



Communication

The isotope effect in H₃S superconductorR. Szcześniak^{a,b}, A.P. Durajski^{a,*}^a Institute of Physics, Częstochowa University of Technology, Ave. Armii Krajowej 19, 42-200 Częstochowa, Poland^b Institute of Physics, Jan Długosz University in Częstochowa, Ave. Armii Krajowej 13/15, 42-200 Częstochowa, Poland

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ABSTRACT

The experimental value of H₃S isotope coefficient decreases from 2.37 to 0.31 in the pressure range from 130 GPa to 200 GPa. We have shown that the value of 0.31 is correctly reproduced in the framework of the classical Eliashberg approach in the harmonic approximation. On the other hand, the anomalously large value of the isotope coefficient (2.37) may be associated with the strong renormalization of the normal state by the electron density of states.

It is the most probable that the metallic hydrogen could be the superconductor with the very high value of the critical temperature (T_C) [1,2]. The expected high T_C is associated with the large Debye frequency (the mass of the proton is very small) and the lack of the electrons on the inner shells, which should significantly cause the increase of the electron-phonon coupling constant (λ) [3–5]. Unfortunately, the pressure of the hydrogen's metallization is very large ($p > 400$ GPa [6,7]). For this reason, the experimental confirmation of the theoretical predictions have not been obtained to this day.

In 2004 Ashcroft suggested the existence of the superconducting state in the hydrogen-rich compounds with the critical temperature comparable to T_C of the pure hydrogen, whereas the metallization pressure might be subjected to the significant decrease due to the existence of the chemical pre-compression [8]. Ashcroft's predictions were confirmed in many later papers. The selected results are presented in Fig. 1.

The superconducting state in the hydrogen sulfide with the exceptionally high value of the critical temperature ($T_C \sim 200$ K) was discovered in 2014 [27,28]. The detailed dependence of the critical temperature on the pressure for the compounds H₃S and D₃S is presented in Fig. 2. The experimental results [27–29] and the theoretical papers [30–36] suggest that the superconducting state in the hydrogen sulfide is induced by the electron-phonon interaction. In particular, the strong isotope effect was observed. However, the values of the isotope coefficient (α) significantly differ from the canonical value of 0.5 predicted by the BCS theory [37,38]. This behaviour can be connected with a flat band/steep band scenario which is realized analogous to the case of MgB₂ [39,40]. Taking into account the *clean limit*, the Bianconi-Perali-Valletta (BPV) theory [41–43] was proposed to describe the high-temperature superconducting state in hydrogen

sulfide [44–46]. The BPV theory considers high- T_C superconductivity made of multiple condensates. The optimum critical temperature is predicted for a first condensate in the BCS regime and the second one in the BCS-BEC crossover near a Lifshitz transition. At the Lifshitz transition, a change of the topology of the Fermi surface is induced by pressure or doping and it has been shown to control high temperature superconductivity [44]. The existence of two superconductivity gaps in H₃S could be confirmed in tunneling experiments at low temperatures [47].

In the presented paper, we have explained the experimental data for α on the basis of the classical and the extended Eliashberg formalism basing on the phonon pairing mechanism where the multiple bands of a metal are reduced to a single effective band (*dirty limit*). In the first step, on the basis of the experimental results, we determined the approximation lines $T_3^H S_C(p)$ and $T_3^D S_C(p)$, which served for the calculation of the isotope coefficient:

$$\alpha_{\text{exp}}(p) = -\frac{\ln[T_3^D S_C(p)] - \ln[T_3^H S_C(p)]}{\ln[m_D] - \ln[m_H]}, \quad (1)$$

where m_D and m_H are respectively the deuterium's and protium's atomic mass. The shape of the function $\alpha_{\text{exp}}(p)$ is plotted in Fig. 3. It can be clearly seen that the isotope coefficient decreases with the increasing pressure. In particular, it was obtained: $\alpha_{\text{exp}}(130 \text{ GPa}) = 2.37$ and $\alpha_{\text{exp}}(200 \text{ GPa}) = 0.31$.

