Resistive transition of hydrogen-rich superconductors

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Abstract

Critical temperature, $T_c$, and transition width, $\Delta T_c$, are two primary parameters of the superconducting transition. The latter parameter reflects the superconducting state disturbance originating from the thermodynamic fluctuations, atomic disorder, applied magnetic field, the presence of secondary crystalline phases, applied pressure, etc. Recently, Hirsch and Marsiglio (2021 Phys. Rev. B 103 134505, doi: 10.1103/PhysRevB.103.134505) performed an analysis of the transition width in several near-room-temperature superconductors and reported that the reduced transition width, $\Delta T_c/T_c$, in these materials does not follow the conventional trend of transition width broadening in applied magnetic field observed in low- and high-$T_c$ superconductors. Here, we present a thorough mathematical analysis of the magnetoresistive data, $R(T, B)$, for the high-entropy alloy (ScZrNb)$_{0.65}$[RhPd]$_{0.35}$ and hydrogen-rich superconductors of Im-3m-H$_3$S, C2/m-LaH$_{10}$ and P6$_3$/mmc-CeH$_6$. We found that the reduced transition width, $\Delta T_c/T_c$, in these materials follows a conventional broadening trend in applied magnetic field.

Keywords: resistive transition, hydrogen-rich superconductors, high-entropy alloys, Debye temperature

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

1. Introduction

In 1970, Satterthwaite and Toepke [1] formulated a conceptual idea, which became mainstream research in superconductivity 45 years later: ‘...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. For high concentrations of hydrogen in 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.’ In 2004, Ashcroft [3] proposed rigorous mathematical routine for this idea.

Satterthwaite and Toepke [1] also showed that their conceptual idea can be proved in experiment. By 1970, it had already been known (mainly by extended experimental work performed at Bell Telephone Laboratories) that conventional metallic hydrides/deuterides VH$_{0.5}$ [4], NbH$_x$ ($x = 0.88, 0.99$), NbD$_y$ ($y = 0.11, 0.13, 0.79, 0.80$) [5], ZrH$_2$ [6], TiH$_2$ [6], and, remarkably, LaH$_{2.45}$ [7] and dihydride of thorium, ThH$_2$ [6], are not superconductors. Thus, Satterthwaite and Toepke [1] searched for metallic superhydrides (or, in their words, ‘high hydrides’) and they found that thorium superhydride, Th$_4$H$_{15}$, and its isotopic counterpart Th$_4$D$_{15}$, are both superconductors with the transition temperature of $T_c = 8.0–8.3$ K. In addition, they found that a prominent isotope effect in Th$_4$H$_{15}$ versus Th$_4$D$_{15}$ phases was not observed [1], which indicates a departure from the expected electron–phonon-mediated mechanism of superconductivity [8, 9]. Wang et al [10] reported on the discovery of a low-$T_c$ phase of Th$_4$H$_{15}$, which exhibits $T_c \approx 6$ K.

It took several decades (from the pivotal prediction by Satterthwaite and Toepke [1] for Drozdov et al [11] to show that superhydrides can be synthesized at high pressure ($P > 100$ GPa) and high temperature ($T > 1000$ K)
inside a diamond anvil cell (DAC). To date, more than a dozen superhydride phases, including ones exhibiting near-room-temperature superconductivity (NRTS), have been synthesized from Pr-H [12], P-H [13], Pt-H [14], Sn-H [15], Ce-H [16], Th-H [17], S-(H, D) [11, 18–22], Y-H [23, 24], La-(H, D) [25–29] and Ba-H [30] binary systems. Recently, Semenok et al. [31] reported on the first observation of superconductivity in a La-Y-H ternary system.

The primary experimental technique to study NRTS is magnetoresistance measurement, \( R(T, B) \). In many reports, this measurement is accompanied or initiated by first-principles calculations [32–40]. The latter is a modern research tool, which uses computing power to predict thermodynamically stable phases, density of states, phonon spectra, superconducting transition temperatures and some other superconducting parameters (such as the superconducting gap energy, coherence length, Ginzburg–Landau parameter, critical fields, etc.).

