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Isotope Effects of Novel Superconducting YH_6 – YD_6 and Th-H – Th-D Systems

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Abstract. This paper is devoted to test the validity of the Eliashberg – McMillan (EM) model and the recent Gor'kov – Kresin (GK) model developed to solve the paradox in the original BCS theory of superconductivity. These models were designed for predicting the isotope effects of some superconducting materials that have a strong coupling coefficient $\lambda > 1.5$. The study is focused on new superconducting materials whose critical temperature is close to room temperature, specifically YH_6 – YD_6 and Th-H – Th-D systems. Although the isotopic coefficient α- values of the two models are close to each other, they deviate from the experimental value with an error percent of 20%. These models, despite their importance and simplicity for predicting the isotope effects, can be considered with critical reliability.

Keywords: Isotope effects, Superconductivity, strong coupling coefficient, High pressure.

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1. Introduction

Superconductive technology is one of the most promising approaches to quantum computing because it offers devices with little dissipation [1]. This feature can be invested in the field of satellite communication, where the most important development takes place in the transition towards all- digital receivers as well as transmitters [2]. Among other applications, the nuclear magnetic resonance, the magnetic levitation train, the transport processing of electrical energy (motors, generators, transformers and power lines, and superconducting magnetic energy storage (SMES) systems are already solutions contributing to the nowadays daily life, but more than that, are solutions that will contribute to improve the quality of life of many human beings in the near future [3].

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, of about 20 GPa, ultimately proved to be incorrect, and it was realized latter that the actual metallization pressure around 500 GPa [5]. For a long time, the critical temperature of conventional superconductors has been limited to T_c = 39 K in MgB_2 [6]. The breakthrough and tremendous progress for reaching high temperature superconductivity came with the discovery of critical temperature at 203 K in H_3S at about 150 GPa [7]. This work initiated extensive high-pressure studies of hydrides, which are conventional phonon mediated superconductors. The most illustrious of these researches is the work of Somayazulu et al in which they reached at 260 K in La H_{10} [8]. The most recent studies among others regarding the superconductors of superhydride have focused on yttrium hexahydride YH_6- YD_6 system and thorium hydrogen ThH_9-ThD_9 system. However, Troyan et al [9] have demonstrated the superconductivity transition temperature of Im-3m- YH_6 = 224 K at 166 GPa. Another research group of Kong et al [10] confirmed this transition temperature with maximal Tc of ≈ 220 K at 183 GPa. This compound was found to be stabilized above 110 GPa [11]. As well, high pressure synthesis of two novel high- T_c hydride superconductors, $P6_3/mmc$-ThH_9 and Fm-3m ThH_{10}, with experimental critical temperature T_c of 146 and 159-161 K at pressures 170- 175 GPa, respectively. The fcc- ThH_{10} has stabilization pressure of 85 GPa, making this material unique among all known high T_c metal polyhydrides [12].

A great breakthrough occurred with the discovery of the isotope effect in the superconductivity of mercury [13] which definitely established as has been suggested independently by Fröhlich [14] that the electron – phonon interactions are primarily responsible for superconductivity. The Maxwell results inferred that the transition of a superconductor is a function of the nuclear mass, the lighter the mass, the higher the transition temperature. These pioneering works motivated Bardeen, Cooper and Schriffer (BCS) to develop a comprehensive theory to explain the main parameters of a conventional superconductivity [15]. The isotope effect coefficient α , according to BCS theory 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 the interaction between electrons resulting from virtual exchange of phonons is attractive. The superconductivity state is formed when this attractive interactions between electrons resulting from virtual exchange of phonons. This attractive interaction dominates the repulsive screened Coulomb interaction. It seems that both couplings play an important role in the mechanism of superconductivity [16]. The isotope effect coefficient predicted by BCS theory has the value $\alpha = 0.5$, which is in excellent agreement with the reported isotope effect in many metals like (Hg, Tl, Zn) and others [17]. The inverse isotope effect ($\alpha < 0$) and ($\alpha < 1/2$) have been reported in many materials [17,18]. These results put strict limits to BCS theory, particularly for the materials having strong coupling coefficients ($\lambda > 1.5$). The significance of studying the isotope effects lies in the possibility of identifying the interatomic forces that control the properties of superconducting materials such as electron-mediated phonons and Coulomb interactions. This paper focuses on the mathematical models which take into accounts the strong coupling superconductors which makes the isotope effect exponent has no longer universal value.

