Superconductivity and Equation of State of Distorted fcc-Lanthanum above Megabar Pressures

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Abstract

Lanthanum (La) is the first member of the rare-earth series of elements that has recently raised considerable interest due to its unique superhydride LaH₁₀⁻δ and its superconducting properties. Although several studies have found superconductivity and phase transitions in metallic La, there has been a lack of experimental evidence for the equation of state and superconductivity over one megabar pressure. Herein, we extend pressure range up to 163 GPa to explore the equation of state and superconductivity of La via electrical transport and X-ray diffraction measurements. Le Bail refinement of experimental XRD patterns indicated that above 130 GPa orthorhombically distorted fcc-La (Fm̅3m→Fmmm) phase emerges with the fitted parameters of 3rd order Birch-Murnaghan equation of state: V₁₀₀ = 16.48 (7) Å³, B₁₀₀ = 208 (26) GPa and B₁₀₀′ = 9 (1) in the range of 100-160 GPa. The superconductivity emerges in the newly distorted fcc-La with an onset critical temperature Tc of 6.1 K at 100 GPa, decreases to 1.5 K at 130 GPa, then begins to grow and reach 3.3 K at 150 GPa. We have extrapolated the upper critical magnetic field μ₀Hc₂ as 0.32 T (Gingburg-Landau) and 0.43 T (WHH formalism) at 140 GPa. Ab initio calculations confirm electron-phonon coupling mechanism with predicted Tc (A-D) = 2.2 K (μ* = 0.195), dTc/dP = 0.11-0.13 K/GPa and μ₀Hc₂(0) = 0.4 T and coherence length ξBCS = 28 nm at 140 GPa.

Keywords: lanthanum, superconductivity, equation of state, high pressure
Introduction

Since the first discovery of superconductivity (SC) in 1911 [1], scientists have searched for materials that can conduct electricity without resistance below a superconducting transition temperature ($T_c$). So far, tremendous efforts have been devoted to exploring high-$T_c$ SC in a variety of materials such as cuprates [2], iron-based superconductors [3], and hydrogen-rich compounds [4]. Among them, SC in hydrides has been successfully realized through the discovery of novel compounds formed in the S-H system at modest pressures with a $T_c$ up to 203 K. The discovery of high-temperature superconductivity in $Im\bar{3}m$-H$_2$S by theoretical methods [5-6] and experimental measurements [7-9] reveals that even room-temperature SC can be achieved in hydrogen-rich materials. Hence, the search for superconducting polyhydrides at very high pressures has raised a new round of research upsurge in physics. Metal hydrides are interesting materials for realizing high-$T_c$ superconductivity by forming unconventionally high stoichiometric ratio. In particular, metal lanthanum (La) can react with hydrogen triggering a non-stoichiometric $Fm\bar{3}m$-LaH$_{10}$ (closer to La$_2$H$_{21}$), which recently has been reported as high-$T_c$ superconductor with critical temperature above 250 K at 170 GPa [10-11]. In the framework of total experimental and theoretical investigation of La-H system, the study of structural and electronic properties of metal La at ultrahigh pressures is necessary for designing of electronic high-performance SC-devices on the basis of La superhydrides.

The unique physical and chemical properties of the rare-earth metals have attracted interest for decades. Mostly trivalent rare-earth metals from La through Lu possess a similar $d$-electron character near the Fermi energy, neither of which is superconducting at ambient pressure except metal La [12]. As the first member of the rare-earth series of elements, La can exist in both double hexagonal-close-packed ($dhcp$) phase and face-centered-cubic ($fcc$) phase, while a $bcc$ structure is favorable at high temperatures near the melting point [13-15]. Due to high electronic density of states at the Fermi surface and specific phonon spectrum, one would expect strong electron phonon coupling, and therefore a reasonable high superconducting transition temperature observed for La. The $dhcp$ and $fcc$ phases at ambient pressure yield superconducting transition temperatures $T_c$ near 5 and 6 K, respectively [16]. Both structures show significant increases in $T_c$ with pressure ($dT_c/dP \sim 0.87-1$ K/GPa) and at 4 GPa reach $T_c \approx 9.3$ K, and even 13 K around 17 GPa [17-18]. The $dhcp$ phase undergoes several structural transformations, firstly transforms to the $fcc$ structure near 2.2 GPa, shifts at about 5.4 GPa to a distorted $fcc$ ($R\bar{3}m$) structure, then returns to the original $fcc$ phase at 60 GPa [16-17]. At the same time, critical temperature demonstrates complex behavior with several “waves” (broad maximums) and anomalies up to 50 GPa [17]. Until now, the equation of state (EoS) and superconductivity of metal La were not studied under high pressure conditions (over 100 GPa).

