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# Study of the Isotope Effects of Novel Superconducting $LaH_{10}$-$LaD_{10}$ and $H_3S$-$D_3S$ Systems

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## Abstract

This paper is directed to study the isotope effects of some superconducting materials that have a strong coupling coefficient $\lambda > 1.5$, and focuses on new superconducting materials whose critical temperature is close to room temperature, specifically $LaH_{10}$-$LaD_{10}$ and $H_3S$-$D_3S$ systems. The Eliashberg-McMillan (EM) model and the recent Gor'kov-Kresin (GK) model for evaluating the isotope effects coefficient α were examined for these systems. The predicted values of α as a function of pressure, as compared to experimental values led to inference that these two models, despite their importance and simplicity, cannot be considered complete. These models can be used to calculate isotope effect of most superconducting materials with strong coupling coefficients but with critical reliability. The significance of studying the isotope effect lies in the possibility of identifying the interatomic forces that control the properties of superconducting materials such as electrons-mediated phonons and Coulomb interactions.

## Keywords

Isotope Effects, Superconductivity, Strong Coupling Coefficient, High Pressure

## 1. Introduction

Superconductivity was first discovered in 1911 [1], in mercury cooled below 4 K. The temperature below which a material becomes superconducting is called the critical temperature. It was quickly appreciated that a state exhibiting zero electrical resistance could be tremendously useful, if materials that have critical temperatures much higher than 4 K could be found. Over the past century, as more superconductors have been discovered, the record for the highest critical temperature achieved has progressed towards the ultimate goal of room temperature.

Since the discovery of Bednorz and Müller in 1986 [2] on oxide superconductors with critical temperature $T_c$ approximately equal to 35 K, there are a great number of laboratories all over the world involved in research of superconductors with high $T_c$ values. The discovery of room temperature superconductors has been long-standing dream of many scientists, because the technological and practical applications of such discovery should be tremendous. Now experimental data confirms superconductivity at higher temperature than even before. Drozdov *et al.* reached superconductivity at 250 K in lanthanum hydride $LaH_{10}$ at pressure of about 170 GPa [3]. This is the highest critical temperature that has been confirmed so far in a superconducting material. This is an encouraging step towards the goal of achieving room temperature superconductivity. In fact, the metallic atomic-like hydrogen phase was proposed by Wigner and Huntington to occur under high pressure conditions [4]. The pressure of metallization, estimated by these authors was about 20 GPa, ultimately proved to be incorrect, and it was realized later found to be much higher than this value [5]. The experimental [6] and theoretical [7] works in high temperature superconductors have stimulated another important hydrogen compounds are the $H_3S$-$D_3S$ system. The superconductivity occurs via formation of a structure of stoichiometry $H_3S$ with S atoms arranged on a body-centered-cubic (bcc) lattice. Two metallic structures with R3m and Im-3m symmetries are reconstructive above 111 and 180 GPa respectively [7]. The knowledge of the microscopic mechanisms of high $T_c$-superconductors should be a theoretical guide. One of these microscopic mechanisms is the isotope effect on the superconductivity transition temperature. Such an effect can be considered as the hallmark of phonon-induced superconductivity in conventional superconductors. Inspired by anomalous isotope effect in iron-based superconductors [8], it is helpful to investigate the competitions between electron-electron and electron-phonon interaction. While phonons are central in explaining the mechanism of conventional superconductors, their role should be taken into accounts for explaining high $T_c$ superconductors, as well. Different theories have been proposed to explain the mechanisms of superconductivity. The macroscopic theories like Meissner effect [9], London equations [10], Ginsberg-Landau theory [11] do fairly explain some properties of superconductors but were fairly inadequate to explain the underlying mechanism of superconductivity. The first microscopic theory of superconductivity was put forth by Bardeen, Cooper and Schrieffer (BCS) in 1957 [12]. This theory explained a second-order phase transition at the critical temperature $T_c$, an electronic specific heat, the Meissner Ochsenfeld effect, and the dependence of $T_c$ on the isotope substitution (change of $T_c$ upon deuterium for hydrogen substitution). After the BCS mechanism was established, it became clear that the isotope coefficient α was determined from the equation $T = Am^{-\alpha}$, where $m$ is the isotope mass and $A$ is a constant. This theory is based on the fact that the interaction between electrons resulting from virtual exchange of phonons is attractive. The superconducting state is formed when this attractive interaction between electrons resulting from virtual exchange of phonons is attractive. The superconducting state is formed when this attractive interaction dominates the repulsive screened Coulomb interaction. The present research motivates further experiments in the search of high-temperature superconductivity at ambient pressure. Recently, the photochemically synthesized C-S-H systems becomes superconductor with its highest critical temperature being $T_c = 287 \pm 1.2$ K at 267 ± 10 GPa [13]. The origin of superconductivity is not purely electron mediated phonon but also electron-electron interactions. It seems that both couplings play an important role in the mechanisms of superconductivity. In unconventional superconductors, isotope effect can be considered as a probe for both couplings [14]. The BCS theory can be considered as a first successful microscopic theory, which explains most of the physical properties observed in conventional superconductors first and is the theoretical ground for interpretations of the properties of superconductors, particularly the isotope effect. The isotope effect coefficient predicted by BCS theory has the value $\alpha \approx 0.5$. Although this value is valid for some metals like Hg, Tl and others, but the spans between less and greater than this value is found in so many superconducting materials indicating that the BCS theory did not completely succeed in explaining isotope effect in superconductors, particularly for the materials having strong coupling coefficient ($\lambda > 1.5$). This paper focusses on the mathematical models which take into accounts the strong-coupling superconductors and the isotope effect exponent has no longer universal value.