2. Isotope Effect Coefficient

2.1 Isotope Effect in BCS Theory

The relation between transition temperature T_c, typical phonon frequency ω and interaction strength $N(E_f)$ V using the weak coupling BCS theory as:

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

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 ħ ω, 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 ω which is proportional to $M^{-1/2}$ [19], where M is the ionic mass. Within the framework of the electron – phonon mechanism, the T_c can be described by the following relation:

$$T_c = A\,M^{-\alpha} \quad (2)$$

Where A is a constant, M is the mass of the element substituted by its isotope and α is the isotope effect coefficient, which is defined as:

$$\alpha = -\frac{\partial \ln T_c}{\partial \ln M} \approx -\frac{M}{T_c}\frac{\Delta T_c}{\Delta M} \quad (3)$$

where ΔT_c is the shift of the critical temperature substitution of isotopic mass and ΔM 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 α = 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 [20] argued that the following equation is more accurate than that in (3):

$$\propto_i = -\frac{lnT_c\,(i+1) - lnT_c(i)}{lnM_{i+1} - lnM_i} \approx -\frac{M_i}{T_{c(i)}}\frac{T_c(i+1) - T_c(i)}{M_{i+1} - M_i} \quad (4)$$

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 α_i.

Vora [21] has deduced from the best fit to the data of about twenty-five materials, the following equation for T_c:

$$T_c = \left(\frac{<\omega>}{10.71}\right)(\lambda - 0.3362) \quad (5)$$

Where <ω> is the average phonon frequency and λ 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 ThH_9 – ThD_9 system as example:

$$\propto = -\frac{M}{\Delta M}\frac{<\omega>_{ThD_9} - <\omega>_{ThH_9}}{<\omega>_{ThH_9}} \quad (6)$$

The isotope effect evaluation using the 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 [22].

It is noticeable that the D – derived optical phonon modes shift towards lower frequencies, relative to the corresponding H – derived modes. For instances, at 100 and 150 GPa, the lowest optical modes Γ point shift from 728 and 957 K in ThH_9 to 486 and 621 K in ThD_9, respectively [12]. According to these phonon frequencies, equation (6) gives α- values: 0.32 and 0.34 for the pressures of 100 and 150 GPa respectively. The average value of α is 0.33, while the predicted value from the critical temperature shift is 0.47 [12]. The error percent between the two methods is 27%. 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.

2.2 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 [23] assumed strong coupling between electrons and phonons and calculated the spectrum and the damping excitations. All superconductors are characterized as having weak ($\lambda_{opt}<<1$), intermediate ($\lambda_{opt}\approx 1$), and strong coupling ($\lambda_{opt}>>1$) [24], McMillan – Dynes [25,26] performed advanced analysis of the problem by utilizing the Eliashberg theory and proposed the critical temperature equation:

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

Here μ* is the effective Coulomb repulsion, which is assumed to be within a range of μ* = 0.1 – 0.2. Equation (7) is highly accurate for a wide range of coupling strength $\lambda_{opt} \leq 1.5$, and its 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:

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

For λ_{opt} >1.5, Allen and Dynes [27] proposed a correction factor should be included in the equation (7), so that it becomes:

$$T_c = T_c^{\circ} f_1 f_2 \quad (9)$$

Where f_1 is the "strong – coupling correction", and f_2 is the "shape correction". f_1 is calculated using the following equation:

$$f_1 = \left[1+\left(\frac{\lambda_{opt}}{2.46(1+3.8\mu^*)}\right)^{3/2}\right]^{1/3} \quad (10)$$

For the f_2 calculation, the empirical relation deduced from **Table1** in ref. 27 is used [24]:

$$f_2 = 1+(0.0241-0.0735\mu^*)\lambda_{opt}^2 \quad (11)$$

This parabolic function is deduced by the fit of tabulated f_2 values for all materials reported by Allen and Dynes [27].

