The electron-phonon coupling constant, the Fermi temperature and unconventional superconductivity in a room-temperature superconductor carbonaceous sulfur hydride

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Abstract

Recently, Snider et al (2020 Nature 586 373) discovered room-temperature superconductivity in highly-compressed carbonaceous sulfur hydride, H\textsubscript{x}(S,C)\textsubscript{y}. In this paper we report results of analysis of experimental temperature dependent magnetoresistance data, \(R(T,B)\), reported by Snider et al. The analysis shows that H\textsubscript{x}(S,C)\textsubscript{y} compound at pressure of \(P = 210\) GPa has the electron-phonon coupling constant \(\lambda_{e-ph} = 2.0\) and the ratio of critical temperature, \(T_c\), to the Fermi temperature, \(T_F\), in the range of \(0.010 \leq T_c/T_F \leq 0.018\). These deduced values are very close to ones reported for H\textsubscript{3}S at \(P = 155-165\) GPa. We also show that H\textsubscript{x}(S,C)\textsubscript{y} sample (compressed at \(P = 267\) GPa) with the record \(T_c = 285\) K exhibits similar \(T_c/T_F\) ratio. This means that in all considered scenarios the room-temperature superconductor H\textsubscript{x}(S,C)\textsubscript{y} falls into unconventional superconductors band in the Uemura plot, where all other highly-compressed super-hydride/deuterides are located. We also stress that our analysis reveals significant problems with raw magnetoresistance data, \(R(T,B)\), reported by Snider et al (2020 Nature 586 373). Thus, independent confirmation for \(T_c\) values, which were deduced from \(R(T,B)\) data, for highly-compressed H\textsubscript{x}(S,C)\textsubscript{y} compound is required.
I. Introduction

In 2015 Drozdov et al [1] reported on the discovery of the first near-room-temperature (NRT) superconductivity in highly-compressed sulphur hydride, H$_3$S. To date, NRT superconductivity has been observed in four super-hydrides/deuterides systems subjected to high pressure: Th-H [2], S-(H,D) [1,3-5], Y-H [6,7], La-(H,D) [8-10].

Recently, Snider et al [11] discovered the room-temperature superconductivity (RTS) in ternary compound H$_x$(S,C)$_y$ which exhibits the transition temperature within a range of $T_c = 275$-287 K to be subjected to external pressure of $P = 258$-270 GPa. While detailed phase/structural and phonon spectrum measurements [1-7,9,12], as well as the first-principle calculation studies [2,6,13-25] are on-going tasks for this first RTS compound [26], in this paper we report results of the analysis of temperature dependent magnetoresistance, $R(T,B)$, from which we deduced:

1. The charge carriers effective mass, $m_{\text{eff}}$;
2. The ground state superconducting coherence length, $\xi(0)$.
3. The Fermi temperature, $T_F$.

In a result, we find that in all considered scenarios room-temperature superconductor H$_x$(S,C)$_y$ has the ratio of critical temperature, $T_c$, to the Fermi temperature, $T_F$, within a range of $0.010 < T_c/T_F < 0.018$. This means that H$_x$(S,C)$_y$ falls in to unconventional superconductors band in the Uemura plot, where heavy-fermions, cuprates, pnictides and all near-room-temperature superconductors are located.

II. Description of the approach

Detailed description of the approach can be found elsewhere [27]. In short, we use the $T_c/T_F$ ratio to locate the position of the superconductor in the Uemura plot [28]. $T_F$ is calculated by:
\[
T_F = \frac{\pi^2}{8 \cdot k_B} \cdot m_{\text{eff}}^* \cdot \xi^2(0) \cdot \left(\frac{\alpha \cdot k_B \cdot T_c}{\hbar}\right)^2,
\]

where \(\hbar = h/2\pi\) is the reduced Planck constant, \(k_B\) is the Boltzmann constant, \(\alpha = \frac{2\Delta(0)}{k_B T_c}\), where \(\Delta(0)\) is the ground state energy gap. We deduce the ground state coherence length, \(\xi(0)\), by the fit of experimental \(R(T,B)\) curves to analytical approximate of Werthamer, Helfand and Hohenberg theory \([29,30]\) proposed by Baumgartner \(et\ al\) \([31]\):

\[
B_{c2}(T) = \frac{\phi_0}{2 \cdot \pi \cdot \xi^2(0)} \cdot \left(\frac{1 - \frac{T}{T_c}}{0.153 \cdot \left(1 - \frac{T}{T_c}\right)^2 - 0.152 \cdot \left(1 - \frac{T}{T_c}\right)^4 / 0.693}\right).
\]

