The dominance of non-electron–phonon charge carrier interaction in highly-compressed superhydrides

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Received 5 April 2020, revised 16 July 2021
Accepted for publication 2 August 2021
Published 15 September 2021

Abstract

The primary mechanism governing the emergence of near-room-temperature superconductivity (NRTS) in superhydrides is widely accepted to be the electron–phonon interaction. If so, the temperature-dependent resistance, $R(T)$, in these materials should obey the Bloch–Grüneisen (BG) equation, where the power-law exponent, $p$, should be equal to the exact integer value of $p = 5$. However, there is a well-established theoretical result showing that the pure electron–magnon interaction should be manifested by $p = 3$, and $p = 2$ is the value for pure electron–electron interaction. Here we aimed to reveal the type of charge carrier interaction in the layered transition metal dichalcogenides PdTe$_2$, high-entropy alloy (ScZrNb)$_{0.65}$[RhPd]$_{0.35}$ and highly-compressed elemental boron and superhydrides H$_3$S, LaH$_x$, PrH$_9$ and BaH$_{12}$ by fitting the temperature-dependent resistance of these materials to the BG equation, where the power-law exponent, $p$, is a free-fitting parameter. The results showed that the high-entropy alloy (ScZrNb)$_{0.65}$[RhPd]$_{0.35}$ exhibited pure electron–phonon mediated superconductivity with $p = 4.9 \pm 0.4$. Unexpectedly, we revealed that all studied superhydrides exhibit $1.8 < p < 3.2$. This implies that it is unlikely that the electron–phonon interaction is the primary mechanism for the Cooper pairs formation in highly-compressed superhydrides, and alternative pairing mechanisms, for instance, the electron–magnon, the electron–polaron, the electron–electron and other pairing mechanisms should be considered as the origin for the emergence of NRTS in these compounds.

Keywords: hydrogen-rich superconductors, near-room-temperature superconductors, charge carriers interaction

(Some figures may appear in colour only in the online journal)

1. Introduction

The discovery of the first superhydride/superdeuteride superconductor Th$_4$H$_{15}$/Th$_4$D$_{15}$ by Satterthwaite and Toepke [1] was based on a very clear idea, which they expressed as [1]: ‘There has been theoretical speculation [2] that metallic hydrogen might be a high-temperature superconductor, in part because of the very high Debye frequency of the proton lattice. With high concentrations of hydrogen in the metal hydrides one would expect lattice modes of high frequency and if there exists an attractive pairing interaction one might expect to find high-temperature superconductivity in these systems also.’ Surprisingly enough, Satterthwaite and Toepke [1] found that isotopic counterparts Th$_4$H$_{15}$ and Th$_4$D$_{15}$ have the same superconducting transition temperature, $T_c$, which is in direct contradiction with the theory of electron–phonon-mediated superconductivity [3, 4], including the case of hydrogen-rich metallic alloys [5]. Later it was found that the PdH-PdD-PdT
system exhibits the inverse isotope effect [6, 7], where heavier counterparts have higher superconducting transition temperatures in comparison with isotopically lighter compounds.

In this regard, we should mention that, despite a milestone experimental discovery reported by Drozdov et al [8] regarding the near-room-temperature superconductivity (NRTS) in highly-compressed sulphur hydride, a recent report by Minkov et al [9] showed that the isotope effect in the H$_3$S-D$_3$S system is not as prominent as was initially reported in 2015. A primary open question here is what the exact stoichiometry of the hydrogen isotopes in NRTS samples is. There is some experimental evidence [10] that hydrogen- and deuterium-based NRTS phases have different hydrogen isotopes stoichiometry and, thus, it is possible that the observed difference in $T_c$ for isotopic counterparts is originated from the stoichiometry, rather than from the fundamental difference in the phonon spectra.

Because the charge carriers can form the Cooper pairs not only by the attractive electron–phonon interaction [11–14], there is a necessity to reaffirm or disprove the electron–phonon-mediated superconductivity in the NRTS superhydrides. In an attempt to do this, here we analysed the temperature-dependent resistance, $R(T)$, in several highly-compressed superhydrides by using the generalized Bloch–Grüneisen (BG) equation [15, 16], where the power-law exponent, $p$, is assumed to be a free-fitting parameter.

In the result we show the dominance of non-electron–phonon charge carrier interactions (in particular, the electron–magnon and the electron–electron interactions) in highly-compressed H$_3$S, LaH$_3$, PrH$_3$, CeH$_3$, and BaH$_{12}$ superhydrides. This result is in good accord with reports [17, 18] where it was shown that NRTS superconductors are in the unconventional superconductors region of the Uemura plot [19, 20]. This implies that superhydride superconductors exhibit a non-electron–phonon pairing mechanism.

2. Model description

To determine the type of the charge carrier interaction in NRTS, here we proposed to use an advanced version of the BG equation [15, 16] which can be represented in the most general form as [21]:

$$R(T) = R_0 + A_1 \cdot T + \sum_{\nu} A_{\nu} \cdot \left( \frac{T}{T_\nu} \right)^p \cdot \int_0^{T_\nu} \frac{x^p}{(e^x - 1) \cdot (1 - e^{-x})} \cdot dx$$  \hspace{1cm} (1)

where $R_0$ is the resistance at $T \rightarrow 0$ K, $T_\nu$ is the Debye temperature, $A_{\nu}$ are weighting parameters, and $p$ is the power-law exponent which has a theoretical integer value for the following interaction mechanisms [21]:

$$p = \begin{cases} 
2 & \text{for the electron–electron interaction} \\
3 & \text{for the electron–magnon interaction} \\
5 & \text{for the electron–phonon interaction}
\end{cases}$$  \hspace{1cm} (2)