A modified form of the isotope effect coefficient α is developed by Gor'kov and Kresin (GK) [28] 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 [28]:

$$\alpha \approx \frac{1}{2}\left[1-\frac{\lambda_{ac}}{\lambda_{opt}}\frac{\rho^2}{(\rho^2+1)^2}\right] \quad (12)$$

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

ϖ_{ac} is the average frequency of the acoustic phonons and λ_{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 (α =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 α – values. For this purpose, new groups of materials with superconductivity close to room temperature and different high pressures were used. 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

3.1. Isotope Effects in the fcc (YH_6 – YD_6) 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 (yttrium) 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 YH_6 – YD_6 system under high compression. The Coulomb pseudopotential $\mu^* = 0.1$ was assumed. The isotope coefficient α was determined by using the EM- model, the equation (8) and the GK- model, the equation (12). 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 100 and 300 K. When calculating the isotope effect from the equation (12), 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 λ_{opt} and λ_{ac}. This distinction allows comparison of the relative contributions of the optical and the acoustic phonons. Depending on [28], $\lambda_{opt} \approx 3\lambda_{ac}$, and $\tilde{\omega}_{opt} \approx 4\ \tilde{\omega}_{ac}$ were estimated. This estimate was used in a single case for sulfur hydride system [28] and was generalized to include, a similar system in properties, the super hydrides system as it is a convincing estimate due to its reliance on practical results for optical phonon frequencies and acoustic phonon frequencies. To the best of our knowledge, this model is used for the first time for predicting the isotope effect in superconducting systems beyond H_3S-D_3S.

Figure 1 demonstrates the variations of the superconducting parameters T_c, $\tilde{\omega}_{opt}$, λ_{opt}, and α with the pressure in gigapascals. The first three parameters mainly represent the superconducting materials properties of interest for predicting the isotope effect coefficient α. The pressure range extends from 100 to 300 GPa. Figure1a shows the dependence of the critical temperature T_c on pressure is weak, this predictions for T_c amount to a remarkable agreement with Ref. [29]. 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 than 1.5. The predicted T_c value is around 215 K at the pressure of 200 GPa. The recent reported value for YH_6 was 211 K at pressure of 201GPa [10]. As shown in (b) and (c) of Figure 1, the weak pressure dependence of T_c results from an almost perfect compensation between the average phonon frequency $\tilde{\omega}$, which increases with pressure and the e-ph coupling constant λ which instead decreases. This balance holds down to a pressure of 165 GPa. The isotope coefficient α was determined from the equations (8) and (12) which were used for the cases of strong electron – phonons coupling $\lambda > 1.5$. Both equations give approximately a constant value of α as a function of pressure (figure 1d). Both models used for calculating α give slight fluctuation as a function of pressure. The average value of α using the equation (8) is 0.488. The experimental value is (0.4) [9], the percent error is 22%. On the other hand, the average value of α using the equation (12) is 0.476, with a percent error of 19% from the experimental value. The results of α – values for YH_6 – YD_6 system shown in Table 1 indicate that the two models are convergent with each other but both of them predicted α – values deviated from the experimental value of about 20%. In order to verify more from both models in calculating α – 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 Th-H – Th-D. Table 2, and Table 3 summarize the data required to calculate α – values under strong coupling condition. Table 2 shows the data used for the $P6_3/mmc$-ThH_9 superconducting compound, while Table 3 demonstrates the data for F- m-3m ThH_{10} structure. The value of the Coulomb potential $\mu^* = 0.1$ was assumed. For ThH_9, the average value of α from equation (8) is 0.495.. On the other hand, when using the equation (12) in calculating the values of α, it was noticed that the average value of α was equal to 0.470, in excellent agreement with predicted value of $\alpha = 0.0.470$ in Ref. [12].

For Fm-3m ThH_{10}, the average α – value is 0.492 when using equation (8) (Table 3), while it was found that the average value of α when using equation (12) is 0. 468.These results clearly indicate that the EM model and the GK model reproduce α values closely to each other.

