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THERMODYNAMIC PROPERTIES OF PRESSURIZED LaH_{10} SUPERCONDUCTOR DUE TO COLLECTIVE EXCITATION OFCOOPER PAIRS

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Abstract

There are two categories of superconductors; s-wave superconductors that are isotropic and d-wave superconductors that are anisotropic. The microscopic theory of superconductivity by Bardeen, Schrieffer and Cooper (BCS theory) explains s-wave pairing of charges under ambient pressure but it fails to explain charge pairing under high pressure. Studies have shown that superconductivity in hydrides is due to electron-phonon mediation. Models have been developed to explain the pressure effect on Tc but so far no unified model has been agreed upon to explain HTSC under pressure using the Bogoliubov-Valatin Transformation (BVT) formalism. The developed theory was used in this work to give more understanding of the superconducting process under pressure and carry on a comparison with other researchers. The systems energy, specific heats, entropy and Sommerfield coefficient were determined. The specific heat capacity for LaH_{10} is 0.0315meV/K at Tc=237.9 and 210GPa. The systems have maximum Cv =4.68meV/K at T=6K. LaH_{10} shows Sommerfield coefficient of 0.00235meV/K^2 at the same stated pressure values. The value of entropy for the hydride at Tc is 0.13meV/K^2 for LaH_{10}. The highest entropy for LaH_{10} is 0.451meV/K^2 occurs at 1000K.

Keywords: Hydrides**,** Pressure, Energy gap**,** Phonon, Collective excitation, BVT

Introduction

Room temperature superconductivity has been achieved under immense pressure. BCS theory accounts for excitation of Cooper pairs at ambient pressure. , Bardeen, *et al*, (1957) developed the microscopic theory of superconductivity based on electron pairing under phonon mediation. The theory explained all the accumulated experimental data on superconductivity. Bogoliubov (1958) showed that the BCS theory could be derived from electronic Hamiltonian. Similar results as those of the BCS theory are obtained when the Bogoliubov-Valatin Transformation (BVT) is used. Despite all the achievements, the BCS theory fails to explain the phenomenon at T_c>30K such as hydride superconductors.

Ashcroft (2004) suggested that compounds with a high hydrogen content might be, in effect, chemically recompressed metallic hydrogen. In a recent development, superconductivity, highest T_c in carbonaceous Sulphur hydride under a pressure of about 2.6million atmospheres (267Gpa) has been achieved (Dias and Salamat, 2021) They used Diamond Anvil Cell (DAC) to achieve this pressure and the material super conducted at 288K (15℃). This development is the most significant breakthrough since the discovery of the high-Tc cuprates (Bednorz and Mueller, 1986). There is every reason to anticipate even higher values of Tc for other hydrides, which means that achieving superconductivity at room temperature now appears perfectly realistic. Determination of the structure of the new hydrides with synchrotron X-ray diffraction is overwhelming evidence of the conventional s-wave superconductivity in the hydrides at high pressures (Drozdov *et al*, 2015). Lanthanum superhydride has recently showed s-wave superconductivity under high pressure. (Sun *et al*, 2021). Purans *et al* (2021) motivated by the discovery of superconductivity above 250 K at high pressure in LaH_{10} and the prediction of overcoming the room temperature threshold for superconductivity in YH_{10} and the urge for a better understanding of hydrogen interaction mechanisms used locally sensitive X-ray absorption fine structure spectroscopy (XAFS) to get insight into the nature of phase transitions and the rearrangements of local electronic and crystal structure in YH_3 under pressure up to 180GPa. They provided evidence of strong effect of hydrogen on the density of yttrium states that increases

with pressure and XAFS data showed evidence a strong anharmonicity.