## 2. Isotope Effect Coefficient

### Isotope Effect in BCS Theory

According to the weak coupling BCS theory, the relation between transition temperature $T_c$, typical phonon frequency $\omega$ and interaction strength $N(E_f)V$ as:

$$k_B T_c = 1.14\hbar\omega \exp\left(-\frac{1}{VN\left(E_f\right)}\right) \tag{1}$$

$V$ is the pairing potential arising from electron-phonon interaction, $N(E_f)$ is the electron density of states at Fermi surface and $k_B$ is the Boltzmann constant. The transition temperature is a strong function of the electron concentration, and its proportional to $\hbar\omega$, which is consistent with the isotope shift. It should be possible to make estimates of the change of $T_c$ with pressure, alloying, etc., from (1). The following approximation can be used to calculate $\omega$ which is proportional to $M^{1/2}$ [15], where $M$ is the ionic mass. Within the framework of the electron-phonon mechanism, the $T_c$ can be described by the following relation:

$$T_c = AM^{-\alpha} \tag{2}$$

where $A$ is a constant, $M$ is the mass of the element substituted by its isotope and

$\alpha$ is the isotope effect coefficient, which is defined as:

$$\alpha = -\frac{\partial \ln T_c}{\partial \ln M} \approx -\frac{M}{T_c}\frac{\Delta T_c}{\Delta M} \tag{3}$$

where $\Delta T_c$ is the shift of the critical temperature substitution of isotopic mass and $\Delta M$ is the mass difference between two isotopes. In the standard BCS theory, $T_c$ is inversely proportional to the square root of the masses of the isotope elements, hence the isotope effect coefficient $\alpha = 0.5$ which has been considered in good agreement with important isotope effect in many metals as mentioned before. In numerical simulation of the Equation (3), Huang [16] argued that the following equation is more accurate than that in (3):

$$\alpha_i = \frac{\ln T_c(i+1) - \ln T_c(i)}{\ln M_{i+1} - \ln M_i} \approx \frac{M_i}{T_c(i)}\frac{T_c(i+1) - T_c(i)}{M_{i+1} - M_i} \tag{4}$$

In the formula (4), two set of adjacent data ($T_c(i)$, $M(i)$), $T_c(i+1)$ and $M_{i+1}$ should be used for an accurate calculation of $\alpha_i$.

Vora [17] has deduced from the best fit to the data of about twenty-five materials, the following equation for $T_c$:

$$T_c = \left(\frac{\langle\omega\rangle}{10.71}\right)(\lambda - 0.3362) \tag{5}$$

where $\langle\omega\rangle$ is the average phonon frequency and $\lambda$ is the electron-phonon coupling strength. As the electron-phonon coupling strength is unaffected by the isotope substitution for harmonic phonon dispersion, and by using Equation (5), the isotope-effect coefficient can be written in terms of the phonon frequency for the $\text{LaH}_{10}$-$\text{LaD}_{10}$ system as example:

$$\alpha = -\frac{M}{\Delta M}\frac{\langle\omega\rangle_{\text{LaD}_{10}} - \langle\omega\rangle_{\text{LaH}_{10}}}{\langle\omega\rangle_{\text{LaH}_{10}}} \tag{6}$$

The isotope effect evaluation using Equation (6) requires only the knowledge of the phonon frequencies which can be measured by the infrared or Raman spectra or predicted by the first principal density functional theory DFT. This equation indicates that the isotope effect causes a phonons frequency shift (energy shift) which differs than the original BCS theory which is given in terms of the superconducting temperature shift. Both of these shifts are due to the internal heavy atom effects.