4. Conclusion

The calculations of the α – values for strong coupling coefficient superconductors indicate that both EM model and GK model give a convergent result with each other. As for the recent GK model, it was previously examined for only one case, which was H_3S-D_3S system, this research

proved the possibility of expanding its use to include other systems, due to its prediction of α – values close to the traditional EM model. The deviation of the isotope effect value in both models from the experimental value, even by 20% indicates that their reliability is critical. Accordingly, the development of a comprehensive theory for most of the conventional and unconventional superconducting materials would expedite the experimental efforts to reach superconductive materials that operate at room temperature and low pressure.

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Appendix

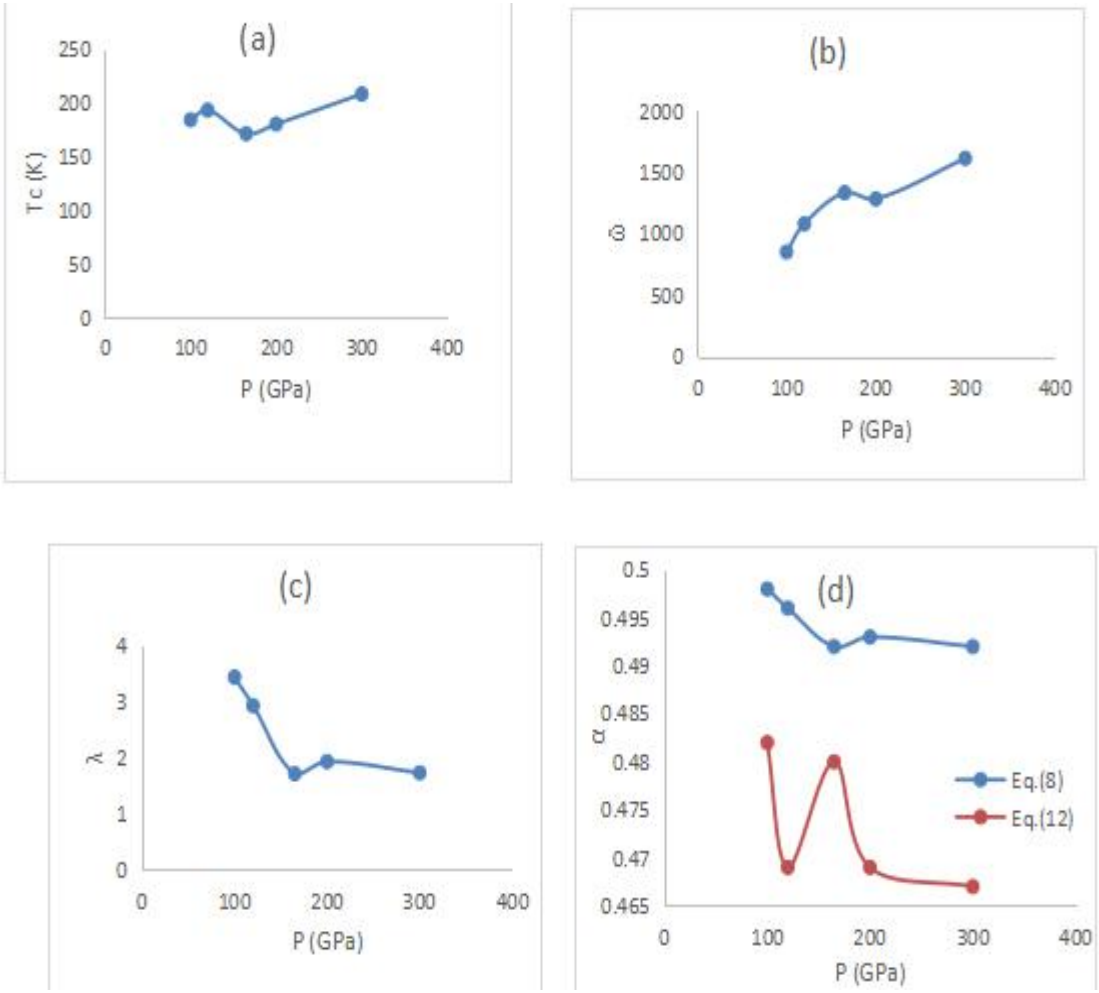

Fig. 1. The calculated superconducting parameters at different pressures for the YH_6 – YD_6 system (a) Calculated T_c of YH_6 as a function of pressure (b) The average values of phonon frequencies as a function of pressure (c) The coupling coefficients as a function of pressure (d) Calculated isotope effect coefficients α as a function of pressure for the EM model, equation (8), and GK model, equation (12).