Therefore understanding the thermodynamic properties and pairing mechanisms of hydride compounds under pressure can help find a metastable form of hydrides for commercialization of the uses**.** A theory to explain the charge pairing mechanisms that pressure brings about is the limiting factor. There has been consistency between theoretical predictions and calculations and most importantly, the general theory of conventional superconductivity on superconductivity in hydrides and the experimental findings. However, some inconsistencies exist between the predictions and experiments, most prominent of all being the experimental electron–phonon coupling under high pressure. Exploring the thermodynamic properties of hydrides under pressure has been done in this research.

Methodology

In this research, the effective Hamiltonian of the system was diagonalized using the BVT (Bogoliubov-Valatin Transformation) formalism and used to further obtain energy of sulphur hydride (H_3S) and other thermodynamic properties. The general Hamiltonian of an electron-electron interaction is given by;

$$H = H_o + H_1$$

Which can written as:

$$H = \sum_k \mathfrak{E}_k n_k + \sum_{k'} \mathfrak{E}_{k'} n_{k'} - \sum_{kk'} V_{kk'} n_k n_{k'} \qquad (1.0)$$

H_o is the unperturbed Hamiltonian system and is sometimes called Bloch energy while H_1 is the interaction. In terms of electron creation and annihilation operators, equation it can be written as

$$H = \sum_k \mathfrak{E}_k c^\dagger_{k\uparrow} c_{-k\downarrow} + \sum_{k'} \mathfrak{E}_{k'} c^\dagger_{k'\uparrow} c_{-k'\downarrow} - \sum_{kk'} V_{kk'} c^\dagger_{k'\uparrow} c^\dagger_{k\uparrow} c_{-k\downarrow} c_{-k'\downarrow} \qquad (1.1)$$

Where $n_k = c^\dagger_{k\sigma} c_{k\sigma}$ Is the electron number operator.

We now introduce quasiparticle creation and annihilation operators known as Bogoliubons where creation and annihilation operators are $\gamma^\dagger$ and γ respectively.

The creation and annihilation operators of electrons and quasiparticles are related as;

$$c^\dagger_{k\uparrow} = u_k \gamma^\dagger_{k\uparrow} + v_k \gamma_{-k\downarrow}$$

$$c_{k\uparrow} = u_k \gamma_{k\uparrow} + v_k \gamma^\dagger_{-k\downarrow} \qquad (1.2)$$

$$c^\dagger_{-k\downarrow} = u_k \gamma^\dagger_{-k\downarrow} - v_k \gamma_{k\uparrow}$$

$$c_{-k\downarrow} = u_k \gamma_{-k\downarrow} - v_k \gamma^\dagger_{k\uparrow}$$

Where the number operators are;

$$\gamma^\dagger_{k\uparrow} \gamma_{-k\downarrow} = m_k$$

$$\gamma_{-k\downarrow} \gamma^\dagger_{k\uparrow} = (1 - m_k) \qquad (1.3)$$

We now substitute (1.2) and (1.3) in (1.1);

$$H = \left\{ \sum_k \mathfrak{E}_k (u_k \gamma^\dagger_{k\uparrow} + v_k \gamma_{-k\downarrow})(u_k \gamma_{-k\downarrow} - v_k \gamma^\dagger_{k\uparrow}) + \sum_{k'} \mathfrak{E}_{k'} (u_{k'} \gamma^\dagger_{k'\uparrow} + v_{k'} \gamma_{-k'\downarrow})(u_{k'} \gamma_{-k'\downarrow} - v_{k'} \gamma^\dagger_{k'\uparrow}) \right.$$
$$\left. - \sum_{kk'} V_{kk'} \begin{pmatrix} (u_{k'} \gamma^\dagger_{k'\uparrow} + v_{k'} \gamma_{-k'\downarrow})(u_k \gamma^\dagger_{k\uparrow} + v_k \gamma_{-k\downarrow}) \\ (u_k \gamma_{-k\downarrow} - v_k \gamma^\dagger_{k\uparrow})(u_{k'} \gamma_{-k'\downarrow} - v_{k'} \gamma^\dagger_{k'\uparrow}) \end{pmatrix} \right\} \qquad (1.4)$$

