}(0)$ attainable for existing pulse magnets. Using strong pulsed magnetic fields up to 68 T, we constructed the magnetic phase diagram of the $(La,Nd)H_{10}$ superhydride; the magnetic phase diagram appears to be surprisingly linear with $H_{C2} \propto |T - T_C|$. This discovery motivated us to look at the behavior of other hydride superconductors as well. Figure 3 shows that several known superhydrides have a linear H_{C2}(T), and the coefficient $\alpha = -dH_{C2}/dT$ varies in a quite narrow range, $\alpha = 0.9 \pm 0.3$. This leads to the interesting conclusion that the upper critical field in many compressed polyhydrides can be expressed as a linear dependence, $H_{C2}(0) = \alpha T_C$.

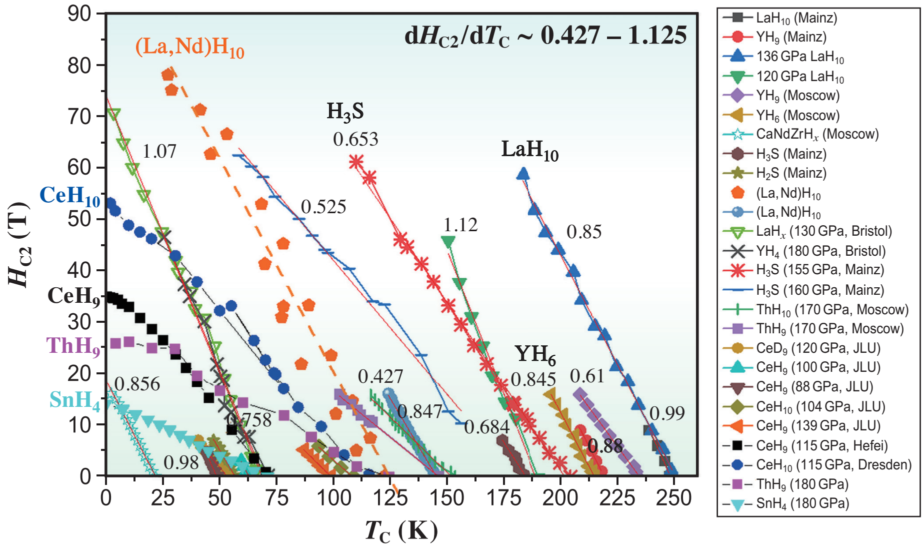

Fig. 3. Magnetic phase diagrams of superhydrides. The legend on the right shows the proposed chemical formula and the research group (city) that investigated the compound.

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