Table 1. Calculated values of electron phonon coupling λ, average phonon frequency ϖ, T_c and the isotope effect coefficient α for superconducting yttrium hydride YH_6 under high compression. The Coulomb pseudopotential μ* = 0.1 was assumed.

Pressure (GPa)	**100**	**120**	**165**	**200**	**300**	
λ_{opt}	3.44[29]	2.93[11]	1.714[9]	1.93[30]	1.73[29]	
ϖ (K)	851[29]	1080[11])	1333[9]	1282[30]	1612[29]	
T_c°(K)	184	193	171	180	208	Eq.(7)
T_c(K)	312	268	218	215	242	Eq.(9)
α	0.498	0.496	0.492	0.493	0.492	Eq.(8)
α	0.482	0.469	0.48	0.469	0.467	Eq.(12)

Table 2. Calculated values of electron – phonon coupling coefficient λ, average phonon frequency ϖ, T_c and the isotope effect coefficient α at different pressure for superconducting thorium hydride ThH_9. The Coulomb pseudopotential μ* = 0.1 was assumed.

Pressure (GPa)	**110**	**150**	
λ_{opt}	2.15	1.73	Ref.(12)
ϖ(K)	981.7	957	Ref.(12)
T_c°(K)	144.7	181	Eq.(7)
T_c(K)	223.45	218	Eq.(9)
α	0.494	0.496	Eq.(8)
α	0.472	0.468	Eq.(12)

Table 3. Calculated values of superconducting parameters at different pressure. Electrons – phonons coupling coefficient λ, average phonons frequency ϖ, T_c and the isotope effect coefficient α for Thorium hydride ThH_{10}. The Coulomb pseudopotential μ* = 0.1 was assumed.

Pressure (GPa)	**100**	**200**	
λ_{opt}	2.19	1.58	Ref.(31)
ϖ (K)	1042	1215.8	Ref.(31)
T_c°(K)	159	145	Eq.(7)
T_c(K)	198	166	Eq.(9)
α	0.494	0.490	Eq.(8)
α	0.472	0.464	Eq.(12)

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مجلة كلية العراق الجامعة للهندسة والعلوم التطبيقية

تأثيرات النظام على المجاميع الجديدة فائقة التوصيلية $YD_6 - Yh_6 - Th - D - Th - H$

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الملخص .تكرس هذه الورقة البحثية لفحص نموذجين رياضيين تم تطويرهما من قبل الياشبرغ – ماكميلان و كوركوف – كريسن من اجل حل التناقض في النظرية الاصيلة للتوصيلية الفائقةBCS . صمم هذين النموذجين لغرض التنبؤ بتاثيرات النظير لبعض المواد فائقة التوصيلية والتي تمتلك معامل اقتران قوي قيمته اكبر من 1.5. ركزت الدراسة على المواد الفائقة التوصيلية المكتشفة حديثا والتي تقترب درجة حرارتها الحرجة الى درجة حرارة الغرفة، وبالتحديد المجموعتين $YD_6 - YH_6$ و Th-H – Th-D. على الرغم من ان قيم معامل النظير التي تم حسابها باستعمال النموذجين كانت متقاربة مع بعضها البعض الا انها تنحرف عن القيمة التجريبية بنسبة خطأ 20%. بناءا على نتائج الدراسة فان هذين النموذجين على الرغم من اهميتهما وبساطتهما في التنبؤ بتأثيرات النظيرلكن الاعتماد عليهما يحصل بموثوقية حرجة.