Since we are interested in the pairing, all terms $\gamma\ \gamma\ \gamma\ \gamma$ will be ignored because they represent single unpaired particles

We determine the values of u_k and $v_k$used for the diagonalization. We equate the off diagonal terms to zero and obtain:

$$\sum_{k}\left(u_k v_k[\gamma_{-k\downarrow}\gamma_{-k\downarrow} - \gamma_{k\uparrow}^{\dagger}\gamma_{k\uparrow}^{\dagger}]\right) - \sum_{kk'} -2u_k v_k^3(-[-\gamma_{-k\downarrow}\gamma_{-k\downarrow} - \gamma_{k\uparrow}^{\dagger}\gamma_{k\uparrow}^{\dagger}]) = 0$$

$$2u_k v_k - 2u_{kv_k^3} = 0$$

$$2u_k v_k = 2u_{kv_k^3}$$

Therefore

$$v_k^2 = 1$$

$$v_k = +1$$

Since we are interested in the pairing, we impose the bosonic probability condition,

$$u_k^2 - v_k^2 = 1$$

Therefore we obtain u_k as,

$$u_k^2 - (+1)^2 = 1$$

$$u_k = \sqrt{2}$$

$$v_k = +1$$

And u_k as,

$$u_k = \sqrt{2}$$

We take positive values since u_k and v_k are real and positive. These values diagonalize the Hamiltonian

$$H_{Diag} = -2\sum_{k}\mathcal{E}_k v_k^2 - \sum_{kk'} V_{Kk'}\, v_k^4 \qquad (1.5)$$

The magnitude of the ground state energy of the system is obtained as,

$$E_o = -2\mathcal{E}_k - V_{Kk'} = -(2\mathcal{E}_k + V_{Kk'})$$

$$[E_o] = 2\mathcal{E}_k + V_{Kk'} \qquad (1.6)$$

For the two hydrides from table 1 and equation 1.6;

$$E_o = 100meV\ or\ 0.10eV\ or\ 1.602\ x\ 10^{-20}J$$

We express the energy of the system at any temperature as a function of temperature by multiplying the ground state energy by the thermal activation factor given by: $exp\left(-\frac{\Delta E}{k_B T}\right)$ where k_B is the Boltzmann's constant and ΔE is the superconducting energy gap. The energy of the quasi particles for superconductivity is very small quantity and is generally 1% of the minimum energy of the system (Ayodo, 2008). Therefore

$$\Delta E = 0.01E_o$$

Therefore at any temperature T, the energy of the system is given by:

$$E(T) = E_o exp\left(-\frac{\Delta E}{k_B T}\right)$$

Which we can write as;

$$E(T) = E_o exp\left(-\frac{2\mathcal{E}_k + V_o}{k_B T}\right) \qquad (1.7)$$

The following hydrides have been studied in this research;

Table 1

Hydride	Formula	$\mathcal{E}_k$ (meV)	V_o (meV)	ω_D (meV)	Δ_o (meV)	Pressure (GPa)	T_c (k)	Reference
Lanthanum hydride	LaH_{10}	24.3	48.6	113	51	210	237.9	S. F. Elatresh et al, 2020

The values of ω_D, Δ_o, pressure and T_c are obtained from experimental results while values of $\mathcal{E}_k$ *and* V_o are mathematically determined using the equation below;

$$V_o = -2\mathcal{E}_k$$

Where $\mathcal{E}_k$ is a single particle energy

$$\omega_D = 2\sqrt{{\mathcal{E}_k}^2 + |\Delta_o|^2} \qquad (Ryo\ Shimano\ et\ al, 2020)$$

From the table, energy of the system, equation 1.9.2 can be given as;

$$E(T) = 97.2exp\left(-\frac{11.27}{T}\right) \qquad (1.8)$$

Where in both cases k_B=0.08628meV/K

Specific heat capacity (c_v)
It's given as the derivative of the energy of the system with respect to the temperature at constant volume. Specific heat capacity of a system is given by.