In this work, we present new data of La by comprehensive studies on the structural and superconducting properties up to 165 GPa. We have reproduced the earlier reported results, and expanded the pressure range of the superconducting properties of La above megabar pressure. The superconductivity survives in La with an onset critical temperature $T_c$ of 6.1 K at 100 GPa and then decreases to 3.3 K at 150 GPa. We have extrapolated the upper critical magnetic field $\sim 0.32-0.43$ T upon applied magnetic field at 140 GPa. The combined high-pressure structure investigations with synchrotron radiation indicated that the superconductivity occurred in a newly reported orthorhombically distorted $fcc$-La ($Fm\bar{3}m \rightarrow Fmmm$) structure above 100 GPa.
Experimental and Theoretical Methods

We have used the target La sample purchased from Alfa Aesar with the purity of 99.9%. The assembly used for electrical resistance measurements is shown in Fig. 1. For the electrical conductivity measurements, we used a piston-cylinder diamond anvil cell (DAC) made by Be-Cu material and the diamonds had a culet of diameter of 100 µm and bevelled at 8° to a diameter of about 300 µm. The sample chamber consisted of a tungsten gasket with Al₂O₃ insulating layer. The excess Al₂O₃ is also used as the pressure-transmitting media (PTM). The piston diamond was coated with four 1 µm thick Mo electrodes, which is connected to the external wires using a combination of 25 µm thick Pt shoes soldered onto brass holders. The 5 µm thick La sample was placed on the Mo electrodes and packed in with aluminum oxide in an argon glovebox. The DAC was clamped inside the argon-protected glovebox. Pressure was determined from the Raman shift of the diamond anvil edge excited with 532 nm laser [19].

FIG. 1. (a) Schematic of the assembly used for electrical resistance measurements. The sample chamber consisted of a tungsten outer gasket (W) with an insulating Al₂O₃ and epoxy. (b) Micro pictures of a sample at 140 GPa irradiated by top light. Inset shows the sample irradiated by bottom light.

The synchrotron X-ray diffraction patterns of La sample in a pneumatic DAC with 50 µm culet were recorded on the ID-27 synchrotron beamline at the European Synchrotron Radiation Facility (Grenoble, France) with the use of a focused (1.7 × 2.3 µm) monochromatic X-ray beam of 33 keV (λ = 0.3738 Å) and a Perkin-Elmer area detector placed at a distance of 364.12 mm from the sample. MgO is used as PTM and pressure gauge [20]. The exposure time was 30–100 s. CeO₂ standard was used for the distance calibration. The experimental X-ray diffraction images were analyzed and integrated using the Dioptas software package (version 0.4) [21].

Calculations of superconducting $T_c$ were carried out using QUANTUM ESPRESSO (QE) package [22]. Phonon frequencies and electron-phonon coupling (EPC) coefficients were computed using density-functional perturbation theory [23], employing plane-wave pseudopotential (PP) method, Perdew-Burke-Ernzerhof and Perdew-Zunger exchange-correlation functionals [24]. In our ab initio calculations of the electron-phonon coupling (EPC) parameter $\lambda$, the first Brillouin zone was sampled using $2\times2\times2$ and $4\times4\times4$ q-points mesh, and a denser $24\times24\times24$ k-points mesh (with Gaussian smearing and $\sigma = 0.025$ Ry, which approximates the zero-width limits in the calculation of $\lambda$). Critical temperature $T_c$ was calculated from the Allen-Dynes equations [25].

We also calculated the EoS for distorted fcc-La, by performing the structural relaxations of cell parameters at various pressures using density-functional theory (DFT) within the generalized gradient approximation.
(Perdew-Burke-Ernzerhof and Perdew-Zunger functionals), and the projector-augmented wave (PAW) method as implemented in the VASP code [26]. Plane wave kinetic energy cutoff was set to 600 eV and the Brillouin zone was sampled using Γ-centered k-points meshes with resolution $2\pi \times 0.05$ Å$^{-1}$.