However, as it is pointed out in [22], equation (1) has a linear limit for $p \rightarrow 1$:

$$\lim_{p \rightarrow 1} \left( \frac{T}{T_\nu} \right)^p \cdot \int_0^{T_\nu} \frac{x^p}{(e^x - 1) \cdot (1 - e^{-x})} \cdot dx \rightarrow \left( \frac{B}{T_\nu} \right) \cdot T$$  \hspace{1cm} (3)

where $B$ is a constant. Thus, the linear term in equation (1) can be represented as an integral part at $p \rightarrow 1$ with weighting factor $A_1$:

$$R(T) = R_0 + \sum_{\nu} A_{\nu} \cdot \left( \frac{T}{T_\nu} \right)^p \cdot \int_0^{T_\nu} \frac{x^p}{(e^x - 1) \cdot (1 - e^{-x})} \cdot dx.$$  \hspace{1cm} (4)

We should stress that equation (4) was never applied to the analysis of experimental $R(T)$ data, because there is a sum of integrals and, thus, the fitting procedure is over-parametrized. However, to the best of the author’s knowledge, in all published work (except, the work by Jiang et al [23] and another recent report [22]) equation (4) was used for the electron–phonon integrand, i.e. $p = 5$ (see, for instance, [24–26]).

One possible way to use equation (4) is to reduce the number of integrals to one, but in this integral the power-law exponent, $p$, will be a free-fitting parameter:

$$R(T) = R_0 + A_p \cdot \left( \frac{T}{T_\nu} \right)^p \cdot \int_0^{T_\nu} \frac{x^p}{(e^x - 1) \cdot (1 - e^{-x})} \cdot dx.$$  \hspace{1cm} (5)

where we introduce the designation of $T_\nu$ for the characteristic temperature for the case when $p$ is a free-fitting parameter, while the designation of $T_\nu$ is kept for $p = 5$.

Thus, the dominant charge carrier interaction mechanism in given materials can be determined from comparison of the deduced free-fitting parameter $p$ with theoretical values for pure cases (equations (1, 2)).

To the best of the author’s knowledge, equation (5) was first used by Jiang et al [23] to reveal the dominant interaction mechanism in Sr$_2$Cr$_3$As$_2$O$_2$ ferrimagnet. The resulting mechanism was established as the electron–magnon interaction (with deduced $p = 3.34$ [23]), which was the first confirmation of the approach validity. More recently, in [22], the approach was applied to pure copper, iron and cobalt (for which $R(T)$ was reported by Matula [27], Teixeira [28], and White and Woods [29]). Also, the approach confirmed the electron–phonon-mediated superconductivity in ReBe$_{22}$ (for which $R(T)$ was reported by Shang et al [24]). On the other hand, for the highly compressed superconducting $\varepsilon$-phase of iron (for which $R(T)$ was reported by Shimizu et al [30] and by Jacard et al [31]), and for twisted bilayer graphene superlattice (for which $R(T)$ was reported by Polshin et al [32]) the non-electron–phonon mechanisms of superconductivity were revealed.
Here, to fit $R(T)$ data we used the recently-proposed equation [33]:

$$R(T) = R_0 + \theta \left( T_c^{\text{onset}} - T \right) \cdot \left( \frac{R_{\text{norm}}}{I_0 \left( F \cdot \left( 1 - \frac{T}{T_{\text{char}}} \right)^{3/2} \right)} \right)^2$$

$$+ \theta \left( T - T_c^{\text{onset}} \right) \cdot \left( R_{\text{norm}} + A \cdot \left( \frac{T}{T_{\text{char}}} \right)^p \int_0^{T_{\text{norm}}} \frac{x^p}{(e^x - 1) \cdot (1 - e^{-x})} \cdot dx \right)$$

(6)

where, instead of fixed $p = 5$ (as it was the case in [33]), here we used $p$ as a free-fitting parameter. $T_c^{\text{onset}}$ is the free-fitting parameter of the onset of superconducting transition, $R_{\text{norm}}$ is the sample resistance at the onset of the transition, $\theta(x)$ is the Heaviside step function, $I_0(x)$ is the zero-order modified Bessel function of the first kind and $F$ is a free-fitting dimensionless parameter. More details about equation (6) can be found elsewhere [22, 34].

It should be noted that there is an alternative approach to analyse $R(T)$ datasets [21, 27, 29, 31, 35] by replacing equation (5) by a simple power-law fitting function:

$$R(T) = R_0 + A_N \cdot T^N$$

(7)

where $N$ is a free-fitting parameter. This approach assumes that the deduced $N$-value (in equation (7)) is an accurate approximation for $p$-value in equation (5). Thus, in this assumption, the fitting equation for superconductors is:

$$R(T, B) = R_0 + \theta \left( T_c^{\text{onset}} - T \right) \cdot \left( \frac{R_{\text{norm}}}{I_0 \left( F \cdot \left( 1 - \frac{T}{T_{\text{char}}} \right)^{3/2} \right)} \right)^2$$

$$+ \theta \left( T - T_c^{\text{onset}} \right) \cdot \left( R_{\text{norm}} + A_N \cdot \left( T^{N - \left( T_c^{\text{onset}} \right)^N} \right) \right).$$

(8)

There is a need to point out a fundamental problem associated with the use of equations (7) and (8), which is the unit of the free-fitting parameter $A_N$. From the logic it should have the unit of $\Omega \cdot K^{-N}$, and if (as we show below) $N = 2.69$ for some material, the unit should be $\Omega \cdot K^{-2.69}$. However, there is no physical parameter or measurable value which has the unit of $\Omega \cdot K^{-2.69}$. This problem can be resolved if equation (7) is transformed to:

$$R(T) = R_0 + A_N \cdot \left( \frac{T}{T_{\text{char}}} \right)^N$$

(9)

where $T_{\text{char}}$ is some characteristic temperature. However, equation (9) cannot be used to fit $R(T)$ data, because the parameter $A_N$ (which has unit of Ohm in equation (9)), $T_{\text{char}}$, and $N$ have mutual dependence $\equiv 1$. This can be resolved if $A_N$ in equation (9) will be fixed to some conventional value. However, there is no clarity as to what this value and the physical meanings for $A_N$ and $T_{\text{char}}$ can be (equation (9)).