The value of the isotope coefficient for $p=200$ GPa can be reproduced in the framework of the classical Eliashberg formalism. To this end, we solved numerically equations [48,49]:

$$\varphi_n = \frac{\pi}{\beta} \sum_{m=-1100}^{1100} \frac{K(i\omega_n - i\omega_m) - \mu^*(\omega_m)}{\sqrt{\omega_m^2 Z_m^2 + \varphi_m^2}} \varphi_m, \quad (2)$$

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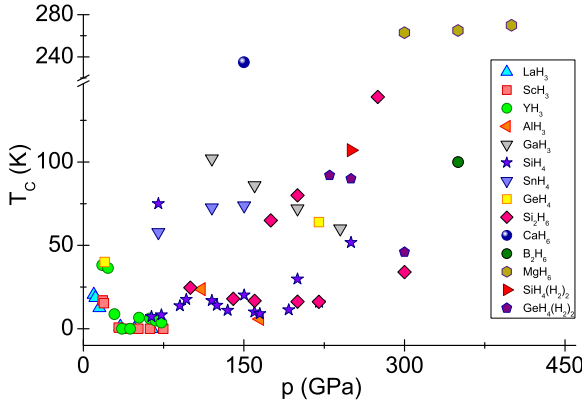


Fig. 1. (Color online) The predicted critical temperatures for the hydrogen-rich compounds. The results for tri-hydrides: LaH_3 , ScH_3 , YH_3 , AlH_3 , and GaH_3 are from the works: [9,10], and [11]. The results obtained for four-hydrides are presented in the papers: SiH_4 [12] (experiment), [13–15], SnH_4 [16], GeH_4 [17]. The results for six-hydrides and eight-hydrides were obtained in the works: Si_2H_6 [18,19], Ba_2H_6 [20], CaH_6 [21], MgH_6 [22] (see also [23]), $\text{SiH}_4(\text{H}_2)_2$ [24], and $\text{GeH}_4(\text{H}_2)_2$ [25,26].

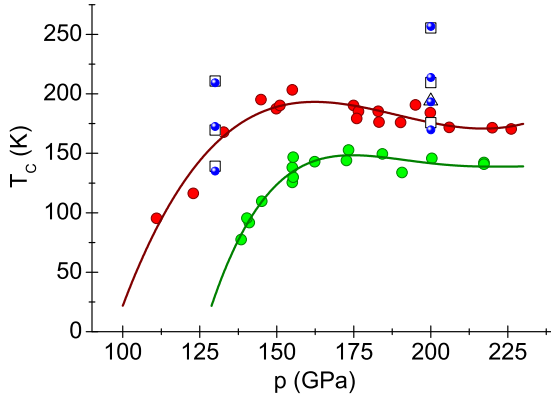


Fig. 2. (Color online) The influence of the pressure on the value of the critical temperature - H_3S (the red circles) and D_3S (the green circles) [29]. The lines were obtained using the approximation procedure. The squares represent the results obtained with the help of the classical Eliashberg equations in the harmonic approximation, the triangle represents the anharmonic analysis, and the blue spheres denote expression (5).

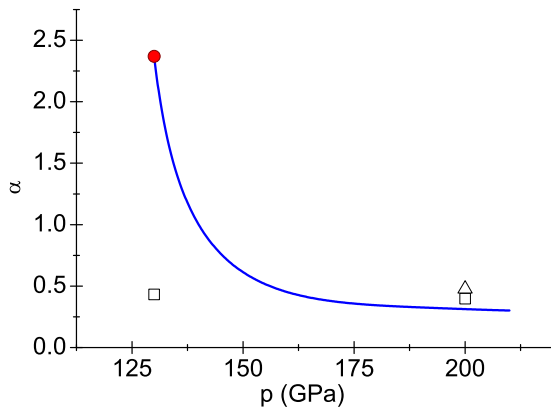


Fig. 3. (Color online) Blue line - the experimental values of the isotope coefficient on the basis of the formula (1). The squares were obtained in the framework of the classical Eliashberg formalism in the harmonic approximation. The triangle corresponds to the classical Eliashberg formalism - the anharmonic analysis. The red circle was obtained assuming the strong renormalization of the normal state by the electron density of states.