**Results and discussion**

The EoS of metallic La has been investigated in the DAC with 50 μm culet and MgO as the PTM. As shown in Fig. 2a, at 107 GPa, the Bragg peaks of La in the experimental patterns can be well indexed by a distorted fcc-La structure (with space group Fmmm) which hasn’t been discovered in other rare-earth metals before [27]. Le Bail refinements of experimental XRD pattern with distorted fcc-La structure at various pressures can be seen in Supporting Information Figure S1. The Rietveld refinement of the experimental highest pressure of 163.3 GPa is also shown in Fig. 2a, indicating that the newly found distorted fcc-La structure can be preserved up to 163.3 GPa without any phase transitions. The refined lattice parameters and the unit cell volume of La sample upon compression run as a function of pressure are presented in Supporting Information Figure S2 and Fig. 2c (also given in Supporting Information Table S1). The diagrams show the anisotropy behavior of La sample during compression: $da/dP$ and $dc/dP$ are close and can be roughly approximated by linear functions $a(P) = 4.2379 - 0.0029 \cdot P$ (Å) and $c(P) = 4.2601 - 0.0027 \cdot P$ (Å), while $db/dP$ has sufficiently different declination and $b(P) = 4.2816 - 0.0021 \cdot P$ (Å) (see Supporting Information Figure S2). The pressure-volume points obtained from the XRD data are plotted in Fig. 2c along with the calculated EoS using density-functional theory (DFT) with three different pseudopotentials. We can see that the calculated results of distorted fcc-La cell depending on the used pseudopotentials. PAW PBE La-pseudopotential expands cell and increases the volume, while Perdew-Zunger pseudopotential compresses the cell. The best consistency with experimental $V(P)$ was observed for PBE Vanderbilt ultrasoft La-pseudopotential, which is up to only 6% lower than the experimental values. In order to determine the main parameters of the EoS, the obtained pressure-volume data was fitted by third-order Birch-Murnaghan equation [28]:

$$P = \frac{3B_0}{2} \left\{ \left( \frac{V}{V_0} \right)^{-\frac{7}{3}} - \left( \frac{V}{V_0} \right)^{-\frac{5}{3}} \right\} \left\{ 1 + \frac{3}{4} \left( B'_0 - 4 \right) \left[ \left( \frac{V}{V_0} \right)^{-\frac{2}{3}} - 1 \right] \right\}$$

(1)

namely $V_0$, $B_0$ and $B'_0$, where $V_0$ is equilibrium cell volume (here at 100 GPa, index is “100”), $B_0$ is bulk modulus and $B'_0$ is derivative of bulk modulus with respect to pressure. The fitted result yields equilibrium volume $V_{100} = 16.48$ (7) Å$^3$ and an isothermal bulk modulus $B_{100} = 208$ (26) GPa with its pressure derivative $B'_{100} = 9$ (1) (see Supporting Information). The bulk modulus of distorted fcc-La Fmmm phase is greatly larger than those reported fcc- and dhcp-La phase, both of whose bulk modulus is just 24 GPa [29-30].
FIG. 2. (a) Le Bail refinement of experimental XRD pattern of La sample at 107 GPa and 163.3 GPa, respectively. The shaded areas represent the unknown peaks (impurities) and were also excluded from the analysis. The positions of the Bragg reflections from La and MgO are marked by black vertical sticks. (b) Pressure-volume relations for La sample from experimental and calculated results. Solid spheres represent data from experiment. The red dashed curve show EoS fit to third-order Birch-Murnaghan equation. The blue circle, green square and yellow triangle indicates the calculated data by using different pseudopotentials.