However, to demonstrate that the widely-used power-law approximation (equations (7) and (8)) cannot be reliable substitution for equations (5) and (6), we also fitted $R(T)$ data for elemental copper and silver, PdTe$_2$, high-entropy alloy (ScZrNb)$_{0.65}$[RhPd]$_{0.35}$ and several highly compressed superconductors to equations (5)–(8) and reported the results herein.

### 3. Results

#### 3.1. Elemental copper

First, we made a comparison of fits to equations (5) and (7) for temperature-dependent resistivity, $\rho(T)$, for pure copper. Experimental $\rho(T)$ data was measured by Teixeira [28] and this dataset was referred to as dataset #269 by Matula [27]. For the analysis we used $\rho(T)$ data in two temperature ranges, $1.2 \, K \leq T \leq 34.2 \, K$ and $1.2 \, K \leq T \leq 70.6 \, K$, and, thus, both analysed $\rho(T)$ datasets were within low-$T$ ranges of $\frac{T}{T_\rho} \leq \frac{1}{10}$ and of $\frac{T}{T_\rho} \leq \frac{1}{3}$ respectively, where $T_\rho = 320–342 \, K$ [27, 36].

$\rho(T)$ data fits to equations (5) and (7) for the temperature range of $\frac{T}{T_\rho} \leq \frac{1}{10}$ are shown in figures 1(a) and (b), where deduced $N = 4.72 \pm 0.02$ and $p = 5.08 \pm 0.03$. These values are in the proximity of the expected value of 5. However, the $N$-value deduced for $\rho(T)$ data measured at twice wider temperature range ($\frac{T}{T_\rho} \leq \frac{1}{3}$) is significantly different, $N = 3.39 \pm 0.04$ (figure 1(c)), while deduced $p = 4.82 \pm 0.02$ remains practically the same (figure 1(d)). This result is in agreement with the general understanding [21, 27–29] that the power-law function (equation (7)) is a good approximation for the BG equation (equation (5)) at $\frac{T}{T_\rho} \leq \frac{1}{30}$.

Deduced Debye temperatures for two temperature ranges, i.e. $T_\rho \,(T < 34 \, K) = 306 \pm 4 \, K$ and of $T_\rho \,(T < 70 \, K) = 347 \pm 1 \, K$, are in reasonable agreement with reported $T_\rho = 320–342 \, K$ for pure copper [36].

#### 3.2. Elemental silver

To demonstrate that the approach described in the previous section is reliable, in this section we performed the analysis for pure elemental silver. In figure 2 we showed experimental $\rho(T)$ data reported by Teixeira [28] (this dataset was referred to as dataset 103 by Matula [27]) and data fits to equations (5) and (7). For the analysis we used $\rho(T)$ data in two temperature ranges of $1.28 \, K \leq T \leq 22.8 \, K$ (which is the low-$T$ region, because $\frac{T}{T_\rho} \leq \frac{1}{10}$ (where $T_\rho = 221–228 \, K$ [27, 36])), and of $1.28 \, K \leq T \leq 113 \, K$ (which is within the medium-$T$ range of $\frac{T}{T_\rho} \leq \frac{1}{3}$).

It can be seen (figure 2(c)) that the fit to equation (7) has low quality and the free-fitting parameter $N = 1.99 \pm 0.04$ is very different from the expected value of 5, while the fit to equation...
Figure 1. $\rho(T)$ data for pure elemental copper reported by Teixeira [28] (this $\rho(T)$ dataset is referred to as dataset #269 by Matula [27]). (a), (b) for temperature range $T < T_\theta \lesssim 1\times10^3$ and (c), (d) for temperature range $T_\theta \lesssim 2 \times 10^3$. (a) Fit to equation (7), deduced $N = 4.72 \pm 0.02$, goodness of fit is 0.9998; (b) fit to equation (5); deduced $p = 5.08 \pm 0.03$, $T_\omega = 306 \pm 4$ K, goodness of fit is 1.0. (c) Fit to equation (7), deduced $N = 3.39 \pm 0.04$, goodness of fit is 0.997; (d) fit to equation (5); deduced $p = 4.82 \pm 0.02$, $T_\omega = 347 \pm 1$ K, goodness of fit is 1.0. 95% confidence bands are shown by shaded areas.

Figure 2. $\rho(T)$ data for pure elemental silver reported by Teixeira [28] (this $\rho(T)$ dataset is referred to as dataset #103 by Matula [27]). (a), (b) for temperature range $T < T_\theta \lesssim 1\times10^3$ and (c), (d) for temperature range $T_\theta \lesssim 2 \times 10^3$. (a) Fit to equation (7), deduced $N = 4.47 \pm 0.01$, goodness of fit is 0.99997; (b) fit to equation (5), deduced $p = 4.54 \pm 0.02$, $T_\omega = 251 \pm 8$ K, goodness of fit is 1.0. (c) Fit to equation (7), deduced $N = 1.99 \pm 0.04$, goodness of fit is 0.994; (d) fit to equation (5); deduced $p = 5.02 \pm 0.03$, $T_\omega = 228.8 \pm 0.5$ K, goodness of fit is 1.0. 95% confidence bands are shown by shaded areas.
Layered transition metal dichalcogenides (TMDCs) possess various unusual electronic properties, including the emergence of superconductivity [37, 38] from the inherent instabilities of the stripe charge ordered phase [38, 39]. The generalized BG equation (equation (5)) has never applied for the analysis of temperature-dependent resistivity in TMDCs and here we performed the analysis for PdTe$_2$. In figure 3 we show $R(T)/R(300 \text{ K})$ data for single crystal PdTe$_2$ at ambient pressure, as recently reported by Yang et al [40]. These authors claimed that their analysis showed an unusual $N = 4$ value for the fit to the power-law function of $R(T)/R(300 \text{ K})$ data. Our fits to the power-law function (equation (7)), shown in figures 3(b) and (e), do not have high quality. To demonstrate that the generalized BG equation (equation (5)) can be a better research tool to analyse temperature-dependent resistivity data in figures 3(b) and (e), we show the data fits to equation (5), where $p$ was fixed to $p = 5$ and in figures 3(c) and (f) the data fits to equation (5), where $p$ was a free-fitting parameter. It is deduced that $T_\theta = 153 – 164 \text{ K}$
and $T_c = 191 - 205$ K varies within 7% despite the fact that the temperature range for the analysed $R(T)/R(300$ K) dataset varies by a factor of two. Free-fitting parameter $p = 3.08 - 3.30$ (figures 3(c) and (f)) also varies in a narrow range and it is definitely well below the $N = 4$ reported by Yang et al [40].