$$Z_n = 1 + \frac{1}{\omega_n \beta} \sum_{m=-1100}^{1100} \frac{\lambda(i\omega_n - i\omega_m)}{\sqrt{\omega_m^2 Z_m^2 + \varphi_m^2}} \omega_m Z_m, \quad (3)$$

where $\varphi_n = \varphi(i\omega_n)$ represents the order parameter function, and $Z_n = Z(i\omega_n)$ denotes the wave function renormalization factor. The

fermion Matsubara frequency is given by the formula: $\omega_n = \frac{\pi}{\beta}(2n - 1)$, $\beta = 1/k_B T$ (k_B is the Boltzmann constant). The electron-phonon pairing kernel has the following form: $K(z) = 2 \int_0^{+\infty} d\Omega \frac{\Omega}{\Omega^2 - z^2} \bar{\alpha}^2 F(\Omega)$. The Eliashberg functions ($\bar{\alpha}^2 F(\Omega)$) for $p=130$ GPa and $p=200$ GPa were calculated by Duan et al. [30].

The depairing electron correlations in the Eliashberg formalism are described with the use of the formula: $\mu^*(\omega_n) = \mu^* \theta(\omega_C - |\omega_n|)$. The quantity μ^* denotes the Coulomb pseudopotential, θ is the Heaviside function. ω_C represents the cut-off frequency: $\omega_C = 3\Omega_{\max}$, where Ω_{\max} is the Debye frequency. It should be noted that the Coulomb pseudopotential was defined by Morel and Anderson [50]:

$$\mu^* = \frac{\mu}{1 + \mu \ln\left(\frac{\omega_e}{\omega_{\ln}}\right)}. \quad (4)$$

The symbol μ is given by the formula: $\mu = \rho(0)U$, whereas $\rho(0)$ is the value of the electron density of states at the Fermi level, and U is the Coulomb integral. The quantity ω_e represents the characteristic electron frequency and the logarithmic phonon frequency is given by: $\omega_{\ln} = \exp\left[\frac{2}{\lambda} \int_0^{\Omega_{\max}} d\Omega \frac{\bar{\alpha}^2(\Omega)F(\Omega)}{\Omega} \ln(\Omega)\right]$. Note that the formalism presented above assumes isotropy of the superconducting state. This has been challenged in papers [44–47] where the Fermi surface of H_3S is predicted to consist of multiple sheets similar to those in MgB_2 . It is worth emphasizing that in the case of anisotropic description of the superconducting state it will be necessary to use the multi-band Eliashberg theory in the same way as it was done for MgB_2 [51–53], CaC_6 [54,55] or molecular hydrogen [3,56] to deal with the effects of possible variations of the superconducting energy gap on the Fermi surface. Accordingly, the isotropic Eliashberg spectral function and isotropic Coulomb pseudopotential should be generalized into a matrix form with the coefficients strongly correlated with the band structure. Of course the best way to analyze the superconducting state is solution of the full anisotropic Eliashberg equations with overt dependence of the order parameter on the wave vector. Such calculations are feasible (eg. MgB_2 [57]), however, they require enormous computational capacity.

In Fig. 2, we marked the values of the critical temperature calculated with the help of the Eliashberg equations. We presented the results for $\mu^* \in \{0.1, 0.2, 0.3\}$. Additionally, we also placed the value of T_C for $p=200$ GPa, determined beyond the harmonic approximation [58]. It turns out that the numerical results can be reproduced using the formula (see also Fig. 1):

$$k_B T_C = \omega_{\ln} \exp\left[\frac{-(1 + \lambda)}{\lambda - \mu^*(1 + 0.4747\lambda)}\right], \quad (5)$$

where the electron-phonon coupling constant should be calculated from: $\lambda = 2 \int_0^{\Omega_{\max}} d\Omega \frac{\bar{\alpha}^2(\Omega)F(\Omega)}{\Omega}$. We note that the Eq. (5) is close to the McMillan expression [59]. However, the numerical parameters possess the new values, which have been obtained on the basis of the least-squares analysis of the 300 values of T_C calculated by using the Eliashberg equations. The classical McMillan formula can not be used because the relative differences between the values of T_C calculated from the McMillan expression and obtained numerically from the Eliashberg equations are of the order of 30%.