The superconductivity of metallic lanthanum has been investigated in the DAC with 100 μm culet and Al₂O₃ as the PTM. After loading the La sample into the DAC (the detailed setup shown in Fig.1), we have measured the evolution of electrical resistance as a function of temperature for La sample at various pressures, as shown in Supporting Information Figure S4 and Fig. 3a. Upon cooling at 0 GPa from 300 K down to 1.6 K, the overall behavior of resistance displays a typical metallic-like feature then the resistance decreases sharply to zero resistance at 5.2 K. The superconducting transition critical temperature \( T_c \) defined by the intersect temperature between the horizontal and vertical resistance line, as shown in Fig. 3b. With increasing pressure, the change of \( T_c \) as a function of pressure is very close to the reported data below 60 GPa in Fig. 5 [15, 17]. Further compression, Fig. 3a shows the evolution of electrical resistance as a function of the temperature above 100 GPa. It is found that the superconducting transition can be triggered from both cooling and heating process. The data collected at 140 GPa showed that resistance dropped to zero at lower temperature with 2.2 K, while at 150 GPa, a clear superconducting transition was observed with \( T_c \) reached about 3.3 K (Fig. 3b). The observed superconducting transitions were sharp with the transition width (from 10% - 90%) of the normal state resistance at \( T_{\text{onset}} \) around 0.5 K, indicating the good homogeneity of the superconducting phase. The beginning of the transition to superconducting state at 140 GPa can also be fixed at the initial point of the resistance growth (~ 4.8 K) which is caused by the presence of distributed SC-NSC grain contacts [31]. However, for 150 GPa such definition of \( T_c \) is no longer applicable. The pressure dependence of superconducting \( T_c \) of metallic La exhibit approximately linear behavior in 100-130 and 130-150 GPa ranges.
The resistance of the La sample as a function of temperature at different pressures during cooling and heating. The shaded region is amplified and displayed in (b), and the electrical resistance during cooling shows a superconducting transition at 140 GPa and 150 GPa, respectively. Dashed lines show the definition of superconducting transition temperature $T_c$.

To assure what has been observed in Supporting Information Figure S4 and Fig. 3 is indeed the superconducting transition, we further conducted the electrical resistance measurements around the transition temperature at variant external magnetic field. We have checked the data at 53 GPa and 140 GPa with applied magnetic $H$ and the measured results of 53 GPa can be found in Supporting Information Figure S5.

Fig. 4a shows the measured resistance at 140 GPa with applied magnetic $H$ of 0.025, 0.05, 0.075 and 0.1 T. The critical temperature $T_c$ decreased with increasing magnetic field, indicating strong evidence that the transition is superconductivity in nature. Much higher fields are required to suppress the superconductivity. To figure out the upper critical magnetic field $H_{c2}(0)$, the extrapolation method combined with the Ginzburg-Landau (LG) equation

$$\mu_0 H_{c2}(T) = \mu_0 H_{c2}(0) \left( 1 - \frac{T^2}{T_c^2} \right)$$

was applied. The extrapolation ($R^2 = 0.98$) of transition temperature gives an estimate of $\mu_0 H_{c2}(0) = 0.32-0.33$ T. Werthamer-Helfand-Hohenberg (WHH) model [32] for critical magnetic field, simplified by Baumgartner [33]:

$$\mu_0 H_{c2}(T) = \frac{\mu_0 H_{c2}(0)}{0.693} \left( 1 - \frac{T}{T_c} \right)^{-0.153} \left( 1 - \frac{T}{T_c} \right)^2 - 0.152 \left( 1 - \frac{T}{T_c} \right)^4$$

leads to $\mu_0 H_{c2}(0) = 0.42-0.43$ T in a good agreement with calculated value (0.4 T, Table 1).