It should be noted that there are two primary requirements for the $R(T)$ dataset to be reliably fitted to equation (5). The first requirement is that $R(T)$ data should be measured at as wide as possible range of $T/T_c$ ratio. Truly, it can be seen in figures 1(b), (d), 2(b), (d) and 3(c), (f) that the uncertainties for the deduced $T_c$ values are dramatically narrower for $R(T)$ datasets taken at a wider temperature range. The origin for this reduction is that equation (5) has the term:

$$
\left( \frac{T}{T_c} \right)^p \int_0^{T_p} \frac{e^x}{(e^x - 1) \cdot (1 - e^{-x})} \cdot dx
$$

and it is obvious that if $R(T)$ data covers a wider $T/T_c$ range, than the full term (i.e. equation (10)) will be more accurately calculated. Also, when the $p$ value is a free-fitting parameter it has some uncertainty and, thus, full integral (equation (10)), where the second free-fitting parameter is $T_c$, can be calculated with wider uncertainty.

Also, there is the second requirement that the $R(T)$ dataset should contain many data points to calculate the same integral (equation (10)) with better accuracy. Both these requirements are not really satisfied for NRTS superconductors, for which $T_c > 1500$ K and $R(T)$ datasets are conventionally measured up to $T = 295$ K only and detailed discussion of this problem for NRTS materials is given below in section 3.6.

3.4. High-entropy alloy (ScZrNb)$_{0.65}$[RhPd]$_{0.35}$

Before equations (5) and (6) will apply to hydrogen-rich superconductors, there is a necessity to show that these equations are applicable for low-temperature superconductors. In figure 4 we applied equations (6) and (8) to fit $R(T)$ data for high-entropy alloy (ScZrNb)$_{0.65}$[RhPd]$_{0.35}$ for which raw experimental $R(T)$ data was reported by Stolze et al [41]. It should be mentioned that recently we fitted this $R(T)$ dataset to equation (5) for fixed $p = 5$ [33], and here we make the $p$ parameter free.

It can be seen (figure 4) that both fitting curves have high quality. However, the fit to the power-law model (equation (8), figure 4(a)) revealed $N = 1.2$, which is remarkably different from the fit to the BG model (equation (6), figure 4(b)) for which the deduced $p = 4.9 \pm 0.4$. This implies that the superconducting state in high-entropy alloy (ScZrNb)$_{0.65}$[RhPd]$_{0.35}$ emerges solely from the electron–phonon interaction. Also, from results presented in figure 4 it can be seen that the power-law function (equation (8)) cannot be accepted as a valid approximation for the integrated form of the BG equation (equations (5) and (6)).

3.5. $\alpha$-Ga phase of highly-compressed boron

Now we turn to highly-compressed superconductors and in figure 5 we showed $R(T)$ data for elemental boron pressurized at $P = 240$ GPa (reported by Eremets et al [42]). Oganov et al [43] showed that elemental boron at pressure above $P = 89$ GPa exists in the so-called $\alpha$-Ga phase and, thus, the superconducting state emerges in this boron phase.

The $R(T)$ fit to equation (8) is shown in figure 5(a), for which deduced $N = 1.32 \pm 0.02$. It should be stressed that the fit to equation (6) can be performed also for fixed $p = 5$ value (figure 5(b)). This fit has a high quality (the goodness of fit is 0.9977) and deduced $T_g = 318 \pm 7$ K is in the expected range.

However, when $p$ was chosen to be a free-fitting parameter, the deduced value $p = 2.3 \pm 0.3$ indicates that the electron–electron interaction is dominant in this superconductor. To explain this unexpected result, one can take into account that elemental boron is situated between metals and insulators in the Mendeleev’s table. Despite the fact that this element exhibits three valence electrons, which, in principle, can lead to metallic conductivity, the electrons are sufficiently localized and at ambient conditions boron is an insulator [43]. Thus, it is not a real surprise that the electron–electron interaction dominates in this material even at high pressure.
There is a need to discuss two issues. The first one is that the power-law approximation (equation (8)) of the BG integral form (equation (6)) cannot be considered a valid substitution because deduced $N = 1.32 \pm 0.02$ (equation (8)) is significantly different from $p = 2.3 \pm 0.3$ (equation (6)). The second issue is that the deduced characteristic temperature, $T_c = 435 \pm 38$ K, has a much wider uncertainty range in comparison with $T_0 = 318 \pm 7$ K. As we already mentioned above, this is because $T_0 > T_c$ and, thus, the $R(T)$ dataset is analysed at an effectively narrower reduced temperature range (which has the upper limit of $\frac{T_c}{T_0}$) in comparison with the case when the same dataset is analysed at a reduced temperature range with the upper limit of $\frac{P}{T_0}$. In addition, the uncertainty of free-fitting parameter $p$ causes the increase in the uncertainty of free-fitting parameter $T_0$, while at fixed $p$ value the uncertainty in calculating the integral in equation (10) is solely dependent on the single parameter $T_0$ (see for details section 3.3).