On this basis, it was found out that the values μ^* corresponding to $[T_C]_{\text{exp}}$ are equal to 0.239 and 0.286, respectively for the pressure at 130 GPa and 200 GPa (the harmonic approximation), and 0.146 (the anharmonic analysis).

The expression on the isotope coefficient was derived using the dependence:

$$\alpha = \frac{\omega_{\ln}}{2T_C} \frac{dT_C}{d\omega_{\ln}}. \quad (6)$$

Thus:

$$\alpha = \frac{1}{2} \left[1 - \frac{(1 + \lambda)(1 + 0.4747\lambda)(\mu^*)^2}{(\lambda - \mu^*(1 + 0.4747\lambda))^2} \right]. \quad (7)$$

The theoretical results have the following form: $\alpha(130 \text{ GPa}) = 0.432$ and $\alpha(200 \text{ GPa}) = 0.397$ (the harmonic approximation), and $\alpha(200 \text{ GPa}) = 0.477$ (the anharmonic approach). It can be easily seen that the theoretical value of the isotope coefficient for $p=200 \text{ GPa}$ in harmonic approximation qualitatively well reproduce the experimental data. The parameter α in anharmonic case does not seem to agree with the experimental data. This disagreement should be clarified if more experimental data will be obtained or if the re-analysis of the Eliashberg function in the anharmonic approach will be made. In the case of $p=130 \text{ GPa}$ the discrepancy between the Eliashberg result and the result of the measure is extremely high, which denotes the collapse of the classical theoretical description.

The high value of the isotope coefficient in the terms of the lower pressures can be tried to explain by the pairing mechanism other than the electron-phonon mechanism [60]. However, the modification of the classical Eliashberg formalism should also be considered. From the theoretical point of view it highlights the big change of the electron density of states at and near the Fermi surface together with the pressure change. The *ab initio* calculations performed for $p=210 \text{ GPa}$ suggest the existence of the sharp peak of $\rho(\epsilon)$ very close to the Fermi surface [61]. The peak moves away from the Fermi level for the lower pressures. Thus the high value of the electron density of states at lower p are not very near the Fermi surface. In the considered case, this means the significant reduction of the number of Cooper pairs forming the superconducting state, because the greatest contribution to the superconducting condensate bring the electrons located near the Fermi level. The significant modification of the normal state in the studied system can be modelled by the renormalized unperturbed fermionic propagator with Green function, which accounts for the gap of arbitrary magnitude and depth.

Let us consider the renormalized Green function of the normal state, in which the depreciation of the electron density of states was taken into account [62]:

$$G_{\mathbf{k}}(i\omega_n) = -\frac{i\omega_n\tau_0 + \epsilon_{\mathbf{k}}\tau_3}{\omega_n^2 + \epsilon_{\mathbf{k}}^2 + B^2}A - \frac{i\omega_n\tau_0 + \epsilon_{\mathbf{k}}\tau_3}{\omega_n^2 + \epsilon_{\mathbf{k}}^2}(1 - A), \quad (8)$$

where τ_0, τ_3 are the Pauli matrices associated with the normal state and $\epsilon_{\mathbf{k}}$ is the electron energy. The parameters $A \in (0, 1)$ and B determine the depth and the width of the decrease in electron density of states with respect to the baseline at the Fermi level. Deriving the Eliashberg equations for the renormalized Green function and using the approximations were discussed in paper [62], the algebraic equation on the critical temperature can be obtained:

$$1 = \frac{\lambda}{1 + \lambda} \ln \left(\frac{\omega_{\text{ln}}}{2\pi k_B T_C} \right) - \frac{\lambda}{1 + \lambda} \left[f_1 \Psi \left(\frac{1}{2} \right) + 2f_2 \operatorname{Re} \Psi \left(\frac{1}{2} + \frac{iB}{2\pi k_B T_C} \right) + 2f_3 \operatorname{Re} \Psi \left(\frac{1}{2} + \frac{iB}{2\pi k_B T_C} \right) \right], \quad (9)$$

where:

$$g = \left[\frac{(1 - A)(1 + \lambda) + A}{1 + \lambda} \right]^{1/2}, \quad (10)$$

$$f_1 = \frac{(1 - A)^2}{g^2}, \quad (11)$$

$$f_2 = \frac{1}{2g^2} \left[g^2 - (1 - A)^2 + \frac{(1 - A - g^2)^2}{1 - g^2} \right], \quad (12)$$

$$f_3 = -\frac{1}{2g^2} \left[\frac{(1 - A - g^2)^2}{1 - g^2} \right]. \quad (13)$$

The symbol Ψ denotes the digamma function.