We also have calculated superconducting parameters of La at different pressures to explore its mechanism. To compare our calculations with experimental results at 50 GPa, we firstly computed superconducting parameters of slightly distorted fcc-La ($R$-3$m$) in PZ and PBE pseudopotentials which led to results: $\lambda = 1.067$, $\omega_{\log} = 112.7$ K and $T_c = 9.3$ K at $\mu^* = 0.1$ (50 GPa). Eliashberg functions $\alpha^2 F(\omega)$ of distorted fcc-La with different $\sigma$-broadening (QE) at 50 GPa is presented in Supporting Information Figure S7. Thus, experimental $T_c$ of 10.5 K (Ref. 17, 50 GPa) is consistent with our experimental results (53 GPa, 10.5 K) and close to the predicted results
(9.3 K). Thus, we are confidence that the current theoretical calculations of $T_c$ and $H_{c2}$ as function of are applied to the results of both our new and earlier studies by Tissen et al. [17] and the much earlier measurements of Wittig et al. [15]. At 150 GPa, we took into account that phonon spectrum of La ends at 350 cm$^{-1}$ ($\approx$ 0.043 eV, Fig. 4b and 4c). On such energies density of states near $E_F \pm \hbar \omega_{\text{max}}$ is almost constant ($\approx$ 20.1 states/Ry/cell-La/atom) that allows us to apply constant DOS approximation [34] and take $\alpha^*$ corresponding to almost zero broadening in QE output. Results obtained with PBE ultrasoft Vanderbilt pseudopotential of La are $\lambda = 0.75$, $\omega_{\text{log}} = 193$ K, and differ from the results received with Goedecker-Hartwigsen-Hutter-Teter PZ pseudopotential: $\lambda = 0.68$, $\omega_{\text{log}} = 287$ K, $T_c$ (A-D) = 9.4 K for $\mu^* = 0.10$, and $T_c$ (A-D) = 5.7 K for $\mu^* = 0.15$ at 150 GPa. Later value is close to $T_c$ defined during the cooling cycle that is why we included $\mu^* = 0.15$ related values in Table 1. There is no significant difference in $T_c$ and $\mu^*$ between used pseudopotentials. Experimental critical temperature 3.3 K corresponds to anomalous value of $\mu^* = 0.195$. At this Coloumb pseudopotential McMillan isotope coefficient is quite small $\beta = 0.21$ (it increases to 0.37 at $\mu^* = 0.15$), while coherence length $\xi_{\text{BCS}} = 0.5 \sqrt{\hbar/\pi e H_{c2}}$ is 23 nm which is about 35% lower than for metallic lanthanum at 0 GPa (36 nm) [35]. Additional calculations shows that at 180 GPa expected experimental $T_c$ increases over 5.2 K (at $\mu^* = 0.195$) and $\mu_0 H_{c2} = 0.9$ T while decreasing pressure to 130 GPa leads to calculated $T_c$ about 1.5 K. The observed change in the sign of $dT_c/dP$ (Fig. 5), $\mu^*$ and $N(E_F)$ over 130 GPa is probably caused by onset of the $Fm\overline{3}m \rightarrow Fmmm$ phase transition in La.

![Figure 4](image_url)

**FIG. 4.** (a) The superconducting transition of the La sample at 140 GPa with applied magnetic field. Inset shows the change of $T_c$ with magnetic field and the solid line in red represents fitting by the WHH and LG equation. (b) The calculated superconducting parameters of distorted fcc-La phase at 150 GPa. (c) The calculated phonon density of state of La at 150 GPa.

| Table 1. Parameters of superconducting state of metallic La at 130-180 GPa calculated by Allen-Dynes (A-D) equation. Here $\gamma$ is Sommerfeld constant, $\Delta C(T_c)$ – is the heat capacity jump, $\mu^*$ is 0.195 (0.15). |
|---|---|---|---|---|
| **Parameters** | **130 GPa** | **140 GPa** | **150 GPa** | **180 GPa** |
| $DOS (E_F)$, states/eV/atom | 0.71 | 0.72 | 0.73 | 0.55 |
| $\lambda$ | 0.59 | 0.67 | 0.68 | 1.04 |
| $\omega_{\text{log}}$, K | 285 | 208 | 287 | 113 |
| $T_c$ (A-D), K | 1.5 (3.4) | 2.2 (4.1) | 3.3 (5.7) | 5.2 (6.9) |
| $\Delta(0)$, meV | 0.2 (0.5) | 0.3 (0.6) | 0.5 (0.9) | 0.8 (1.2) |
| $\mu_0 H_{c2}(0)$, T | 0.3 (0.6) | 0.4 (0.75) | 0.6 (1.0) | 0.9 (1.2) |
ΔC/Tc, mJ/mol·K² | 7.6 (7.8) | 8.2 (8.4) | 8.3 (8.4) | 9.1 (9.9)  
γ, mJ/mol·K²     | 5.3      | 5.6      | 5.85     | 5.2       
Rₐ = 2Δ(0)/kₙTc | 3.53 (3.55) | 3.55 (3.6) | 3.55 (3.6) | 3.75 (3.9) 

To sum up, Fig. 5 shows the evolution of the crystal structure and the superconducting transition temperature as a function of pressure in the form of a phase diagram. For clear comparison, we have listed our data and earlier results together. It can be seen that the differences in Tc between our work and earlier studies [15, 17] are very small, and this tiny difference could be contributed to the different hydrostatic conditions with various PTM. For the electrical conductivity measurements, we used excess Al₂O₃ as the PTM. As an example, we have investigated the pressure distribution in the superconducting electrical resistance measurements at about 132 GPa, as shown in the Supporting Information Figure S9. From this figure, we can see that the largest pressure difference is 7 GPa away from the sample center with 20 micron. Besides, considering that MgO solid is softer than Al₂O₃ and probably offering a better hydrostatic environment at ultrahigh pressures, so we proposed that the pressure difference in the XRD experiments is no more than 7 GPa. Thus the present experiments show a good quasihydostatic pressure condition.