It should be noted that we define \(T_c\) and \(B_{c2}\) by employing the most strict criterion, i.e. \(\frac{R(T)}{R_{\text{norm}}} \to 0\) (detailed discussion of the problem can be found elsewhere \([32]\)). We will designate this model as B-WHH model.

Because \(\alpha = \frac{2\Delta(0)}{k_B T_c}\) cannot be deduced from available experimental data, we perform our calculations by assuming that \(\alpha\) has lower and upper limits within \(s\)-wave superconducting gap symmetry:

\[
3.5 \leq \alpha \leq 4.5
\]

where the lower limit is the weak-coupling limit of Bardeen-Cooper-Schrieffer theory \([33]\) and the upper limit is the value for anharmonic phonons computed for precursor compound \(\text{H}_3\text{S}\) by first principle calculations approach \([17,34]\).

The charge carriers effective mass, \(m_{\text{eff}}^*\), has been calculated by the use of the Eliashberg’s theory expression \([35]\):

\[
m_{\text{eff}}^* = (1 + \lambda e^{-\mu \cdot \lambda_{e-ph}}) \cdot m_e
\]

where \(m_e\) is the electron mass and the electron-phonon coupling constant \(\lambda_{e-ph}\) is deduced by the use of the advanced McMillan equations \([32,36]\):

\[
T_c = \left(\frac{1}{1.45}\right) \cdot T_0 \cdot e^{-\left(\frac{1.04 \cdot (1 + \lambda_{e-ph})}{\lambda e^{-ph} - \mu^* \cdot (1 + 0.62 \lambda_{e-ph})}\right)} \cdot f_1 \cdot f_2^*
\]
where $T_\theta$ is the Debye temperature, and

$$f_1 = \left( 1 + \left( \frac{\lambda_{e-ph}}{2.46(1+3.8\cdot\mu^*)} \right)^{3/2} \right)^{1/3}$$

$$f_2^* = 1 + (0.0241 - 0.0735 \cdot \mu^*) \cdot \lambda_{e-ph}^2.$$  

where $\mu^*$ is the Coulomb pseudopotential parameter (ranging from $\mu^* = 0.13 - 0.16$ [32,36]) for which in this paper we use an average value of $\mu^* = 0.13$. And the Debye temperature was deduced from the fit of $R(T, B = 0)$ data to Bloch-Grüneisen (BG) equation [37,38]:

$$R(T, B = 0) = R_0 + A \cdot \left( \frac{T}{T_\theta} \right)^5 \int_{0}^{\frac{T_\theta}{T}} \frac{x^5}{(e^x-1)(1-e^{-x})} \cdot dx$$

where $T_\theta$, $R_0$ and $A$ is a free-fitting parameter. It should be noted, that the procedure to deduce the electron-phonon coupling constant, $\lambda_{e-ph}$, by combined use of the Bloch-Grüneisen and the McMillan equations is widely used in the field [39,40].

It is important to note that analysed $R(T,B)$ datasets were directly extracted from Figs. 1,2 of Ref. 11, in the same routine as it has been done for $R(T,B)$ datasets of other highly-compressed superconductors which we analysed in our recent papers [27,32]: black phosphorous (raw data reported by Shirotani et al [41] in their figure 5), boron (raw data reported by Eremets et al [42] in their figure 2), germanium arsenide (raw data reported by Liu et al [43] in their figure 3), silane (raw data reported by Eremets et al [44] in their figure 2), $\zeta$-phase of O$_2$ (raw data reported by Shimizu et al [45,46,47]), sulphur (raw data reported by Shimizu et al [48] in their Fig. 10), lithium (raw data reported by Shimizu et al [49] in their Fig. 2), sulphur hydride/deuteride (raw data reported by Einaga et al [50] in their Fig. 3(a,b); by Drozdov et al [1] in their figure 2(b); and by Mozaffari et al [51] in their figure 1), lanthanum hydride/deuteride (raw data reported by Drozdov et al [52] in their figures 1,2,4, and extended data figures 2,3,5).
III. Results and discussion