We simultaneously solved the Eqs. (6) and (9) in a numerical way. The input parameters for the pressure at 130 GPa were assumed ($\alpha = 3.37$ and $T_C = 157.9 \text{ K}$). It should be noted that in order to accurately determine the values of the parameters A and B , the calculations should start relatively close to the exact solution since the equation set possesses the many local minima. It turns out that the equations allow to reproduce the experimental values of the critical temperature and the isotope coefficient for $A=0.904$ and $B=29.12 \text{ meV}$. In the considered case, the renormalized Green function corresponds to the electron density of states in which in the vicinity of the Fermi surface does not appear the sharp maximum. This result simulates offset the electron density peak from the Fermi surface. The obtained values of A and B agree with the offset of the $\rho(\epsilon)$ peak from the Fermi surface presented in the papers [30,31], and [46].

The new approach to clarification the large value of the isotope coefficient in H_3S compound should be also discussed in the context of the classical Eliashberg theory. We notice that in the standard Eliashberg model all depairing processes are modelled parametrically by the Coulomb pseudopotential. Taking into account explicitly the reason responsible for the decrease of the critical temperature (in our case, it is the renormalization of the Green function by the offset of the electron density peak from the Fermi surface), we obtain the drastic reduction of μ^* . For this reason, we excluded μ^* from the Eq. (9). However, the adoption of the low value of Coulomb pseudopotential in the expression (9) only slightly lowers T_C .

In general, it can be said that the new approach to the problem of high value of the isotope coefficient means the modification of the Eliashberg equations (in particular, the equation for the critical temperature), by explicitly taking into account the renormalization of the normal state Green function. In the present case, the wave function renormalization factor is equal to $1 + \lambda$, and does not depend on the value of μ^* , due to the fact that for $T = T_C$ the order parameter vanishes in Eq. (3).

In conclusion, basing on the experimental data we determined the range of variation of the isotopic coefficient for H_3S superconductor in the function of the pressure. We showed that the isotope coefficient accepted the anomalously high values in the area of the lower pressures ($\sim 130 \text{ GPa}$). On the other hand, for the higher pressures ($\sim 200 \text{ GPa}$), the values of α are lower than those in the BCS theory. The conducted theoretical analysis has prove that the low values of the isotope coefficient could be reproduced in the framework of the classical Eliashberg formalism. The anomalously high values of α can be induced by the strong renormalization of the normal state associated with the significant changes of the electron density of states with the change in the pressure. Note that the proposed model does not require the non-phonon pairing mechanism.

Finally, we notice that due to greater amplitude of the zero-point vibrations of H atoms the anharmonic effects should significantly affect the value of T_C and α . The generalized Eliashberg theory have suggested that the anharmonicity could change the superconducting properties of the system for the following reasons: i) the existence of the Debye-Waller factors in the ionic potential, ii) the change in the spectral density of the one-phonon Green function induced by the phonon-phonon interactions, iii) the many-phonon process, and iv) the interference effects between one- and many-phonon processes [63]. Particularly, the results included in paper [58] suggest that the harmonic approximation overestimates the value of the electron-phonon coupling constant in H_3S by about 30% in comparison with the anharmonic result. Additionally, the critical temperature is much strongly dependent on the pressure than in the anharmonic case ($p > 200 \text{ GPa}$). However, Akashi et al. showed that taking into account also the plasmon mechanism the anharmonic effects should be compensated [36]. The detailed comparison between the harmonic and anharmonic Eliashberg functions which are available in literature the readers can find in the supplementary information for paper [34]. The above issues will be studied by us in the future in the context of the

new experimental data.

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