### 3.6. Im-3m-phase of highly-compressed H$_3$S

Now we turn to the analysis of $R(T)$ data for NRTS H$_3$S for which extended raw datasets are freely available by Mozaffari et al [44]. In their figure 1, Mozaffari et al [44] reported $R(T)$ curves for Im-3m-H$_3$S samples subjected to a pressure of $P = 155$ GPa and of $P = 160$ GPa. In figure 6 we showed $R(T)$ curves and fits to equation (8) for sample 07/2018 compressed at $P = 155$ GPa which was aged for two (09/2018) and four (11/2018) consecutive months.

It should be noted that $R(T)$ curves for this sample have multiple-step transitions, which are shown in figure 7. We fitted the transition for sample 07/2018 to a two-step fitting function in our previous work (figure 9 in [33]). Here, to analyse three $R(T)$ datasets, we cut off $R(T)$ curves at $T_{cut\,off}$ (figure 7) and fitted $R(T > T_{cut\,off})$ to equation (8) (figure 7) and equation (6) (figure 8).

It should be noted that Mozaffari et al [44] fitted the $R(T)$ dataset for sample 07/2018 to the parabolic function:

$$R(T) = R_0 + A \cdot T^2$$  \hspace{1cm} (11)

and based on the fact that the fit has a good quality, the authors [44] came to the conclusion that the charge carrier interaction in H$_3$S is dominated by the strong interaction between electrons and very high energy optical phonons. This mechanism was initially proposed by Capitani et al [45].

However, the fit to equation (11) cannot prove that $R(T)$ has a parabolic dependence because the power-law exponent is already fixed to $N = 2$. In figure 6 we presented fits to equation (8) for samples 07/2018, 09/2018 and 11/2018, where $N$ was a free-fitting parameter. Based on results of fits (figure 6), one can conclude that there is no experimental evidence that $R(T)$ of H$_3$S has parabolic temperature dependence.

To further investigate this issue we return to the work by Capitani et al [45] who fitted $R(T)$ curves for two H$_3$S samples (sample A ($P = 160$ GPa) and sample B ($P = 150$ GPa) [45]) to a model based on the so-called elastic residual scattering rate $\gamma_r$. Capitani et al [45] deduced $\gamma_r = 28$ meV and $\gamma_r = 135$ meV for sample A and sample B respectively, which means that the main fitting parameter is different by a factor of $\sim 5$ for these two samples.

In figure 8 we presented the fits to equation (8) of $R(T)$ datasets for sample A and sample B [45]. Because sample A exhibits a two-step transition the $R(T)$ dataset was cut off (figure 8(a)). From figures 6 and 8 one can conclude that $R(T)$
curves of highly-compressed H$_3$S cannot be characterized by parabolic temperature dependence.

We showed in previous papers [33, 34], that full $R(T)$ transition curves for several NRTS materials can be fitted to the BG equation (equation (6)) where $p = 5$ (fixed). Here we extended this list by H$_3$S samples of 07/2018, 09/2018 and 11/2018 [44] (all compressed at $P = 155$ GPa, figure 9) and samples 07/2018 (P = 160 GPa [44]), sample A (P = 160 GPa [45]) and sample B (P = 150 GPa [44]) (which are shown in figure 10). In table 1 we collected the main deduced parameters for $Im$-3m-H$_3$S samples for which $R(T)$ data is available to date. It can be seen that the Debye temperature, $T_\theta$, varies within a narrow range for all H$_3$S samples.

It can be seen in figures 10(b) and (c) that sample A and sample B exhibit very close $T_\theta$ values, which correlate with their close $T_c^{onset}$ values. However, this result contrasts with a model proposed by Capitani et al [45] who assumed that the
primary parameter of their model, which is the elastic residual scattering rate $\gamma_r$, should be $\gamma_r = 28$ meV and $\gamma_r = 135$ meV for sample A and sample B respectively. We argue herein that the advanced BG model (equation (6)) is sufficient to deduce the primary parameter of NRTS (which is $T_\theta$ or $T_\omega$) and to fit $R(T)$ curves for these materials [33, 34].

It can be seen in figures 9, 10 and table 1 that fits to equation (6) at fixed $p = 5$ exhibit high qualities and deduced $T_\theta$ are in agreement with respective characteristic temperatures for the phonon spectrum reported by first-principles calculations for this compound [46–52]. However, in the case when $p$ is a free-fitting parameter, its value is reduced to close proximity to the electron–electron interaction value of $p = 2.44 \pm 0.03$.

However, only one of six $R(T)$ datasets (i.e. 07/2018) can be fitted to equation (6) (figure 11) when $p$ is a free-fitting parameter. 

We claim herein that the advanced BG model (equation (6)) is sufficient to deduce the primary parameter of NRTS (which is $T_\theta$ or $T_\omega$) and to fit $R(T)$ curves for these materials [33, 34].

Figure 8. Resistance data, $R(T)$, and fits to equation (8) for highly-compressed $Im-3m$-$H_3S$ phase. Raw data reported by (a) Mozaffari et al [44] and (b), (c) Capitani et al [45]. (a) Sample 07/2018 ($P = 160$ GPa), $N = 2.77 \pm 0.04$, goodness of fit is 0.9397; (b) sample A ($P = 160$ GPa), $N = 2.69 \pm 0.09$, goodness of fit is 0.9337; (c) sample B ($P = 150$ GPa), $N = 3.30 \pm 0.09$, goodness of fit is 0.9987. 95% confidence bands are shown by shaded areas.