However, some features of the superconducting transition, and in particular the transition width, \( \Delta T_c \), remain to be revealed by experimental data analysis only. In this regard, we mention a recent report by Hirsch and Marsiglio [41] who analyzed \( R(T, B) \) curves in NRTS materials and found that the reduced transition width, \( \frac{\Delta T_c}{T_c} \), is independent of an applied magnetic field within the range of 0 \( \leq \frac{B_{app}}{B_{c1}(0)} \leq 0.15 \), where \( B_{c2}(0) \) is the ground-state upper critical field. This dependence is different to the one in Nb3Sn [42], K3Ca0 [43], NbN [44], MgB2 [45], YBa2CuO7−δ [46], BaFe2−xRxSx (x \( =0.71 \)) [47], La-doped CaFe2As2 [48] and β-phase Mo1−xRex [49], where \( \frac{\Delta T_c}{T_c} \) broadens on the increase in \( \frac{B_{app}}{B_{c1}(0)} \) [42–49].

Here, we report the results of a thorough analysis of \( R(B,T) \) data for high-entropy alloy (ScZrNb)0.65[RhPd]0.35 and hydrogen-rich compounds of Im-3m-H3S, C2/m-LaH10 and P63/mmc- CeH6. To perform the analysis, we propose a new model to fit the experimental \( R(T, B) \) data, which was also successfully applied for \( R(T, B = 0) \) data of highly compressed elemental sulphur. We find that the dependence of \( \Delta T_c(T) \) versus \( \frac{B_{app}}{B_{c1}(0)} \) (in ScZrNb)0.65[RhPd]0.35, Im-3m-H3S, C2/m-LaH10 and P63/mmc-CeH6 follows a conventional trend of resistive transition broadening with the increase in applied magnetic field.

Experimental \( R(T) \) data for the R3m-phase of sulphur hydride and Im-3m-phase of sulphur deuteride were kindly provided by Dr M Einaga (Osaka University, Japan), \( R(T) \) data for the Fm-3m-phase of lanthanum hydride were kindly provided by Dr M I Erements and Dr V S Minkov (Max-Planck Institut für Chemie, Mainz, Germany) and \( R(T, B) \) data for the Im-3m-phase of H3S were kindly placed [50] as an open data set by Dr S Mozafarri and co-authors (National High Magnetic Field Laboratory, Florida State University, USA).

### 2. Model description

Here, we propose a single equation, which describes the full \( R(T, B) \) curve, including the normal state part, which is well above the onset of the resistive transition, \( T \gg T_c^{\text{onset}} \), the exact temperature at which the transition starts, i.e. \( T_c^{\text{onset}} \) and the transition part, \( T \leq T_c^{\text{onset}} \). To the best of our knowledge, this equation is not known, because existing models describe either the resistive part of \( R(T, B) \) curves [51–56] or \( R(T, B) \) near the \( T_c^{\text{onset}} \) [41, 57–59].

Our model is built on recent results [55, 60], revealing that the normal part of \( R(T, B = 0) \) curves for a range of NRTS can be fitted to the Bloch–Grüneisen (BG) equation [56]:

\[
R(T, B = 0) = R_0 + A \cdot \left( \frac{T}{T_0} \right)^5 \cdot \int_0^{\frac{T_0}{T}} \frac{x^5}{(e^x - 1) \cdot (1 - e^{-x})} \, dx,
\]

where \( R_0, A \) and \( T_0 \) are free-fitting parameters and the latter is the Debye temperature. The deduced Debye temperature, \( T_0 \), and the observed transition temperature, \( T_c \), are linked through the McMillan equation [61], which can be represented in the following advanced form [55]:

\[
T_c = \left( \frac{1}{1.45} \right) \cdot T_0 \cdot e^{\frac{1.04 \cdot (1 + \lambda_e \cdot ph)}{\lambda_e \cdot ph - n^* \cdot (1 + 0.62 \cdot \lambda_e \cdot ph)}} \cdot f_1 \cdot f_2,
\]

\[
f_1 = \left( 1 + \frac{\lambda_e \cdot ph}{2.46 \cdot (1 + 3.8 \cdot \mu^*)} \right)^{5/2} \cdot 1/3,
\]

\[
f_2^* = 1 + (0.0241 - 0.0735 \cdot \mu^*) \cdot \lambda_e \cdot ph,
\]

where \( \mu^* \) is the Coulomb pseudopotential parameter (ranging from \( \mu^* = 0.10–0.16 \)) and \( \lambda_e \cdot ph \) is the electron–phonon coupling constant.