**FIG. 5.** Phase diagram and superconducting transition temperature Tc of La as a function of pressure. The blue squares and pentagram represents data from Tissen et al. [17] and Wittig et al. [15], and the purple spheres and wine diamonds are from our experimental and calculated results. The fcc to dist. fcc (Fmmm) phase transition indicated by red dashed line is marked at 100 GPa from the starting pressure of our XRD work, while the other transition ranges are determined from earlier studies [15-17].

**Conclusions**

In this work, we have studied crystal structure of metallic La at pressures up to 163 GPa, and demonstrated that fcc-La undergoes orthorhombic distortion in this pressure range. The third-order Birch-Murnaghan EoS is defined with parameters \( V_{100} = 16.48 \) (7) \( Å^3 \), \( B_{100} = 208 \) (26) GPa and \( B'_{100} = 9 \) (1) above megabar pressure. The superconducting transitions are detected in distorted fcc-La at 6.1 K (100 GPa) and at 3.3 K (150 GPa), by means of four-probe resistance measurements. Investigated influence of external magnetic field (0-0.1 T) on \( T_c \) at 140
GPa allows us to estimate upper critical magnetic field $\mu_0H_{c2}(0)$ from 0.32 to 0.43 T according to Ginzburg-Landau and Werthamer-Helfand-Hohenberg models, respectively. Calculations of electron-phonon interaction within classical BCS mechanism show good stability and point to increased Coloumb pseudopotential $\mu^*=0.195$, while calculations with common $\mu^*$ interval (0.1-0.15) lead to overestimated $T_c$ (5.7-9.4 K at 150 GPa). The calculated upper critical magnetic field $\mu_0H_{c2}(0) = 0.4$ T and gradient $dT_c/dP = 0.11-0.13$ K/GPa are quite close to experimental values at 140 GPa. Thus, the superconducting properties of metallic lanthanum exhibit approximately linear behavior in 100-130 and 130-150 GPa ranges and can be described within the classical electron-phonon pairing mechanism.

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SUPPORTING INFORMATION

Superconductivity and Equation of State of Distorted fcc-Lanthanum above Megabar Pressures

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CONTENT

1. Equations for calculating $T_c$ and related parameters…………………………………………………………S2
2. Structural information and results of X-ray diffraction studies……………………………………………………S3
3. Additional physical properties…………………………………………………………………………………………S6
1. Equations for calculating $T_c$ and related parameters

Superconducting transition temperature $T_c$ was estimated by using Allen-Dynes formula:

$$T_c = \omega_{\log} \frac{f_1 f_2}{1.2 \exp \left( \frac{-1.04(1 + \lambda)}{\lambda - \mu^* - 0.62 \lambda \mu^*} \right)} \quad \text{(S1)}$$

where multiplication of Allen-Dynes coefficients are:

$$f_1 f_2 = \sqrt{1 + \left( \frac{\lambda}{2.46(1 + 3.8 \mu^*)} \right)^{\frac{3}{2}}} \cdot \left( 1 - \frac{\lambda^2 (1 - \omega_2 / \omega_{\log})}{\lambda^2 + 3.312 (1 + 6.3 \mu^*)^2} \right) \quad \text{(S2)}$$

The EPC constant $\lambda$ and logarithmic average frequency $\omega_{\log}$ and the mean square frequency were calculated as:

$$\lambda = 2 \int_0^{\omega_{\max}} \frac{a^2 F(\omega)}{\omega} d\omega \quad \text{(S1)}$$

and

$$\omega_{\log} = e^{\frac{2}{\lambda} \int_0^{\omega_{\max}} \frac{d\omega}{a^2 F(\omega) \ln(\omega)}} , \quad \omega_2 = \sqrt{\frac{1}{\lambda} \int_0^{\omega_{\max}} \left[ \frac{2 a^2 F(\omega)}{\omega} \right] \omega^2 d\omega} \quad \text{(S2)}$$