Figure 9. Resistance data, $R(T)$, and fits to equation (6) (for $p = 5$ (fixed)) for highly-compressed $Im-3m$-$H_3S$ phase (raw data reported by Mozaffari et al [44]). (a) Sample 07/2018, $T_\theta = 1427 \pm 2$ K, goodness of fit is 0.9995; (b) sample 09/2018, $T_\theta = 1560 \pm 5$ K, goodness of fit is 0.9972; (c) sample 09/2018, $T_\theta = 1552 \pm 6$ K, goodness of fit is 0.9936. 95% confidence bands are shown as shaded areas.
parameter. Other fits either diverged, parameter uncertainties were larger than the deduced values, or parameter dependency reached value $\equiv 1$.

We already discussed some issues of this problem in sections 3.2 and 3.3. In addition to the already mentioned issues, we should stress that to be fitted to equations (5) and (6) the $R(T)$ dataset should be measured in a substantially wide temperature range of $T_{\text{onset}} \leq T \leq T_{\omega}$. For example, $R(T)$ datasets considered in sections 3.1 and 3.2 for pure Cu and Ag cover $\frac{1}{360} \leq \frac{T}{T_{\omega}} \leq \frac{1}{2}$, which allows convergence of fits and the deduction of $p$ and $T_{\omega}$ values with small uncertainties.

For superconductors, the lower $\frac{T}{T_{\omega}}$ boundary is naturally limited by the normal part of the $R(T)$ curve:

$$\frac{T_{\text{onset}}}{T_{\omega}} \lesssim \frac{T}{T_{\omega}} \lesssim 0.9997; \ (b) \ \text{sample A (}\Im - 3m\text{, }H_3S\text{ sample)} \text{ for highly-compressed } \text{Im-3m-H}_3\text{S phase. } R(T) \text{ data reported by (a) Mozaffari et al [44] and (b), (c) by Capitani et al [45].}$$

$$\theta_0 = 1.487 \pm 19 \text{ K}, \ T_{\text{c onset}} = 190.4 \pm 0.1 \text{ K}$$

$$T_0 = 1.618 \pm 50 \text{ K}, \ T_{\text{c onset}} = 200.8 \pm 0.1 \text{ K}$$

$$T_0 = 1.764 \pm 61 \text{ K}, \ T_{\text{c onset}} = 194.4 \pm 0.1 \text{ K}$$

Because other $H_3S$ samples in table 1 have higher $T_{\omega}$ it is natural to propose that $T_{\omega}$ for those samples will also be higher, and thus the ratio of $\frac{T_{\text{onset}}}{T_{\omega}}$ will be even lower than for sample 07/2018 (equation (15)). It should be mentioned that the lower boundary for 07/2018 samples is:

$$0.005 \lesssim \frac{T_{\text{onset}}}{T_{\omega}} \lesssim 0.04,$$

and cannot create a significant problem to accurately calculate the $T_{\text{upper exp}}$ in equation (6). For strong-coupling superconductors (including, in accordance with the most widely accepted point of view, all NRTS [46–52, 57]) this lower limit is much higher, and for $H_3S$ this limit is (see table 1):

$$0.11 \lesssim \frac{T_{\text{onset}}}{T_{\omega}}.$$
A crucial point, why the fit still converged, despite a narrow range of available \( R(T) \) data (equation (17)), is that the \( R(T) \) dataset contained more than 12,000 raw resistive data points in the range indicated by equation (17). Thus, it is no surprise that the uncertainty in the deduced value of \( T_s \) (figure 11(c)) is much larger than the uncertainty for \( T_0 \) (figure 11(b), \( p = 5 \)), because the latter was deduced at a nearly twice wider reduced temperature range:

\[
0.138 \lesssim \frac{T}{T_0} \lesssim 0.205. \tag{18}
\]

In addition, it should be mentioned that each additional free-fitting parameter in any fitting function causes an increase in the uncertainties of other free-fitting parameters.

The primary outcome of this part of the analysis is that \( p \) and \( T_s \) can be deduced for NRTS if experimental \( R(T) \) datasets are measured at a much wider temperature range, which means that the \( R(T) \) dataset should not be limited de facto by the upper boundary of \( T \sim 295 \) K. In other words, \( R(T) \) measurements should be performed at a substantially wider \( f_s \) range, and for the case of \( H_3S \) the upper limit should not be lower than \( T_{upper \exp} \sim 700 \) K or 0.07 \( \lesssim \frac{T}{T_0} \lesssim 0.25 \).

To prove this point of view, we can mention \( \text{PdTe}_2 \) (section 3.3) and \( (\text{ScZnNb})_{0.65}(\text{RhPd})_{0.35} \) (section 3.4) compounds for which raw \( R(T) \) data was measured within a range of \( 0.06 \lesssim \frac{T}{T_0} \lesssim 0.90 \). As a result, both fits were converged and deduced \( p \) and \( T_s \) have small uncertainties (figures 3 and 4).

On the other hand, highly-compressed boron (section 3.5) for which \( R(T) \) data was measured at a temperature range nearly twice as narrow, 0.03 \( \lesssim \frac{T}{T_0} \lesssim 0.55 \), has larger uncertainties for \( p \) and \( T_s \) (figure 5) in comparison with \( \text{PdTe}_2 \) and \( (\text{ScZnNb})_{0.65}(\text{RhPd})_{0.35} \).

Returning now to the sample 07/2018 we can report that the \( \text{Im}-3m \)-phase of \( H_3S \) exhibits \( T_s = 2707 \pm 184 \) K and \( p = 2.44 \pm 0.03 \). The latter value is in close proximity to the electron–electron interaction value of \( p = 2.0 \).

This result can be explained as a manifestation of the covalent-like atomic bonds between ions in the \( \text{Im}-3m \)-phase of \( H_3S \). Detailed discussion of this feature of highly-pressurized sulphur hydride can be found elsewhere [57].