The system of equations (2)–(4) has a unique solution in respect of \( \lambda_e \cdot ph \), if \( T_0, T_c \) and \( \mu^* \) are known. For the latter, in most cases, the mean value of \( \mu^* = 0.13 \) can be a good approximation, if \( \mu^* \) was not computed for the given material by first principles calculations. As a result, the electron–phonon coupling constant, \( \lambda_e \cdot ph \), was found by manual calculations due to the substitution of relevant values in equations (2)–(4).

There is need for two clarifications associated with this approach (equations (1)–(4)):

(a) the absence of the criterion to define a lower temperature limit for which the \( R(T, B = 0) \) data set should be fitted to equation (1);
(b) the absence of the criterion to define the transition temperature, \( T_c \), from the curve \( R(T, B = 0) \), which can be used to calculate the electron–phonon coupling constant, \( \lambda_e \cdot ph \), by the use of equations (2)–(4).

The latter problem has general implication, because it is equally applied to the results of first-principles calculations, where \( T_c \) is one of the outcome parameters. However, there is no clarity as to which point in the experimentally recorded \( R(T) \) curve corresponds to the computed \( T_c \), because it can represent the temperature at the onset of the transition, \( T_c^{\text{onset}} \), or zero resistance point, \( T_{c, \text{zero}} \), or any temperature between
these two experimental values, because $T_c$ can be defined by any ratio in the range:

$$0 \leq \left( \frac{R(T)}{R(T_c^{\text{onset}})} \right) \leq 1. \quad (5)$$

(a detailed discussion of the problem is found elsewhere [55]).

Thus, the task is to find a function, which reasonably well approximates the resistive transition, $R(T)$, and simultaneously self-stitches with the BG function (equation (1)) at the onset of the superconducting transition, $T_c^{\text{onset}}$.

By experimenting with several functions, which potentially can approximate the resistive transition and smoothly stitch the BG function, we report herein the result for a function which is similar, but not exact for a function proposed by Tinkham [59]:

$$R(T, B) = \frac{R_{\text{norm}}}{I_0 \left( \frac{C \cdot \left( 1 - \frac{T}{T_c} \right)^{3/2}}{2 \cdot B_{\text{appl}}} \right)^2} \quad (6)$$

$$R(T, B) = \frac{R_{\text{norm}}}{I_0 \left( \frac{D \cdot \left( 1 - \frac{T}{T_c} \right)^{3/2}}{2 \cdot B_{\text{appl}} / B_{c2}(0)} \right)^2} \quad (7)$$

where $I_0(x)$ is the zero-order modified Bessel function of the first kind and $C$ is a free-fitting parameter of Tesla units. This function was recently modified by Hirsch and Marsiglio [41]:

where $D$ is a dimensionless free-fitting parameter.

It should be stressed that there are several unavoidable problems associated with equations (6) and (7). First of all, we can mention that a pivotal resistance curve $R(T, B_{\text{appl}} = 0)$ cannot be fitted to these equations, because division by zero is prohibited. Also, as this was stated by Tinkham [59], $T_c$ in equation (6) (and in equation (7)) is independent of the applied magnetic field, which is (from his point of view [59]) an unphysical assumption of the model.