$$c_v = \frac{dE_T}{dT}$$

When the equation is simplified, it becomes;

$$c_v(T) = \left(\frac{1095}{T^2}\right) exp\left(-\frac{11.27}{T}\right) \qquad (1.9)$$

Sommerfield coefficient (γ) or Electronic specific heat
It's given as the quotient of the specific heat capacity at constant volume and the temperature T. it's given by;

$$\gamma = \frac{c_v}{T}$$

When simplified the above equation becomes;

$$\gamma(T) = \left(\frac{1095}{T^3}\right) exp\left(-\frac{11.27}{T}\right) \qquad (2.0)$$

Entropy (s)
It's the measure of disorder of a system. It's given as the integral of the specific heat capacity at constant volume with respect to temperature. It's given by;

$$S = \int \frac{c_v}{T} dT$$

Which we can simplify to become;

$$S = \left(K_B + \frac{E_o}{K_B T}\right) exp\left(-\frac{E_o}{100 k_B T}\right)$$

Or

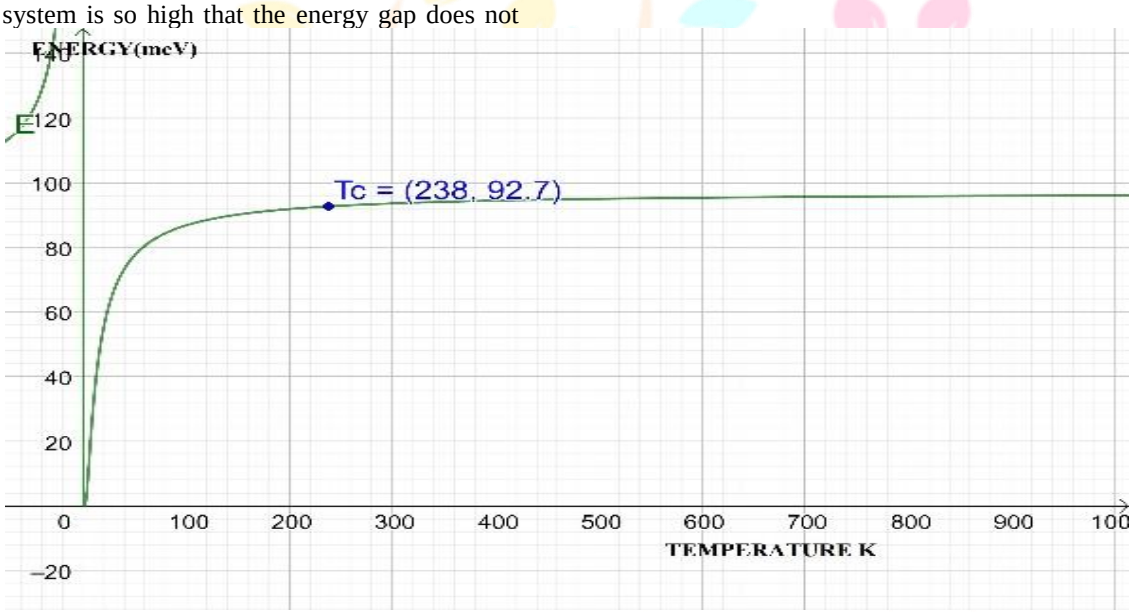
$$S = K_B + \frac{E_o}{K_B T}\left(exp\left(-\frac{E_o}{k_B T}\right)\right)$$

Substituting in the equation;

$$S(T) = 0.08628 + \frac{1127}{T}\left\{exp\left(-\frac{1127}{T}\right)\right\} \qquad (2.1)$$

Results and discussions

The total energy of a system results from the interaction between the particles of the system. The energy due to interaction between the particles increases with increase in the temperature of the system. At a temperature above the Tc the energy of the system is so high that the energy gap does not exist anymore due to the increased agitation of the particles. At this point the material changes from superconducting to normal state.