### 3.7. Highly-compressed \( \text{LaH}_x \) (\( p = 150 \) GPa)

Drozdov et al [10] and Somayazulu et al [58] reported on the discovery of NRT superconductivity in highly-compressed lanthanum superhydride. The NRTS phase was identified as the \( \text{Fm}-3m \)-phase of \( \text{La(H,D)}_{10} \). Due to the highest temperature at which \( R(T) \) data was measured for \( \text{La(H,D)} \) compounds is \( T_{upper \exp} \sim 295 \) K and the fact that \( \text{La(H,D)} \) superconductors exhibit \( T_{c,\text{nom}} \sim 240 \) K (which is higher than \( T_{c,\text{nom}} \sim 200 \) K in \( H_3S \)) the problem of narrow \( f_s \) range (discussed in section 3.6) is more severe for \( \text{La(H,D)} \) superconductors than for \( H_3S \).

We fitted available \( R(T) \) datasets for \( \text{La(H,D)} \) superconductors [10] to equation (5) when \( p = 5 \) in our earlier paper [34]. Here we report that there is only one \( R(T) \) dataset (from all available to date \( R(T) \) datasets [10]) of \( \text{LaH}_x \) which can be fitted to equation (6) where \( p \) is a free-fitting parameter. This dataset is for sample 11 [10]. This sample has hydrogen deficiency, \( \text{LaH}_x \) (where \( x \geq 3 \)) and exhibits \( T_c \sim 70 \) K. The fits to equations (6) and (8) are shown in figure 12.

It can be seen in figure 12(a), that the fit to equation (8) returns \( N = 1.45 \pm 0.01 \) which is approximately twice smaller than \( N \)-values deduced for \( H_3S \) samples (figures 6 and 8).

At the same time, free-fitting parameter \( p = 2.27 \pm 0.05 \) is in close proximity to the value of \( p = 2.44 \pm 0.03 \) deduced for \( H_3S \) sample 07/2018 (figure 11(c)). From this one can draw the conclusion that sample 11 of \( \text{LaH}_x \) exhibits dominant electron–electron charge carrier interaction, similarly to highly-compressed boron and \( H_3S \). We can point out that the electron–electron charge carrier interaction in \( \text{LaH}_x \) is more likely also originated from the covalent-like atomic bonds.

It should be mentioned that the deduced \( p \) and \( T_s \) have relative uncertainties within 2%. These high accuracies in deduced free-fitting parameters can be explained by:

(a) \( R(T) \) dataset being measured at a wide temperature range:

\[
0.084 \lesssim \frac{T}{T_s} \lesssim 0.327 \tag{19}
\]

where \( T_{upper \exp} = 285.45 \) K, \( T_{c,\text{nom}} = 73.07 \pm 0.05 \) K, and \( T_s = 873 \pm 13 \) K;

(b) \( R(T) \) dataset having more than 18,500 data points within the temperature range (equation (19)).
3.8. Highly-compressed \textit{F}-43m-PrH$_9$ and \textit{P}6$_3$/mmc-PrH$_9$

Zhou et al [59] reported the discovery of low-$T_c$ superconductivity in highly-compressed praseodymium hydride. All synthesized samples contained a mixture of \textit{F}-43m-PrH$_9$ and \textit{P}6$_3$/mmc-PrH$_9$ phases.

Due to the fact that our results showed (sections 3.1–3.7) that power-law fit (equations (7) and (8)) is not a reliable research tool, in figure 13 we show that only $R(T)$ fits to equation (6) where $p$ is a free-fitting parameter measured at different pressure in the range of ($P = 135$–145 GPa [59]).

In the studied pressure range ($P = 135$–145 GPa [59]), the free-fitted parameter $p$ is varied in the range of...
1.80 \leq p \leq 1.99. This \( p \) range is close to the theoretical value for pure electron–electron interaction, i.e. \( p = 2.0 \).

It should be mentioned that all three \( R(T) \) datasets were measured at a wide temperature range:

\[
0.01 \lesssim \frac{T}{T_\omega} \lesssim 0.25
\]

where \( T_{upper\ exp} = 150 \text{ K}, T_c^{onset} \approx 7 \text{ K}, \) and \( T_\omega \approx 600 \text{ K}, \) and \( R(T), \) and for this reason, deduced \( p \) and \( T_\omega \) for all three samples have relative uncertainties of less than 2%.

3.9. Highly-compressed pseudocubic \( \text{BaH}_{12} \)

Chen et al [60] reported the discovery of low-\( T_c \) superconductivity in highly-compressed barium hydride \( \text{BaH}_{12} \) which exhibits pseudocubic crystalline structure at pressure from 75 to 173 GPa. In figure 14 we analysed \( R(T) \) curves (showed in figure S41(c) [60]) for the sample subjected to pressure \( P = 100 \text{ GPa} \) and two consequent laser annealing/heating cycles. It can be seen (figure 14(a)) that \( R(T) \) can be with a very good quality fitted for fixed \( p = 5 \) value. However, when \( p \) is a free-fitting parameter it decreases to the value close to the pure case for the electron–magnon interaction, i.e. \( p = 3 \).

Deduced \( p \) and \( T_\omega \) have relative uncertainties of less than 3% (figure 14(b)) mainly because the \( R(T) \) dataset was measured at a wide temperature range:

\[
0.02 \lesssim \frac{T}{T_\omega} \lesssim 0.67
\]

where \( T_{upper\ exp} = 280 \text{ K}, T_c^{onset} \approx 8 \text{ K}, \) and \( T_\omega = 415 \text{ K}. \)
4. Discussion

Overall, in all considered highly-compressed superconductors, i.e., α-Ga phase of boron, H$_2$S, LaH$_3$, PrH$_3$, BaH$_2$, and ε-phase of Fe (considered in [22]), we have found essentially the same result that the power-law exponent, $p$, in the generalized BG equation (equation (6)) is well below 5, i.e., well below the value designated for pure electron–phonon charge carrier interaction in conductors.