Here, we propose a simpler function:

$$R(T, B) = \frac{R_{\text{norm}}}{I_0 \left( F \times \left( 1 - \frac{T}{T_c} \right)^{3/2} \right)^2} \quad (8)$$

which has three free-fitting parameters, $R_{\text{norm}}$, $T_c$ and $F$. The primary rationale to use equation (8) is that multiplicative pre-factors:

$$\frac{1}{2 \cdot B_{\text{appl}}} \quad \text{in equation (6)}, \quad (9)$$

and

$$\frac{1}{2 \cdot B_{\text{appl}} / B_{c2}(0)} \quad \text{in equation (7)}, \quad (10)$$

only renormalize the value of free-fitting parameters $C$ and $D$ in equations (6) and (7), respectively. As a result, equations (6)–(8) provide essentially the same fits for the $R(T, B \neq 0)$ data sets. However, equation (8) does not diverge at $B \rightarrow 0$.

We also need to stress that neither Tinkham [59] nor Hirsch and Marsiglio [41] propose any physical interpretation for the parameters $C$ and $D$ in their equations (equations (6) and (7), respectively), and thus equation (8) can be derived following the same approach as equations (6) and (7).

As a result, the full equation, which we propose to fit $R(T, B)$ curves in materials where charge carrier scattering on phonons is the dominant dissipation mechanism in the normal state, can be expressed in the following form:

$$R(T, B) = R_0 + k \cdot T + \theta \left( T_c^{\text{onset}} - T \right) \cdot \frac{R_{\text{norm}}}{I_0 \left( F \times \left( 1 - \frac{T}{T_c^{\text{onset}}} \right)^{3/2} \right)^2} + \theta \left( T - T_c^{\text{onset}} \right) \cdot \frac{R_{\text{norm}}}{I_0 \left( F \times \left( 1 - \frac{T}{T_c} \right)^{3/2} \right)^2} \cdot \left( \frac{T}{T_0} \right)^5 \times \int_0^{\frac{T_a}{T_0}} \frac{x^5}{(e^x - 1) \cdot (1 - e^{-x})} \cdot dx - \left( \frac{T_c}{T_0} \right)^5 \times \int_0^{\frac{T_a}{T_0}} \frac{x^5}{(e^x - 1) \cdot (1 - e^{-x})} \cdot dx \right) \quad (11)$$

where $\theta(x)$ is the Heaviside step function, while $R_0$ and $k$ are free-fitting parameters, which accommodate possible experimental onsets/uncertainties of the electronic measurement system, particular electrode configurations in DAC and metallic weak links in the sample.

It should be noted that in order to be reliably fitted to equation (11), the normal part of the $R(T, B)$ curve should be
measured in a reasonably wide temperature range, and, thus, if
this is not the case (i.e. the measurements perform within a nar-
row temperature range above \( T_c^{\text{onset}} \)), then a simpler equation
can be used:

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

(12)

where \( k \) is a free-fitting parameter describing the slope of the
normal part of the \( R(T, B) \) curve.

If the studied sample does not have metallic weak links,
then equation (12) is reduced to an equation with the same
number of free-fitting parameters (which are \( T_c^{\text{onset}}, R_{\text{norm}},
F, k \)) as the standard fitting equation for the pinning force
density [62–65]:

\[
F_p(B_{\text{appl}}) = F_{p, \text{max}} \cdot \left( \frac{B_{\text{appl}}}{B_{c2}} \right)^p \cdot \left( 1 - \frac{B_{\text{appl}}}{B_{c2}} \right)^q,
\]

(13)

where \( F_{p, \text{max}}, B_{c2}, p \) and \( q \) are free-fitting parameters. For these
samples, full equation (11) exhibits five free-fitting parameters,
where the additional parameter is the Debye temperature,
\( T_\theta \), which is a fundamental characteristic of a superconductor,
and, thus, it cannot be asserted that \( T_\theta \) is an unnecessary
parameter.

To fit the \( R(T, B) \) data sets, we use a non-linear data fitting
tool from the ORIGIN package (version 2017). In all plots, we
show 95% confidence bands for the fitted curves, which calcu-
lated by the ORIGIN package and these bands represent 2\( \sigma \)
uncertainty for the fitted curve calculated from 2\( \sigma \) uncertainties
of all free-fitting parameters at each point. Rigorous mathema-
tical definition of 95% confidence bands is found else-
where [66].