LaH_{10} (Fg 1): Energy versus Temperature

The electrons are non-interacting at these zero temperatures hence coupling energy is zero (Figure 1). The lower states are filled up and above superconducting gap states are empty. As temperature increases, an increase in energy is noted and approaches plateau. Kibe (2015) and Odhiambo (2016) observed sigmoid curves when at different time related energy of a system to temperature. Under ambient pressure (1atmos), LaH_{10} does not superconduct. As pressure increases the Cooper pairs are collectively excited to cause an exponential increase in energy. There is an increase in density of states at the Fermi level. LaH_{10} with a Tc=237.9K at 210GPa (Elatresh *et al*, 2020) has been observed to be $92.7meV\ or\ 0.0927eV$.

Specific Heat Capacity at Constant Volume.

The specific heat capacity of LaH_{10} is 0.0315meV/Mol.K at Tc=237.9 and 210GPa. The systems have maximum of The systems has maximum Cv =4.68meV/Mol.K at T=6K for LaH_{10} and at this point the system is very unstable and the superfluid state is just like the 2nd order phase transition turning from the ordinary state to a superconducting state. At the Tc more quasiparticles are easily excited because the energy gap is closed. Specific heat becomes very high than normal state (Figure 2). Beyond the Tc, an increase in temperature leads to a decrease in specific heat capacity because more cooper pairs break than form as energy gap is closed.

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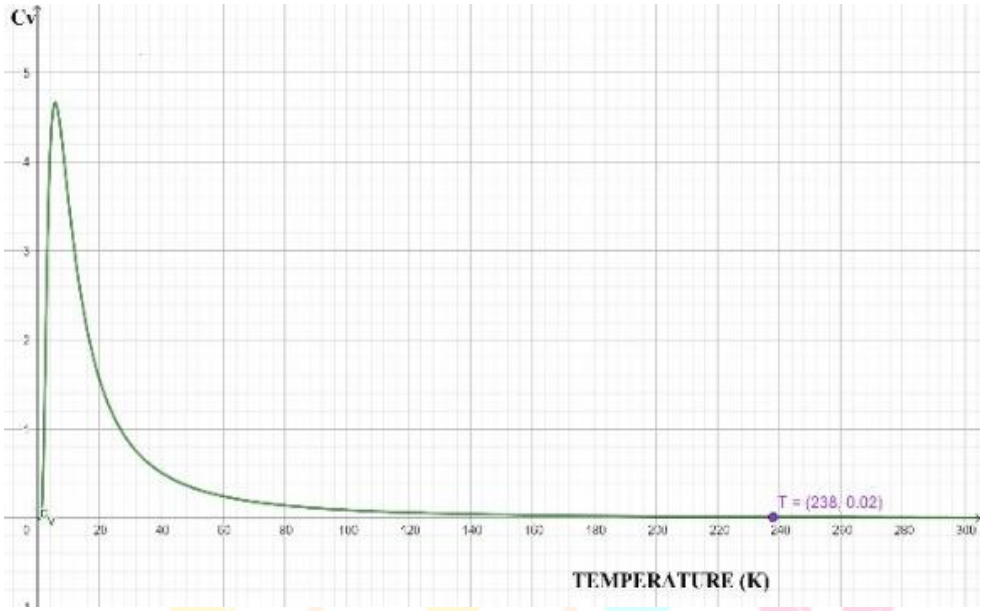
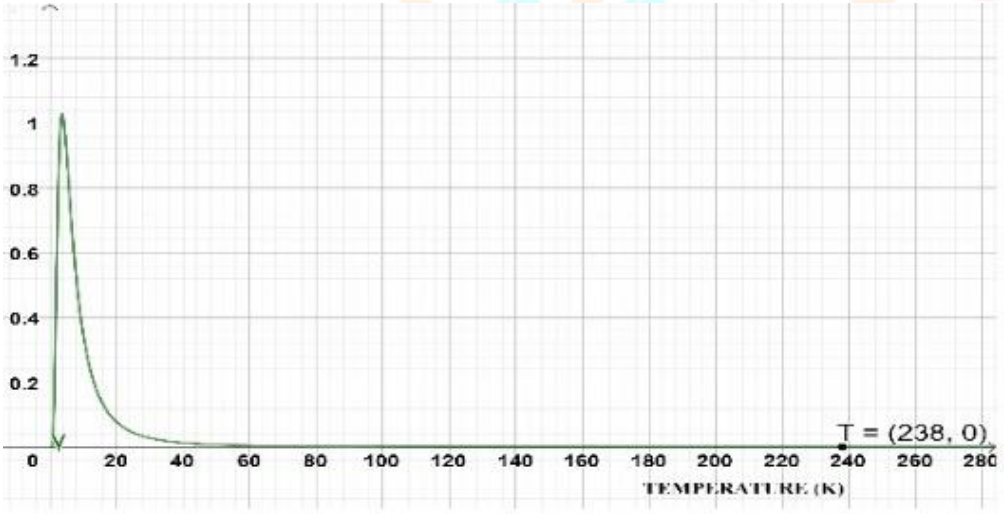