To calculate isotopic coefficient $\beta$ the Allen-Dynes interpolation formulas were used:

$$\beta_{M/M} = -\frac{d}{d \ln M} \frac{\ln T_c}{T_c} = \frac{1}{2} \left[ 1 - \frac{1.04(1 + \lambda)(1 + 0.62 \lambda)}{[\lambda - \mu^*(1 + 0.62 \lambda)]^2} \mu^* \right]$$

$$\beta_{AD} = \beta_{M/M} - \frac{2.34 \mu^* \lambda^{3/2}}{(2.46 + 9.25 \mu^*) \cdot ((2.46 + 9.25 \mu^*)^{3/2} + \lambda^{3/2})} - \frac{130.4 \cdot \mu^* \lambda^* (1 + 6.3 \mu^*) \left( 1 - \frac{\omega_{\log}}{\omega_2} \right) \frac{\omega_{\log}}{\omega_2}}{8.28 + 104 \mu^* + 329 \mu^*^2 + 2.5 \cdot \lambda^2 \left( \frac{\omega_{\log}}{\omega_2} \right)^2} \quad \text{(S5)}$$

where the last two correction terms are usually small (~0.01).

Sommerfeld constant was found as

$$\gamma = \frac{2}{3} \pi^2 k_B^2 N(0)(1 + \lambda) \quad \text{(S7)}$$

and was used to estimate the upper critical magnetic field and the superconductive gap by well-known semi-empirical equations of the BCS theory (see J. P. Carbotte, Rev. Mod. Phys., 62(4), 1990, equations 4.1 and 5.11), working satisfactorily for $T_c / \omega_{\log} < 0.25$:

$$\frac{\gamma T_c^2}{(\mu_0 H_{c1}(0))^2} = 0.168 \left[ 1 - 12.2 \left( \frac{T_c}{\omega_{\log}} \right)^2 \ln \left( \frac{\omega_{\log}}{3T_c} \right) \right] \quad \text{(S8)}$$

$$\frac{2 \Delta(0)}{k_B T_c} = 3.53 \left[ 1 + 12.5 \left( \frac{T_c}{\omega_{\log}} \right)^2 \ln \left( \frac{\omega_{\log}}{2T_c} \right) \right] \quad \text{(S9)}$$
### 2. Structural information and results of X-ray diffraction studies

**Table S1.** Experimental cell parameters for orthorhombically distorted $fcc$-La.

| Pressure, GPa | $a$, Å  | $b$, Å  | $c$, Å  | $V$, Å$^3$/cell | $V$, Å$^3$/atom |
|--------------|---------|---------|---------|-----------------|-----------------|
| 107          | 3.945   | 4.059   | 3.993   | 63.93           | 15.98           |
| 108.5        | 3.942   | 4.055   | 3.985   | 63.68           | 15.92           |
| 110.5        | 3.928   | 4.048   | 3.971   | 63.14           | 15.78           |
| 113.6        | 3.911   | 4.041   | 3.959   | 62.56           | 15.64           |
| 114          | 3.902   | 4.053   | 3.954   | 62.53           | 15.63           |
| 118          | 3.889   | 4.036   | 3.939   | 61.83           | 15.46           |
| 119          | 3.877   | 4.0434  | 3.927   | 61.55           | 15.39           |
| 125.4        | 3.849   | 4.037   | 3.899   | 60.586          | 15.15           |
| 138.5        | 3.815   | 3.981   | 3.866   | 58.71           | 14.68           |
| 141.2        | 3.821   | 3.968   | 3.865   | 58.6            | 14.65           |
| 148.4        | 3.796   | 3.939   | 3.852   | 57.61           | 14.40           |
| 163.3        | 3.752   | 3.94    | 3.812   | 56.37           | 14.09           |

**Table S2.** Experimental equation of state for $Fm\overline{3}m$-MgO.

| Pressure, GPa | $a$, Å  | $V$, Å$^3$ |
|--------------|---------|------------|
| 107          | 3.763   | 53.267     |
| 108.5        | 3.758   | 53.108     |
| 110.5        | 3.754   | 52.901     |
| 113.6        | 3.911   | 62.554     |
| 114          | 3.746   | 52.593     |
| 118          | 3.736   | 52.155     |
| 119          | 3.735   | 52.093     |
| 125.4        | 3.720   | 51.494     |
| 138.5        | 3.693   | 50.374     |
| 141.2        | 3.688   | 50.156     |
| 148.4        | 3.674   | 49.592     |
| 163.3        | 3.646   | 48.489     |
Table S3. The calculated cell parameters for orthorhombically distorted \( fcc\)-La by Perdew-Zunger LDA pseudopotential (QE).