From the converged fit, \( T_c \) can be defined by any chosen
\( R(T) \mid R(T_c^{\text{onset}}) \) criterion (equation (5)). In this work, we use the value of
\( T_c^{0.03} \):

\[
\frac{R(T)}{R(T_c^{\text{onset}})} = 0.03,
\]

(14)

which was used to calculate the electron–phonon coupling constant,
\( \lambda_{e-\text{ph}} \) (equations (2)–(4)). This value, \( T_c^{0.03} \), was also
used to deduce the width of the resistive transition, \( \Delta T_c \):

\[
\frac{\Delta T_c}{T_c} = \frac{T_c^{\text{onset}} - T_c^{0.03}}{T_c^{\text{onset}}},
\]

(15)

The reason for choosing \( \frac{R(T)}{R(T_c^{\text{onset}})} = 0.03 \) for the analysis
of experimental \( R(T, B) \) data of NRTS originates from the
requirement that this ratio should be as small as possible
(extended discussion of this important issue is given in [55])
on the one hand. On the other hand, this ratio should be well
above the level of noise of experimental \( R(T, B) \) as the anal-
ysis will be performed for real measured \( R(T, B) \) data and not
for extrapolated tails of given mathematical functions. Based
on the available \( R(T, B) \), the ratio of \( \frac{R(T)}{R(T_c^{\text{onset}})} = 0.03 \) (equation
(13)) fulfills both requirements.

3. Results

3.1. Highly compressed superconductors in zero magnetic
field

3.1.1. Elemental sulphur. Before the model (equation (11))
will be applied to NRTS materials, we demonstrate its applic-
ability to highly compressed elemental sulphur (figure 1).
Experimental \( \frac{R(T)}{R(T_c^{\text{onset}})} \) curves were reported by Shimizu et al
[67] (in their figure 10 [67]).

The model perfectly found \( T_c^{\text{onset}} \) for all \( \frac{R(T)}{R(T_c^{\text{onset}})} \) data sets
(figure 1). All fits converged with excellent quality (where the
goodness of fit, \( R \), varies within a narrow range of 0.997–0.998
for all fits in figure 1). The free-fitting parameters of the model
\( T_\theta \), as well as the calculated parameters \( T_c^{0.03} \) and \( \lambda_{e-\text{ph}} \) (for
which we used equations (2)–(4) and (14)) also smoothly vary
within narrow ranges (figure 1). The superconducting transition
width, \( \frac{\Delta T_c}{T_c} \), has a trend to increase versus the increase in
applied pressure.

3.1.2. \( R3m \)-phase of sulphur hydride. Einaga et al [18]
studied the pressure dependence of the transition tempera-
ture, \( T_c(P) \), in the \( R3m \)-phase of highly compressed sulphur
hydride. In figure 2, we show the raw \( R(T, P = 133 \text{ GPa}) \)
curve for this phase (reported in figure 3(a) [18]), and data fit to
equation (11).

The deduced \( \lambda_{e-\text{ph}} (P = 133 \text{ GPa}) = 2.3 \pm 0.1 \) is
very close to the first principles calculation value of
\( \lambda_{e-\text{ph}} (P = 130 \text{ GPa}) = 2.07 \) reported by Duan et al [68]
in their first pivotal paper on the highly compressed H–S sys-
tem. The superconducting transition has a moderate width of
\( \Delta T_c/T_c \approx 0.095 \).

3.1.3. \( \text{Im}-3m \)-phase of sulphur deuteride. Einaga et al [18]
also studied the pressure dependence of the transition tempera-
ture, \( T_c(P) \), in the \( \text{Im}-3m \)-phase of sulphur deuteride. In figure
3, we show the raw \( R(T, P = 190 \text{ GPa}) \) data for this
phase (reported in figure 3(b) [18]) and data fit to equation
(11).