On the other hand, for two conventional superconductors ReBe$_{22}$ [22] and high-entropy alloy (ScZrNb)$_{0.65}$[RhPd]$_{0.35}$ (figure 4) deduced free-fitting parameter $p$ is practically indistinguishable from 5. This implies that the electron–phonon interaction is the primary mechanism for the emergence of superconductivity in ReBe$_{22}$ and (ScZrNb)$_{0.65}$[RhPd]$_{0.35}$. However, all highly-compressed superconductors exhibit different dominant charge carrier interaction and are a consequence of this non-electron–phonon pairing mechanism in the superconducting state.

It should be mentioned that there are many theoretically possible mechanisms for charge carrier pairing in the condensed matter [11–14], and from the deduced power-law exponent presented herein, it is more likely that the primary mechanism is related to the electron–electron interaction. This finding is in good accord with the empirical finding that all hydrogen-rich superconductors are located [17, 18, 61, 62] in the unconventional superconductors region of the Uemura plot [19, 20].

Taking into account a possible argument that first-principles calculations have predicted reasonably accurately, the superconducting transition temperatures of several (and what is important to stress, not of all) hydrogen-rich superconductors, the discussion can be related to the issue that sufficiently strong electron–phonon interaction can be accommodated as a consequence of primary and very strong electron–electron interaction. Thus, by varying the Coulomb pseudopotential parameter, $\mu^*$, and the electron–phonon coupling constant, $\lambda_{e-ph}$, in equations similar to the ones proposed by McMillan [53], and Allen and Dynes [54–56]:

$$T_c = \left( \frac{1}{1.20} \right) \cdot \left( \frac{\hbar}{k_B} \right) \cdot \omega_n \cdot e^{-\left( \frac{\lambda_{e-ph}}{\lambda_{ph} - \nu n^* (1 + \frac{\lambda_{e-ph}}{\lambda_{ph} - \nu n^*})} \right)} \cdot f_1 \cdot f_2$$  \hspace{1cm} (22)

where $k_B$ is the Boltzmann constant, and $\hbar$ is the reduced Planck constant, $\omega_n$ is logarithmic phonon frequency, and $f_1$ and $f_2$ are correction functions, it is possible to find correlational dependence between a primary mechanism (which is non-electron–phonon) with the associated electron–phonon interaction.

There is a necessity to discuss the applicability of the high-temperature resistance saturation model [63, 64] to the NRST superconductors. The model [63, 64] has been applied to describe the resistivity of many metallic alloys when the electron mean-free path, $l$, is of the order of the interatomic spacing, $a$, in the compound. It was shown in [17], that H$_2$S exhibits the inequality:

$$a \ll \xi (0) = 2.5 \text{ nm} \ll l (T \sim 200 \text{ K})$$  \hspace{1cm} (23)

Thus, the high-temperature resistance saturation model [63, 64] cannot be applicable for the analysis of H$_2$S samples because the condition of $a \approx l$ not satisfied.

Speaking more broadly, in metallic compounds the condition of $a \approx l$ is satisfied when $T > T_0$. If one takes into account that available-to-date $R(T)$ datasets for NRST is limited by $T_{\text{superco}} \lesssim 0.205$ (equation (18)) and the fact that diamond anvil cells cannot operate at temperatures of $T > T_0 > 1,400 \text{ K}$, it is clear that the high-temperature resistance saturation model [63, 64] is not applicable for the analysis of $R(T)$ curves of NRST superconductors. However, there is a possibility that the model [63, 64] can be applicable for some low-$T_c$ hydrogen-rich superconductors (for instance, BaH$_{12}$ for which deduced $T_0 = 348 \text{ K}$ (section 3.9)) if experimental $R(T)$ curves will be measured.

There is another important issue that has been never been discussed in the literature. This is the possibility that NRTS phases are amorphous. Taking into account that NRTS superconductors are synthesized at extreme heating/cooling rates at high pressure, there is a chance that the NRTS state exhibits in amorphous phases which co-exist with the crystalline phases. The crystalline hydrogen-rich phases can be detected by XRD, however, it is a challenging task to detect amorphous hydrogen-rich phases in samples inside of a diamond anvil cell by XRD.

It is a well-established experimental fact that amorphous tungsten exhibits a superconducting transition temperature of $T_c = 4.8 \text{ K}$ [65], while its crystalline counterpart has $T_c = 0.012 \text{ K}$ [66]. In this regard it is important to mention the recently-discovered high-pressure Ge$_2$Sb$_{2}$Te$_5$ superconductor which exhibits the superconducting state in amorphous phase at nearly ambient pressure in the decompression run [67].

This means that the translational symmetry, the key assumption of the Bardeen–Cooper–Schrieffer (BSC) [3] and Eliashberg’s [4] theories, is not necessarily a condition for superconductivity as a physical phenomenon. The recent experimental discovery of superconductivity in quasicrystals [68], where the translational symmetry does not exist, supports this point of view.

5. Conclusions

In conclusion, in this paper we analysed $R(T)$ data for layered TMDC PdTe$_2$, high-entropy alloy (ScZrNb)$_{0.65}$[RhPd]$_{0.35}$, and highly-compressed α-Ga phase of boron, H$_2$S, LaH$_3$, PrH$_3$ and BaH$_2$ and showed that the dominant charge carrier interaction in the studied highly-compressed superconductors has a non-electron–phonon nature.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

Acknowledgments

The author thanks Dr K Stolze (Leibniz-Institut für Kristallzüchtung) for providing $R(T)$ data for high-entropy...
alloy (ScZrNb)0.65[RhPd]0.35, Dr M I Eremets and Dr V S Minkov (Max-Planck Institut für Chemie, Mainz, Germany) for providing R(T) data for the Im-3m-phase of sulphur hydride and for the Fm-3m-phase of lanthanum hydride, and Dr S Mozafarri and co-authors (National High Magnetic Field Laboratory, Florida State University, USA) for open access magnetoresistance data for the Im-3m-phase of H2S [34]. The author thanks the anonymous referee for proofreading of the manuscript.

The author is grateful for financial support provided by the Ministry of Science and Higher Education 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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