It is noticeable that the D-derived optical phonon modes shifts towards lower frequencies, relative to the corresponding H-derived modes. For instances, at 250, 300 and 350 GPa, the lowest optical modes Γ point shift from 109.44, 118.36 and 123.12 meV in $\text{LaH}_{10}$ to 77.52, 83.44 and 86.93 in $\text{LaD}_{10}$, respectively [18]. According to these phonon frequencies, Equation (6) gives $\alpha$-values: 0.293, 0.297 and 0.295 for the pressures of 250, 300 and 350 GPa respectively. The average value of $\alpha$ is 0.295, while the experimental value from the critical temperature shift is 0.35 [3]. The error percent between the two methods is 15.7%. This discrepancy is due to the neglect of the acoustic phonon frequencies contribution, which are much less than the optical phonon frequencies. Equation (6) only calls the optical phonon frequencies as a proxy for critical temperatures.

**Isotope effect in strong coupling constant**

The BCS theory did not completely succeed in explaining isotope effect in superconductors, but it paved the way for a deeper understanding of electron-phonon coupling. Eliashberg model [19] assumed strong coupling between electrons and phonons and calculated the spectrum and the damping excitations. All superconductors are characterized as having weak ($\lambda_{opt} \ll 1$), intermediate ($\lambda_{opt} \approx 1$), and strong coupling ($\lambda_{opt} \gg 1$) [20]. McMillan-Dynes [21] [22] performed advanced analysis of the problem by utilizing the Eliashberg theory and proposed the critical temperature equation:

$$T_c^{\circ} = \frac{\varpi_{opt}}{1.2} \exp\left[ -\frac{1.04\left(1+\lambda_{opt}\right)}{\lambda_{opt} - \mu^{*}\left(1+0.62\lambda_{opt}\right)} \right] \tag{7}$$

Here $\mu^{*}$ is the effective Coulomb repulsion, which is assumed to be within a range of $\mu^{*}$ = 0.1 – 0.2. Equation (7) is highly accurate for a wide range of coupling strength $\lambda_{opt} \leq 1.5$, and it is widely used to evaluate the $T_c$ in the phonon mediated superconductors. The isotope effect exponent α is determined from Eliashberg-McMillan's (EM) model:

$$\alpha = \frac{1}{2}\left[ 1 - \left( \mu^{*} \ln \frac{\hbar\omega}{1.45 k_B T_c} \right)^2 \left( \frac{1+0.62\lambda_{opt}}{1+\lambda_{opt}} \right) \right] \tag{8}$$

For $\lambda_{opt}$ >1.5, Allen and Dynes [23] proposed a correction factors should be included in the Equation (7), so that it becomes:

$$T_c = T_c^{\circ} f_1 f_2 \tag{9}$$

where $f_1$ is the "strong-coupling correction", and $f_2$ is the "shape correction". $f_1$ must scale as $\lambda_{opt}^{1/2}$. For the $f_2$ calculation, the empirical relation deduced from **Table 1** in ref. 23 is used [20]:

$$f_2 = 1 + \left(0.0241 - 0.0735\mu^{*}\right)\lambda_{opt}^2 \tag{10}$$

This parabolic function is deduced by the fit of tabulated $f_2$ values for all materials reported by Allen and Dynes [23].

A modified form of the isotope effect coefficient α is developed by Gor'kov and Kresin (GK) [24] and shown to provide the relative contributions of optical and acoustic branches of infrared or Raman spectrum. The GK model is based on a hypothesis that the isotope effect originates from high frequency phonons and differs in the two phases. The value of the isotope coefficient is written as [24]:

$$\alpha \approx \frac{1}{2}\left[ 1 - \frac{\lambda_{ac}}{\lambda_{opt}} \frac{\rho^2}{\left(\rho^2+1\right)^2} \right] \tag{11}$$

Here $\rho = \dfrac{\varpi_{ac}}{\pi T_c^{\circ}}$.