The fit in figure 3 is a compelling example of the model
validity, because it covers a wide temperature range of \( 14 \text{ K} \leq
T \leq 290 \text{ K} \) with remarkably high quality of 0.99994.

The deduced \( \lambda_{e-\text{ph}} = 1.85 \) is also within the expected
range reported for this value by first principles calculations,
\( \lambda_{e-\text{ph}} = 1.86 \) [36], and the superconducting transition has a
moderate width of \( \Delta T_c/T_c = 0.14 \).

3.1.4. Hydrogen-deficient lanthanum hydride \( \text{LaH}_x \) (\( x > 3 \)).
Somayazulu et al [25] and Drozdov et al [26] independently
reported on the discovery of NRTS in several phases of the
La–H system, from which the highest transition temperature of $T_c = 245 - 280$ K was observed in the $Fm-3m$-phase of LaH$_{10}$ [26]. The transition temperature depends on the hydrogen stoichiometry and the $R(T)$ curve for a highly hydrogen-deficient sample (sample 11 [26]) is shown in figure 4 together with the $R(T)$ fit to equation (11). The quality of the fit is excellent and the sample has a moderately wide superconducting transition width of $\frac{\Delta T}{T_c} = 0.12$. The deduced electron–phonon coupling constant is $\lambda_{e-ph} = 1.68$ ($P = 150$ GPa, $T_c = 73$ K).

It should be noted that the sample (sample 11, LaH$_x$ ($x > 3$), $P = 150$ GPa [26]) has a complex XRD pattern and the dominant phase in this sample is unknown [26]. This is the reason a direct comparison of the deduced $\lambda_{e-ph}$ value with its contemplated calculated for stoichiometric hydrogen-rich phases of LaH$_{10}$ [32, 69] cannot be made at the quantitative level, because stoichiometric
phases have $T_c > 240$ K versus $T_c = 73$ K for sample 11 (figure 4). However, there is a possibility for qualitative comparison. A lower limit for the electron–phonon coupling constant was reported by Durajski and Szczesniak [70], who calculated $T_c = 22.5$ K and $\lambda_{\text{e-ph}} = 0.845$ for LaH$_3$ compressed at $P = 11$ GPa. The same research group reported $T_c = 215$ K and $\lambda_{\text{e-ph}} = 2.18$ for LaH$_{10}$ compressed at $P = 150$ GPa [69], which can be assumed to be the upper limiting values. Thus, as deduced herein, $T_c = 73$ K and $\lambda_{\text{e-ph}} = 1.68$ for LaH$_x$ ($x > 3$) compressed at $P = 150$ GPa are in between the and upper limits [69, 70].

3.15. Fm-3m-phase of lanthanum hydride. Drozdov et al [26] also reported $R(T)$ data for the stoichiometric $Fm$-3m-phase of LaH$_{10}$ (sample 1 [26]), which is shown in figure 5. Due to the normal part of the $R(T)$ curve being measured in a narrow temperature range, the fit was performed to equation (12) (figure 5). Despite a high transition temperature, $T_{c,0.03} = 236$ K, this sample has a narrow transition width of $\Delta T_c = 0.056$.

3.2. Superconductors in applied magnetic field

3.2.1. High-entropy alloy (ScZrNb)$_{0.66}$[RhPd]$_{0.35}$ in applied magnetic field. In order to extend the dependence of $\Delta T_c$ versus $B_{\text{ext}}/B_{\text{nom}}$ [41] for a wider class of materials, we applied our model to $R(T, B)$ measured in high-entropy alloy (ScZrNb)$_{0.66}$[RhPd]$_{0.35}$ [71]. Details of the experiment have been reported elsewhere [71] and here in figure 6 we show the $R(T, B = 0)$ data fit to equation (11). The fit is excellent and the deduced $\lambda_{\text{e-ph}} = 1.03$ is close to the recently reported $\lambda_{\text{e-ph}} = 1.10$ for another high-entropy alloy (TaNb)$_{0.67}$[HFzTt]$_{0.33}$ [72].