| Pressure, GPa | \( a_{\text{DFT}}, \text{Å} \) | \( b_{\text{DFT}}, \text{Å} \) | \( c_{\text{DFT}}, \text{Å} \) | \( V_{\text{DFT}}, \text{Å}^3 \) |
|--------------|----------------|----------------|----------------|----------------|
| 104          | 3.921          | 3.948          | 3.925          | 15.19          |
| 108          | 3.918          | 3.940          | 3.904          | 15.07          |
| 118          | 3.876          | 3.929          | 3.874          | 14.75          |
| 130          | 3.834          | 3.918          | 3.836          | 14.40          |
| 140          | 3.804          | 3.905          | 3.808          | 14.14          |
| 144          | 3.804          | 3.879          | 3.806          | 14.04          |
| 154          | 3.774          | 3.874          | 3.776          | 13.80          |
| 180          | 3.704          | 3.862          | 3.706          | 13.25          |
| 200          | 3.663          | 3.837          | 3.665          | 12.88          |

Table S4. The calculated cell parameters for orthorhombically distorted \( fcc\)-La by PBE Vanderbilt ultrasoft pseudopotential (QE).

| Pressure, GPa | \( a_{\text{DFT}}, \text{Å} \) | \( b_{\text{DFT}}, \text{Å} \) | \( c_{\text{DFT}}, \text{Å} \) | \( V_{\text{DFT}}, \text{Å}^3 \) |
|--------------|----------------|----------------|----------------|----------------|
| 104          | 3.911          | 4.144          | 3.910          | 15.84          |
| 108          | 3.896          | 4.131          | 3.895          | 15.67          |
| 118          | 3.860          | 4.096          | 3.860          | 15.26          |
| 130          | 3.845          | 4.019          | 3.843          | 14.84          |
| 140          | 3.814          | 3.989          | 3.813          | 14.50          |
| 144          | 3.777          | 4.023          | 3.777          | 14.35          |
| 154          | 3.754          | 3.985          | 3.754          | 14.04          |
| 180          | 3.690          | 3.914          | 3.690          | 13.32          |
| 200          | 3.644          | 3.865          | 3.644          | 12.83          |

Table S5. The calculated cell parameters for orthorhombically distorted \( fcc\)-La by PAW PBE pseudopotential (VASP).

| Pressure, GPa | \( a_{\text{DFT}}, \text{Å} \) | \( b_{\text{DFT}}, \text{Å} \) | \( c_{\text{DFT}}, \text{Å} \) | \( V_{\text{DFT}}, \text{Å}^3 \) |
|--------------|----------------|----------------|----------------|----------------|
| 104          | 4.031          | 4.074          | 4.040          | 16.59          |
| 108          | 4.032          | 4.065          | 4.001          | 16.39          |
| 118          | 3.976          | 4.071          | 3.967          | 16.05          |
| 130          | 3.923          | 4.061          | 3.931          | 15.65          |
| 140          | 3.891          | 4.043          | 3.907          | 15.36          |
| 144          | 3.890          | 4.023          | 3.898          | 15.25          |
| 154          | 3.867          | 4.000          | 3.875          | 14.98          |
| 180          | 3.805          | 3.966          | 3.812          | 14.38          |
| 200          | 3.769          | 3.929          | 3.776          | 13.98          |
Figure S1. Le Bail refinements of experimental XRD pattern of La sample at various pressures. Side reflexes were excluded from the analysis.

Figure S2. The refined cell parameters of La sample at various pressures.
3. Additional physical properties

Figure S3. Photo of the DAC with 50 μm culet and La sample used in X-ray diffraction studies at ESRF.

Figure S4. The electrical resistance of the La sample as a function of temperature during cooling below 100 GPa.
Figure S5. The superconducting transition of the La sample at 53 GPa with applied magnetic field. Inset shows the change of $T_c$ with magnetic field and the solid line in red and blue represents fitting by the LG and WHH equation.

Figure S6. DFT calculated (PAW PBE, VASP) electron density of states of distorted fcc-La at 145 GPa.
Figure S7. Eliashberg functions $\alpha^2 F(\omega)$ of distorted fcc-La with different $\sigma$-broadening (QE) at 50 GPa.

Figure S8. Eliashberg functions $\alpha^2 F(\omega)$ of distorted fcc-La with different $\sigma$-broadening (QE) at 130 GPa.
Figure S9. The pressure distribution in the superconducting electrical resistance measurements at about 132 GPa. Inset shows the corresponding sample positions for the pressure measurements.