$\varpi_{ac}$ is the average frequency of the acoustic phonons and $\lambda_{ac}$ is the acoustic coupling constant. We should remark that the EM model and the GK model indicate that the isotope effect exponent has no longer a universal value ($\alpha = 0.5$) as predicted by the BCS theory, but it may take values less or greater than 0.5. In this paper, the two models were examined to evaluate their validity in calculating $\alpha$-values. For this purpose, new groups of materials with superconductivity close to room temperature and different high pressures were selected. This study paves the way to know the interatomic forces that control the superconductivity at room temperature which is the main goal of current research to achieve superconductivity at room temperature.

## 3. Results and Discussion

### Isotope Effects in the fcc ($LaH_{10}$-$LaD_{10}$) System

First-principles calculations based on density functional theory suggested that a new family of superconducting hydrides that possess clathrate-like structure in which the host atom (lanthanum) is at the center of a cage formed by hydrogen atoms. This nearly spherical structure can be considered as standard for the study of the electrons-electrons and the electrons-phonons interactions and then the isotope effects. Table 1 shows the data used for calculating the isotope effects in superconducting $LaH_{10}$-$LaD_{10}$ system under high compression. The Coulomb pseudopotential $\mu^* = 0.2$ was assumed. The isotope coefficient $\alpha$ was determined by using the EM-model, Equation (8) and the GK-model, Equation (11). Both models mainly depend on the values of the critical temperatures, but new variables, acoustic phonon frequency and the acoustic coupling coefficient were added in the GK model. The predicted critical temperatures $T_c$ were between 150 and 266 K. The reported superconductivity critical temperature of around 250 K at about 170 GPa [3]. When calculating the isotope effect from the Equation (11), the phonon contributions were taken into accounts using the optical and the acoustic branches which they have different frequencies and coupling constants. On this basis, it was introduced the average frequencies $\tilde{\omega}_{opt}$ and $\tilde{\omega}_{ac}$, also the coupling constants $\lambda_{opt}$ and $\lambda_{ac}$. This distinction allows comparison of the

**Table 1.** Calculated values of electron phonon coupling $\lambda$, average phonon frequency $\varpi$, $T_c$ and the isotope effect coefficient $\alpha$ for superconducting lanthanum hydride under high compression. The Coulomb pseudopotential $\mu^* = 0.2$ was assumed.

| Pressure (GPa) | 150 | 250 | 300 | 350 | |
|---|---|---|---|---|---|
| $\lambda_{opt}$ | 2.05 (this work) | 2.38 | 1.82 | 1.54 | Ref. [18] |
| $\varpi$ (K) | 1160 (this work) | 1270 | 1373 | 1428 | Ref. [18] |
| $T_c^{\circ}$ (K) | 133 | 164 | 144 | 119 | Equation (7) |
| $T_c$(K) | 250 | 266 | 195 | 151 | Equation (9) |
| $\alpha$ | 0.442 | 0.480 | 0.460 | 0.447 | Equation (8) |
| α | 0.35 | 0.36 | 0.35 | 0.35 | Equation (11) |

relative contributions of the optical and the acoustic phonons. Depending on [24], $\lambda_{opt} \approx 3\lambda_{ac}$, and $\tilde{\omega}_{opt} \approx 4\tilde{\omega}_{ac}$ were estimated. This estimate was used in a single case for sulfur hydride system [24] and was generalized in this study to include, a similar system in properties, the lanthanum hydride system as it is a convincing estimate due to its reliance on practical results for optical phonon frequencies and acoustic phonon frequencies.

**Figure 1** demonstrates the variations of the superconducting parameters $T_c$, $\tilde{\omega}_{opt}$, $\lambda_{opt}$, and $\alpha$ with the pressure in gigapascals. The first three parameters mainly represent the superconducting materials properties of interest for predicting the isotope effect coefficient $\alpha$. The pressure range extends from 150 to 350 GPa. **Figure 1(a)** shows that the critical temperature $T_c$ is decreasing with the pressure in the range from 200 to 350 GPa, in agreement with [25] [26]. The correction factors of the critical temperature $f_1$ and $f_2$ were taken into accounts because the values of the electrons-phonons coupling constants are greater