As the $R(T, B)$ measurements were performed in a narrower temperature range of $1.7$ K $\leq T \leq 12$ K, we fit these data sets to equation (12). All fits are of excellent quality (with goodness of fit $> 0.99990$) and some of these fits are shown in figure 7. The resultant plot of $\Delta T_c$ versus $B_{\text{ext}}/B_{\text{nom}}$, with $B_{\text{c2}}(0) = 10.7$ T [71], is shown in figure 8, where the conventional trend of the transition width broadening in applied magnetic field is apparent.
Figure 7. $R(T, B)$ data for high-entropy alloy $(\text{ScZrNb})_{0.65}[(\text{RhPd})_{0.35}$ and data fit to equation (12). Goodness of fit for all fits is better than 0.99990. 95% confidence bars are narrower than the thickness of the fitting curve.

3.2.2. $Im$-3m-phase of $H_3S$ in applied magnetic field. Mozaffari et al [50] reported high-field magnetoresistance measurements, $R(T, B)$, for the $Im$-3m-phase of $H_3S$ compressed at $P = 155$ and 160 GPa. Here, we analyze data for a sample compressed at $P = 155$ GPa, which exhibits two resistive transitions at zero applied field (figure 9). The $R(T)$ data were fitted to a two-step function:
resistance (mΩ/g58)

$20 \quad 50 \quad 60$ 185 190 195 200 205 210 215

$Sulphur hydride (P = 155 GPa)$

Figure 8. Magnetic field dependence on the reduced superconducting transition width, $\Delta T_c/T_c$, for high-entropy alloy (ScZrNb)$_{0.65}$[RhPd]$_{0.35}$.

$R(T, B) = R_0 + \theta \left( (T_{c1}^{onset} - T) \right) \times \left( \frac{\Delta T_c}{T_c} \right) \times \left( \frac{R_{norm,1}}{I_0 \left( F_1 \cdot \left( 1 - \frac{T}{T_{c1}^{onset}} \right)^{3/2} \right)^2} \right) + \theta \left( (T_{c2}^{onset} - T) \right) \times \left( \frac{R_{norm,2}}{I_0 \left( F_2 \cdot \left( 1 - \frac{T}{T_{c2}^{onset}} \right)^{3/2} \right)^2} \right) + \theta \left( (T - T_{c2}^{onset}) \right) \times \left( R_{norm,1} + R_{norm,2} + k \cdot T \right).$

where subscripts 1 and 2 indicate the transition.

The fit reveals that both transitions have very narrow transition widths of $\frac{\Delta T_{c1}}{T_c} = 0.009$ and $\frac{\Delta T_{c2}}{T_c} = 0.007$.

In figure 10, we fit to equation (12) (where $k$ was fixed to zero) four $R(T, B)$ data sets of the Im-3m-phase of H$_3$S ($P = 155$ GPa) reported by Mozaffari et al [50] in their supplementary figures 1 and 2. An important feature of the result is that the fits of the $R(T, B)$ data sets for the Im-3m-phase of H$_3$S ($P = 155$ GPa) were processed by the same mathematical routine and the same criterion was applied to deduce $\frac{\Delta T_c}{T_c}$ versus $\frac{R_{onset}}{R_{c(0)}}$ dependence. Based on this, the result is not distorted by any variation, which could have appeared if manual data processing had been implemented.

It can be seen in figure 11 that $\frac{\Delta T_c}{T_c}$ versus $\frac{R_{onset}}{R_{c(0)}}$ (where we adopt $B_{c2}(0) = 88$ T reported by Mozaffari et al [50] for this sample), for the Im-3m-phase of H$_3$S follows the general trend of the transition width broadening with the increase in the magnetic field. For instance, $\frac{\Delta T_c}{T_c}$ versus $\frac{R_{onset}}{R_{c(0)}}$ curve for H$_3$S displays a similar trend to the ones for Nb$_3$Sn and BaFe$_{2−x}$Ru$_x$As$_2$ ($x = 0.71$) (figure 11).