Snider et al [11] in their Fig. 1 reported several $R(T,B=0)$ curves for three highly-compressed $H_x(S,C)_y$ samples. To be reliably fitted to Eq. 8, the $R(T,B=0)$ curve should be measured at reasonably wide temperature range and also should not to be distorted by any experimental artefact, which can change the shape of $R(T,B=0)$ curve. From 10 presented in Figs. 1,2 [11] experimental $R(T,B)$ curves, the only one dataset designated as “Run 3” at $P = 210$ GPa (Fig. 1) was converged by the fit to Eq. 8. It should be noted that fits of other datasets to Eq. 8 either do not converge, either after the converging, deduced values have some large uncertainty boundaries. This is very unusual behaviour of $R(T,B=0)$ datasets for $H_x(S,C)_y$ samples reported by Snider et al [11], because 27 $R(T,B=0)$ datasets for a variety of highly-compressed superconductors (ranging from elements to superhydrides/superdeuterides) reported by different research groups [1,41-52] were fitted to Eq. 8 with a good quality and accuracy [27,32]. In this paper, we analyse raw $R(T,B)$ datasets for $H_x(S,C)_y$ samples reported by Snider et al [11] in an assumption that these datasets are accurate and correct. However, it should be stressed that independent confirmation/disprove primary results reported by Snider et al [11], including $T_c$ values, is required [53].

In Fig. 1 we show the fit of $R(T,B = 0)$ curve (Run 3, $P = 210$ GPa) to Eq. 8. Deduced Debye temperature is $T_\theta = 1497 \pm 8 \ K$. By the use of Eqs. 5-7, and taking in account $T_c = 190 \ K$, one can calculate $\lambda_{e-ph} = 2.0$. This value is in a very good agreement with computed $\lambda_{e-ph} = 1.84 (200 \ GPa)$ and $\lambda_{e-ph} = 1.71 (250 \ GPa)$ reported by Errea et al [17] for precursor binary compound $H_3S$. Based on pivotal result of Duan et al [54] who calculated $\lambda_{e-ph} = 2.19 (200 \ GPa)$ for $H_3S$, we can conclude that deduced by us $\lambda_{e-ph} = 2.0$ for $H_x(S,C)_y$ (Run 3, $P = 210$ GPa) is very close to $\lambda_{e-ph}$ of binary $H_3S$ compound, which perhaps means in turn, that studied sample has composition of $H_3S$, rather than claimed
$H_x(S,C)_y$. In addition, this sample has $T_c = 190$ K, which is, again, in the $T_c$ range for $H_3S$ binary compound.

Based on deduced $\lambda_{e-ph}$ value, the charge carriers effective mass is:

$$m_{eff}^* = (1 + \lambda_{e-ph}) \cdot m_e = 3.0 \cdot m_e$$

(9)

It should be noted that this value is in a good agreement with $m_{eff}^* = 2.76 \cdot m_e$ reported by Durajski [16] for the precursor compound $H_3S$.

![Figure 1](image-url)

**Figure 1.** Resistance data, $R(T)$, and fit to BG model (Eq. 8) for $H_x(S,C)_y$ sample compressed at $P = 210$ GPa (raw data is from Ref. 11, where the sample designated as Run 3). The fit quality is $R = 0.9998$. 95% confidence bar is shown. Green balls show bounds for which $R(T)$ data was used for the Eq. 8 fit.

Snider et al [11] in their Fig. 2(b) reported $B_{c2}(T)$ data for this sample. We extract $B_{c2}(T)$ data directly from Fig. 2(b) of Ref. 11. The fit of $B_{c2}(T)$ to Eq. 2 is shown in Fig. 2(a).