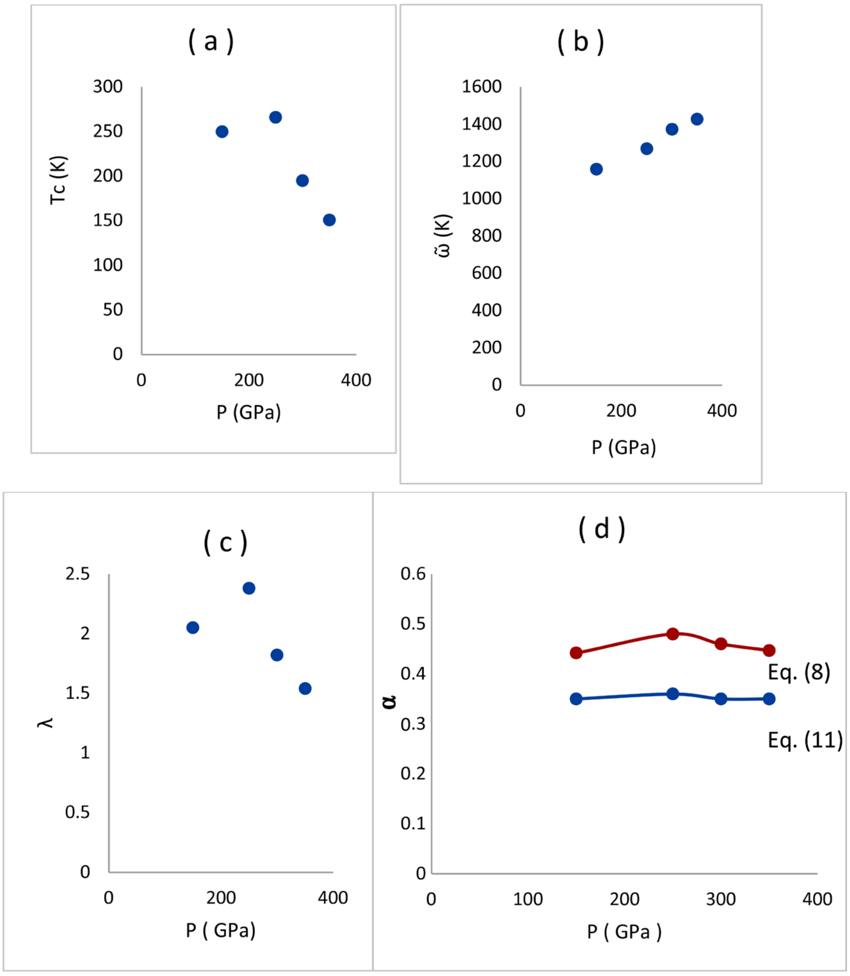

**Figure 1.** The calculated superconducting parameters at different pressures for the LaH$_{10}$-LaD$_{10}$ system (a) calculated $T_c$ of LaH$_{10}$ as a function of pressure; (b) the average values of phonon frequencies as a function of pressure; (c) the coupling coefficients as a function of pressure; (d) calculated isotope effect coefficients $\alpha$ as a function of pressure for the EM model, Equation (8), and GK model, Equation (11).

than 1.5. The predicted $T_c$ values are around 266 K at the pressure of 250 GPa and 151 K at the pressure of 350 GPa. The recent reported value for $LaH_{10}$ was 280 K at pressure of 190 GPa [27]. It was found that $LaH_{10}$, Fm3m structure is stabilized above 170 GPa. This finding supports theoretical estimates regarding the dynamic stability of $LaH_{10}$ above 200 GPa [18]. **Figure 1(b)** shows that the optical phonon frequency increases linearly with the increase of the pressure. This is the contribution of the optical phonon frequencies arise from H-atoms. The acoustical phonon frequency modes with lower frequencies below 45 meV (660 K) arise from La atoms [28]. Interestingly, as pressure increases, the total $\lambda$ decreases monotonously (**Figure 1(c)**), consistent with the decrease of $T_c$ measured by a recent experiment of fcc $LaH_{10}$ [3]. The isotope coefficient $\alpha$ was determined from Equations (8) and (11) which were used for the cases of strong electron-phonons coupling $\lambda > 1.5$. Both equations give approximately a constant value of $\alpha$ as a function of pressure (**Figure 1(d)**). Both models used for calculating $\alpha$ give approximately a constant value as a function of pressure. The average value of $\alpha$ using Equation (8) is 0.457 in excellent agreement with experimental value (0.46), which was measured for $T_c = 249$ K (fcc-$LaH_{10}$) and $T_c =$ 180 K (fcc-$LaD_{10}$) at a pressure of around 150 GPa [3]. On the other hand, the average value of $\alpha$ using Equation (11) is 0.352, with a percent error of 23.4% from the experimental value. The results of $\alpha$-values for $LaH_{10}$-$LaD_{10}$ system shown in **Table 1** indicate that the advantage of the EM model over the GK model. In order to verify more from both models in calculating $\alpha$-values, another system was examined that differs from the first system in structure, but is similar to it in that it operates at high temperatures and under high pressures. The second hydride system that underwent examination of both models is $H_3S$-$D_3S$. **Table 2** and **Table 3** summarize the data required to calculate $\alpha$-values under strong coupling condition. **Table 2** shows the data used for the R3m structure, while **Table 3** demonstrates the data for Im-3m structure. The value of the Coulomb potential $\mu^* = 0.1$ was assumed. For R3m structure, the values of $\alpha$ from Equation (8) are almost constant with the compressed pressure values and equal to 0.49 as shown in **Table 2**. On the other hand, when using the Equation (11) in calculating the values of $\alpha$, it was noticed that the average value of $\alpha$ was equal to

