

Anatomy of the band structure of the newest apparent near-ambient superconductor $\text{LuH}_{3-x}\text{N}_x$

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Introduction. The discovery of high temperature superconductivity under high pressure around 100–250 GPa with T_c about 203 K in a hydrogen-containing H_3S system [1] gave rise to a flow of experimental and theoretical works (see reviews [2–7]). The subsequent discovery of near-room temperature superconductivity in hydrides brings us back to the classical electron-phonon superconducting pairing, very likely consistent with the Bardeen–Cooper–Schrieffer (BCS) theory. Remarkably, there appeared many different classes of hydrides under pressure and band structure calculations (density functional theory – DFT) have shown the ability to predict crystal structures and T_c values for many of those hydrides (for review see [7]).

Here we address the issue of superconductivity in the recently discovered nitrogen-doped lutetium hydride LuH_xN_y where $T_c = 294$ K was claimed to occur at 10 kbar [8]. This result has sparked intensive theoretical and experimental research. In this paper we performed DFT+U band structure calculations for both parent LuH_3 and nitrogen doped $\text{LuH}_{2.75}\text{N}_{0.25}$. In particular, our analysis reveals the contribution of nitrogen bands to the density of states at the Fermi level which might facilitate formation of superconducting state. We also provide simple BCS-analysis of T_c for materials under consideration.

Computational details. The calculations were performed within the DFT+U approximation in the VASP software package [9]. The generalized gradient approximation (GGA) in the form of the Perdew–Burke–Ernzerhof (PBE) exchange-correlation functional [10] was employed. The strong onsite Coulomb repulsion of Lu-4f electrons was described with the DFT+U scheme with the Dudarev approach [11] ($U = 5.0$ eV).

We consider LuH_3 and nitrogen doped $\text{LuH}_{2.75}\text{N}_{0.25}$ (spatial symmetry group $Fm\bar{3}m$) [8] at ambient pressure. The H atoms are located in positions with tetrahedral (0.25, 0.25, 0.25) and octahedral (0.5, 0, 0) surroundings of Lu atoms. The Lu atoms are located at the point of origin, (0, 0, 0). To get $\text{LuH}_{2.75}\text{N}_{0.25}$ only one of the four hydrogen atoms is replaced by the nitrogen atom, since, such substitution, as shown in [8], leads to a metal. The ion relaxation was done for $\text{LuH}_{2.75}\text{N}_{0.25}$. Wannier functions were obtained using the Wannier90 [12] package with projection onto H-1s in octahedral environment and N-2p orbitals.

Results. The comparison of GGA+U total densities of states (DOS) and band dispersions between LuH_3 and $\text{LuH}_{2.75}\text{N}_{0.25}$ is shown on Fig. 1.

Nitrogen doping leads to formation of a wide peak in the total DOS just below the Fermi level, in contrast to the case of stoichiometric LuH_3 . The most important is that the total DOS at the Fermi level increases almost by a factor of two in $\text{LuH}_{2.75}\text{N}_{0.25}$, from 1.5 to 2.6 states/eV/u.c. (left panel of Fig. 1). Substitution of hydrogen atom by nitrogen one gives four additional holes per unit cell which leads to ~ 1.8 eV lowering of the Fermi level in $\text{LuH}_{2.75}\text{N}_{0.25}$ as compared to LuH_3 . Although the bands can not be superposed by their simple shift, some features are quite similar for both systems.

In the case of $\text{LuH}_{2.75}\text{N}_{0.25}$, several bands cross the Fermi level at each high-symmetry direction, while for LuH_3 only couple of bands near Γ -point cross the Fermi level. For $\text{LuH}_{2.75}\text{N}_{0.25}$ at the Fermi level there are pronounced flat band regions in the vicinity of R and X points, which are missing for LuH_3 .