Deduced ground state coherence length is $\xi(0) = 2.39 \pm 0.04 \, nm$. It should be noted that Eq. 2 is a good extrapolative tool, that can be proved for the case of highly-compressed LaH$_{10}$. Drozdov et al. [9] reported first $B_{c2}(T)$ dataset for this NRT superconductor for applied field up to $B_{appl} = 9$ T. This dataset was used to extrapolate $B_{c2}(0)$ value for LaH$_{10}$ in
Ref. [55]. Recently, Sun et al [56] report new experimental $B_{c2}(T)$ dataset for LaH$_{10}$, which was measured on the world-top magnetic field facilities with applied field up to $B_{\text{appl}} = 60$ T. It can be seen that new experimental data is pretty much reproduced extrapolated values obtained by the use of Eq. 2 for dataset measured up to $B_{\text{appl}} = 9$ T.

![Figure 2](image)

**Figure 2.** Superconducting upper critical field, $B_{c2}(T)$, and data fit to B-WHH model (Eq. 2) for two $H_x(S,C)_y$ samples compressed at (a) $P = 210$ GPa and (b) $P = 267$ GPa (raw data is from Ref. 11). (a) the fit quality is $R = 0.9993$; (b) the fit quality is $R = 0.9949$. 95% confidence bars are shown.

From deduced $\xi(0)$, $m^*_e$, and $T_c$, we calculate value ranges for $T_F$ and $T_c/T_F$, by assuming $\alpha$ within its upper and lower limits (Eq. 3). The results are in Table 1.
Table I. Deduced and calculated parameters for RTS carbonaceous sulfur hydride. For calculations we use deduced $m_{eff}^* = 3.0 \cdot m_e$. The highest and the lowest values for $\frac{T_c}{T_F}$ are marked in bold.

| Pressure (GPa) | Deduced $T_c$ (K) | Deduced $\xi(0)$ (nm) | Assumed $\frac{2\Delta(0)}{k_B T_c}$ | $T_F (10^4 \text{ K})$ | $T_c/T_F$ |
|---------------|------------------|----------------------|---------------------------------|-------------------|------------|
| 210 | 189.8 ± 0.5 | 2.39 ± 0.04 | 3.5 | 1.05 ± 0.03 | **0.018 ± 0.001** |
| 267 | 286.5 ± 0.5 | 2.20 ± 0.09 | 3.5 | 1.68 ± 0.17 | 0.017 ± 0.001 |

Snider et al [11] in their Fig. 2 reported $R(T,B)$ curves for RTS sample designated as “II: 267 GPa” from which we deduced $B_{c2}(T)$ dataset by employing mention above criterion of $\frac{R(T)}{R_{norm}} \rightarrow 0$. The fit of this dataset to Eq. 2 is shown in Fig. 2(b). Deduced ground state coherence length is $\xi(0) = 2.00 \pm 0.09 \text{ nm}$. Normal part of this $R(T, B = 0)$ curve was measured at very narrow temperature range that the fit of $R(T, B = 0)$ to BG model does not converge. Thus, we use $m_{eff}^* = 3.0 \cdot m_e$ deduced for sample compressed at $P = 210 \text{ GPa}$, because as this was shown in many first-principle calculation studies [15,17] performed on precursor binary compound $\text{H}_3\text{S}$, that in the range of $P = 200-250 \text{ GPa}$ the charge carriers effective mass, $m_{eff}^*$, is changing by less than 10%. Because the Uemura plot [28] is log-log scale plot, the use of deduced $m_{eff}^* = 3.0 \cdot m_e$ in Eq. 1 cannot significantly perturb the result. From known $\xi(0)$, $m_{eff}^*$, and $T_c$ values we calculate $T_F$ and $T_c/T_F$ (Table 1).