LaH_{10} (Fig 2): Specific heat capacity versus Temperature

Electronic specific heat or Sommerfield coefficient.



LaH_{10} (Fig 3)): Sommerfield coefficient versus Temperature

Based on the graphs, at $T = 0K$, Sommerfield coefficient is zero. This suggests that the electrons in the system do not correlate to form Cooper pairs that contributes to superconducting condensate but remain normal (De Visser 2019). At low temperature, the systems have a wider energy gap. Due to the lower internal energy, the system is exponentially suppressed and absorbs negligible energy hence the low Sommerfield coefficient below Tc. Initially there is a sharp increase in Sommerfield coefficient up to a point where T=3.66k (Figure 3). The higher pressure makes the multi-gap to overlap, the quasiparticles are easily and collectively excited and the system attains higher Sommerfield coefficient before Tc. Beyond this temperature, the Sommerfield coefficient drops forming a Gaussian shaped curve skewed to the left and hump-like shoulder observed by (Cronner *et al*, 1960). The gapless material gains energy slowly, the Sommerfield coefficient decreases. From the figures 3, LaH_{10} shows a Sommerfield coefficient of 0.00235meV/K^2 for LaH_{10} at $Tc = 237.9K$.

$$\frac{2\Delta}{K_B T_c} = \frac{51\ x\ 2}{0.08628\ x\ 237.9} = 4.97$$

Durajski *et al* (2021) found $\frac{2\Delta}{K_B T_c} = 5.04$ for LaH_{10} with Tc=234K under pressure of 250GPa. These values proved phonon mediated superconductivity in LaH_{10}. This value proved phonon mediated superconductivity in sulphur hydride (H_3S)

Specific heat jump from this study is found to be;

$$\frac{\Delta C(T)}{\gamma T_c} = \frac{51x0.0315}{0.00235x237.9} = 2.874$$

Errea *et al* (2020) found the $\frac{\Delta C(T)}{\gamma T_c} = 2.97$ for LaH_{10} at 214GPa at same$Tc = 237.9K$.

Nicol and Carbotte (2015) while studying compressed sulfur hydride found $\frac{\Delta C(T)}{\gamma T_c} = 2.53$ which is closer to those obtained from this study. The thermodynamic universal BCS ratios are near those for Pb and Nb_3Sn (Conventional superconductors.). These ratios are in agreement with those obtained by Elatresh *et al*, 2020 and these values show that the two hydrides are s-wave conventional superconductors. They are within the range of the classic ratios set by the BCS theory. Therefore the pairing mechanisms can be explained using BCS formalism.