In order to define the minimal orbital basis set for $\text{LuH}_{2.75}\text{N}_{0.25}$, needed to reproduce electronic bands near the Fermi level we performed the projection onto Wannier functions. The orbital character of the bands built

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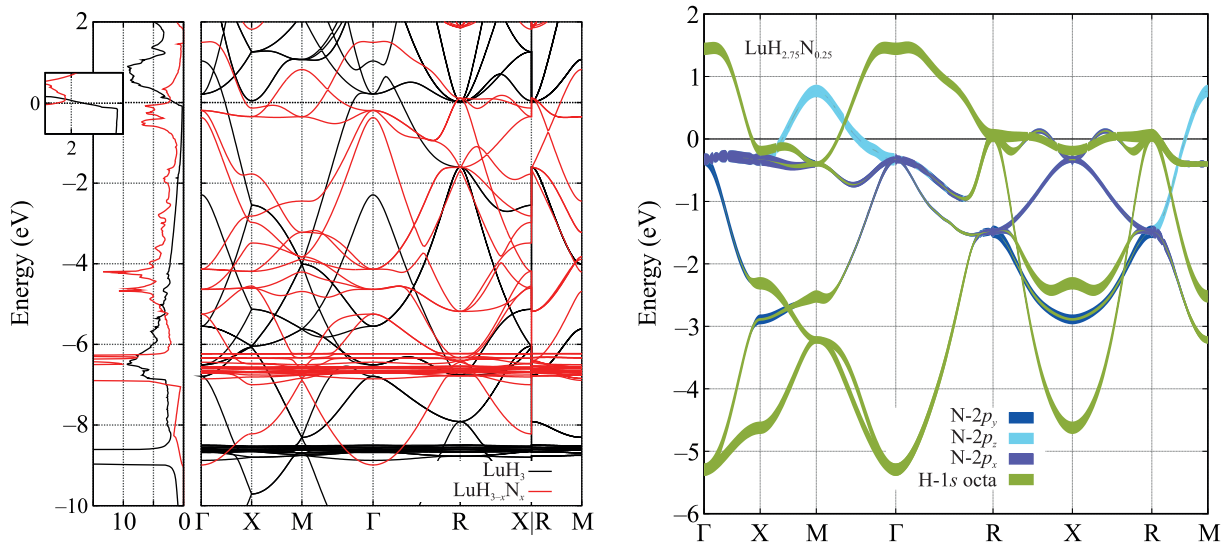


Fig. 1. (Color online) Left panel: GGA + U total DOS (right side) and band dispersions of LuH₃ (black lines), LuH_{2.75}N_{0.25} (red lines on the right side). The inset (left panel) shows DOS near the Fermi level $[-0.5; 0.5]$ eV. Right panel: The bands projected on Wannier function with linewidth showing contributions of H-1s octahedral and N-2p states

on the projected Wannier functions are presented in Fig. 1.

The most important parameter of superconducting materials is its critical superconducting temperature T_c . To get its simplest rough theoretical estimate, we use the well known BSC equation $T_c = 1.14 \omega_D \exp(-1/\lambda)$. We employ $T_c^{\text{LuH}_3} = 62$ K value [13] obtained with anisotropic Migdal–Eliashberg formalism with $\omega_D = 19.3$ meV = 220 K. With the values we can exclude the pairing constant g from BCS equation and then estimate T_c^{LuHN} value for nitrogen doped material, assuming that ω_D and g do not change significantly. The increase of DOS at the Fermi level gives $T_c \sim 111$ K for LuH_{2.75}N_{0.25}.

Conclusion. Our findings suggest that nitrogen doping of LuH₃ can significantly alter the electronic properties of the material (bringing light elements N-2p and H-1s states at the Fermi level), facilitating the occurrence of superconductivity. In particular, nitrogen doping doubles the value of DOS at the Fermi level. Simple BCS analysis suggests that for LuH_{2.75}N_{0.25} the critical temperature might exceed 100 K, and one can even increase it with further hole doping by nitrogen up to 180 K.

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