Both analysed samples of highly-compressed $\text{H}_x(\text{S,C})_y$ are shown in the Uemura plot (Fig. 3). It can be seen, that in all considered scenarios the carbonaceous sulfur hydride has $0.010 \leq T_c/T_F \leq 0.018$ and falls in the unconventional superconductors band, where heavy-fermions, pnictides, cuprates and highly-compressed near-room-temperature superconductors are located.
The equation: temperature of Eq. 3, from which one can obtain: 43 meV < 2.0 parameter superconductors are shown for clarity. The amplitude of the ground state superconducting gap, $\Delta(0)$, cannot be derived in a way presented in Extended Data Fig. 3b [11]. We can make estimation for the ground state London penetration depth, $\lambda(0)$, by taking in account that practically all non-elemental superconductors are type-II superconductors, and most of them have the Ginsburg-Landau parameter $\kappa(0) = \frac{\lambda(0)}{\xi(0)}$ within a range of $60 < \kappa(0) < 120$ [61-67]. Based on deduced $\xi(0) = 2.0$-$2.4$ nm, expected value for $\lambda(0)$ is within a range: $120$ nm < $\lambda(0)$ < $290$ nm.

The amplitude of the ground state superconducting gap, $\Delta(0)$, can be calculated by the use of Eq. 3, from which one can obtain: $43$ meV < $\Delta(0)$ < $56$ meV. It should be noted that temperature and field dependences of superconducting gap, $\Delta(T,B)$, cannot be calculated by the equation:
\[ \Delta(T) = \Delta(0) \cdot 1.76 \cdot k_B \cdot T_c \cdot \sqrt{1 - \frac{T}{T_c}}, \]  

which was used by Snider et al [11] in their Extended Data Fig. 3,d. An accurate analytical expression for \( \Delta(T,B=0) \) for s-wave symmetry has been given by Gross-Alltag et al [68]:

\[ \Delta(T) = \Delta(0) \cdot \tanh \left[ \frac{\pi k_B T_c}{\Delta(0)} \cdot \sqrt{\eta \cdot \frac{\Delta C}{C} \cdot \frac{T_c}{T} - 1} \right] = \Delta(0) \cdot \tanh \left[ \frac{2\pi}{\alpha} \cdot \sqrt{\eta \cdot \frac{\Delta C}{C} \cdot \left( \frac{T_c}{T} - 1 \right)} \right], \]  

where \( \Delta C/C \) is the relative jump in electronic specific heat at \( T_c \), and \( \eta = 2/3 \).

IV. Conclusions

The discovery of superconductivity in highly-compressed \( \text{H}_3\text{S} \) by Drozdov et al [1] heralded the era of room-temperature superconductivity. To date, several superhydride/superdeuteride superconductors have been synthesised: \( \text{BaH}_{12} \) [23], \( \text{PrH}_9 \) [69], \( \text{ThH}_9/\text{ThH}_{10} \) [2], \( \text{YH}_4/\text{YH}_6 \) [6,7], \( \text{LaH}_{10}/\text{LaD}_{11} \) [8,9], \( \text{H}_x(\text{S,C})_y \) [11]. The latter compound, in accordance with recent report by Snider et al [11], exhibits the record transition temperature of \( T_c = 285 \) K at pressure of \( P = 267 \) GPa. In this paper we extract data reported in Figs. 1,2 of Ref. 11 and deduce the electron-phonon coupling constant, \( \lambda_{e-ph} = 2.0 \), the ground state coherence length, \( \xi(0) = 2.20 \pm 0.09 \) nm, and the Fermi temperature, \( T_F = (1.7-2.8) \times 10^4 \) K, for the \( \text{H}_x(\text{S,C})_y \) superconductor. From these deduced values, we calculate the \( T_c/T_F \) ratio in \( \text{H}_x(\text{S,C})_y \) compound and find that in all considered scenarios this RTS compound should be classified as unconventional superconductors.

We should stress that our report is based on non-independently confirmed data reported by Snider et al [11], which we, in this paper, assume to be correct and accurate, despite our analysis reveals significant problem with raw \( R(T,B) \) data reported by Snider et al [11]. Thus, independent confirmation of the magnetoresistance data, including \( T_c \) values, for highly compressed \( \text{H}_x(\text{S,C})_y \) compound is required [53].
Acknowledgement

Author thanks financial support provided by the state assignment of Minobrnauki of Russia (theme “Pressure” No. AAAA-A18-118020190104-3) and by Act 211 Government of the Russian Federation, contract No. 02.A03.21.0006.

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