Entropy

The curve indicates that entropy is nearly zero (0.0882) when temperature T=0K (Figure 4). The system with a large energy gap absorbs low energy. The system energy absorption is suppressed; the system is more ordered hence low entropy. At Tc, the systems have lowest entropy but rises exponentially as temperature rises. The system becomes more disordered as more energy is absorbed beyond Tc. Higher pressure speeds up closing of the gaps. It plateaus as temperature rises because pressure cannot sustain the increased disorder anymore. The value of entropy for the hydride is 0.13meV/K^2 at Tc. The highest entropy for H_3S is 0.450meV/K^2 and occurs at 1000K. This suggests LaH_{10} could be more ordered at a higher temperature than H_3S (S=0.15meV/K^2) and could be better candidate for room temperature superconductor. The free energy of the normal state is much higher than the free energy of the superconducting state at temperatures below the critical temperature value and this therefore makes superconducting phase to appear belowTc. A decrease in internal energy of the system is observed with the diminishing or reduction of the state of the disorder and so this implies that increase in temperature provides more kinetic energy causing increase in thermal entropy (Mumali *et al*, 2016)

From figure 4, state of disorder in normal state is generally higher than that of superconducting state at all temperatures below the Tc $237.9K$ for LaH_{10} showing that superconducting state is more ordered. These results agree with those of other theoretical studies involving s-wave pairing mechanisms like in heavy fermions (Kibe *et al*, 2015 and Waswa *et al*, 2017).

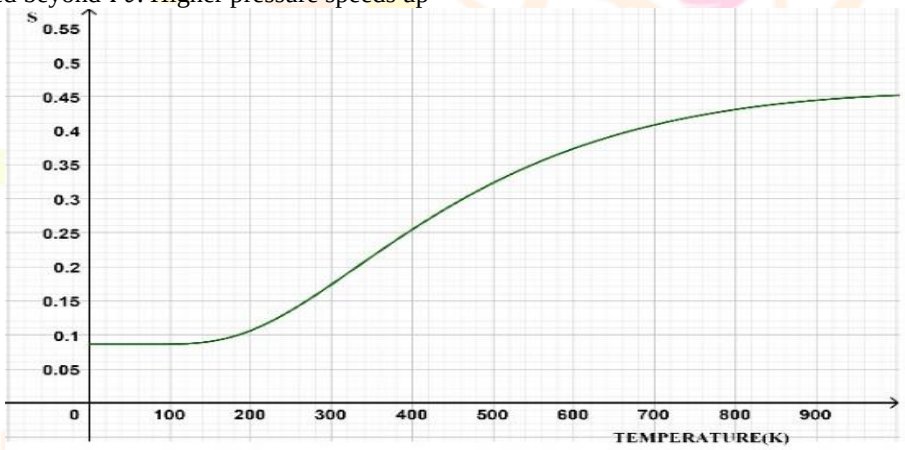


Figure 4: Entropy versus Temperature for LaH_{10}

Conclusions

Energy of LaH_{10} under pressure is due to collective excitation of Cooper pairs that results to exponential rise with temperature; as pressure reduces the cell volume, the lattice is distorted or transformed; there is a small energy difference between the orbitals which enhances electron-phonon coupling strength due to collective excitation of particles. Under pressure, superconductivity is a bulky phenomenon shown by hump-like feature where the dependence of specific heat capacity is 0.0315meV/K at Tc=237.9 and 210GPa. This value is way much higher than for other conventional superconductors under normal pressure. Therefore pressure raises specific heat capacity and Sommerfield coefficient of LaH_{10}. Entropy is lowest at Tc to mean superconducting state under pressure is still ordered like other superconductors. LaH_{10} has lower entropy than hence more ordered and a better candidate for RTS. We further found out BCS ratios and Specific heat jump are in close proximity to other related researches ascertaining that LaH_{10} is a conventional s-wave superconductor. Pressure increase on hydrides causes collective excitation of Cooper pairs that raises Tc.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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