**Table 2.** Calculated values of electron-phonon coupling coefficient $\lambda$, average phonon frequency $\varpi$, $T_c$ and the isotope effect coefficient $\alpha$ at different pressure for R3m structure of sulfur hydride. The Coulomb pseudopotential $\mu^* = 0.1$ was assumed.

| Pressure (GPa) | 110 | 130 | 150 | 160 | |
|---|---|---|---|---|---|
| $\lambda_{opt}$ | 2.08 | 2.07 | 2.57 | 2.48 | Ref. [30] |
| $\varpi$(K) | 981.7 | 1125.1 | 1043.8 | 1124.3 | Ref. [30] |
| $T_c^*$ (K) | 144.7 | 165.7 | 174.5 | 184.5 | Equation (7) |
| $T_c$(K) | 223.45 | 255.84 | 310.49 | 203.5 | Equation (9) |
| $\alpha$ | 0.49 | 0.49 | 0.49 | 0.49 | Equation (8) |
| $\alpha$ | 0.33 | 0.33 | 0.35 | 0.35 | Equation (11) |

**Table 3.** Calculated values of superconducting parameters at different pressure. Electrons-phonons coupling coefficient $\lambda$, average phonons frEquationuency $\varpi$, $T_c$ and the isotope effect coefficient α for Im-3m structure of sulfur hydride. The Coulomb pseudopotential $\mu^* = 0.1$ was assumed.

| Pressure (GPa) | 180 | 185 | 200 | 250 | 300 | |
|---|---|---|---|---|---|---|
| $\lambda_{opt}$ | 2.73 | 2.48 | 2.19 | 1.70 | 1.54 | Ref. [30] |
| $\varpi$ (K) | 1079.4 | 1175.5 | 1334.6 | 1566.8 | 1680.2 | Ref. [30] |
| $T_c^{\circ}$ (K) | 186.0 | 192.6 | 203.5 | 199.6 | 195.9 | Equation (7) |
| $T_c$ (K) | 345.21 | 334.92 | 325.05 | 272.56 | 252.28 | Equation (9) |
| $\alpha$ | 0.49 | 0.49 | 0.49 | 0.49 | 0.49 | Equation (8) |
| $\alpha$ | 0.43 | 0.43 | 0.42 | 0.40 | 0.40 | Equation (11) |

0.34, in excellent agreement with measured value of $\alpha = 0.35$ [29] and with the theoretical prediction [24].

For Im-3m structure, the $\alpha$-values are constant with a pressure change and are 0.49 when using Equation (8) (**Table 3**), while it was found that the average value of $\alpha$ when using Equation (11) is 0.41. The error percent for the experimental value is 17%. In $H_3S$-$D_3S$ system, it is noticed that the GK model achieves $\alpha$-values better than the EM model.

## 4. Conclusion

The isotope effect on the superconductivity transition temperature $T_c$ is one of the hallmarks of phonon-induced superconductivity in conventional superconductors. The dependence of the superconductivity transition temperature on the isotope mass provides an important probe of the pairing mechanism. Precise values of $\alpha$ and $T_c$, together with other parameters, allow a stringent test of superconductivity pairing mechanism, in particular electron-phonon models. The results of the calculations of the $\alpha$-values discussed in this paper indicate that the EM model achieved accurate $\alpha$ values for the ($LaH_{10}$-$LaD_{10}$) system in comparison with the experimental measurements, while the GK model gave better results for $\alpha$ values for ($H_3S$-$D_3S$) system compared to the experimental results. Therefore, it can be concluded that both models are not fit to be a reliable inclusive model, which can apply to most superconducting materials having a strong coupling constant. Accordingly, there is now a need to develop a versatile model that is suitable for treating the isotope effect of most superconducting materials with a strong coupling coefficient.

## Conflicts of Interest

The author declares no conflicts of interest regarding the publication of this paper.

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