3.2.3. C2/m-phase of LaH$_{10}$ in applied magnetic field.

Sun et al [29] reported on the discovery of new C2/m-phase of LaH$_{10}$ compound, which exhibits zero resistance at $T = 170–185$ K at a pressure range of $P = 120–130$ GPa. Sun et al [29] also reported high-field magnetostriction measurements, $R(T, B)$, for this C2/m-phase compressed at $P = 120$ GPa, which we analyzed in our figure 12.

It can be seen in figure 11 that the $\frac{\Delta T_c}{T_c}$ versus $\frac{R_{onset}}{R_{c(0)}}$ dependence (for which we adopt $B_{c2}(0) = 133.5$ T reported by Sun et al [29] for this phase) follows the general trend of the transition width broadening with increased applied magnetic field.

3.2.4. P6$_3$/mmc-phase of CeH$_6$ in applied magnetic field.

Chen et al [16] synthesized a new high-temperature superconducting P6$_3$/mmc-phase of CeH$_6$, which exhibits $T_c = 45–85$ K at relatively low pressure of $P = 88–140$ GPa. Chen et al [16] reported extensive magnetostrictive studies of this new phase, from which we analyzed Sample Cell #1H compressed at $P = 88$ GPa. The raw $R(T, B)$ data are reported in figure S7 [16].

All fits to equation (12) for this sample are shown in figure 13 and all fits have excellent quality. The deduced $\frac{\Delta T_c}{T_c}$ versus $\frac{R_{onset}}{R_{c(0)}}$ dependence (for which we adopt $B_{c2}(0) = 33$ T reported by Chen et al [16] for this sample), is shown in figure 11, and the usual trend of the transition with broadening versus the increase in applied magnetic field is obvious (figure 11).
Figure 10. \(R(T, B)\) data for the highly compressed Im-3m-phase of \(\text{H}_3\text{S}\) \((P = 155 \text{ GPa})\) and data fit to equation (12) \((k\) was fixed to zero). Green balls show \(T_{c, 0.03}\). 95% confidence bars are narrower than the thickness of the fitting curve; goodness of fit for all curves is better than 0.9991.

Figure 11. Field dependence of the superconducting transition width, \(\Delta T_c/T_c\), versus reduced applied field, \(B_{\text{appl}}/B_c^2(0)\), for materials processed in this paper (depicted in bold) and several representative materials of major superconducting families (data for these materials were adopted from [41]).

4. Discussion

We show above that hydrogen-rich superconductors exhibit the usual trend of broadening of the resistive transition width, \(\Delta T_c/T_c\), on the increase in applied magnetic field, \(B_{\text{appl}}/B_c^2(0)\). This result is in contrast with a recent report by Hirsch and Marsiglio [41], who found that the transition width, \(\Delta T_c/T_c\), remains constant (or is even suppressed in the case of \(\text{YH}_9\)) on the increase of \(B_{\text{appl}}/B_c^2(0)\).

To explain this apparent contradiction, we need to point out several important issues. First of all, Hirsch and Marsiglio [41] analyzed \(R(T, B)\) data measured at low applied magnetic fields:

\[
\frac{B_{\text{appl}}}{B_c^2(0)} \leq 0.1,
\]

(17)

where \(B_c^2(0)\) was obtained by extrapolation of \(B_c^2(T)\) data also measured in a narrow temperature range near \(T_c\):

\[
0.9 \leq \frac{T}{T_c} \leq 1.0,
\]

(18)

which both create reasonable uncertainty in the analysis [41].

However, more important is the fact that, to the best of our knowledge, Hirsch and Marsiglio [41] did not provide a \(\Delta T_c\) definition used for their analysis, and thus, there is no clarity on how \(\Delta T_c\) was deduced from the experimental data.

In addition, the NRTS materials analyzed in [41] were multiphase samples and if these phases have close transition temperatures at \(B = 0\) T (see, for instance, figure 9), then this can lead to an effective broadening of the total resistive transition \(R(T, B = 0)\). However, different superconducting
Figure 12. $R(T, B)$ data for the highly compressed $C2/m$ phase of LaH$_{10}$ ($P = 120$ GPa) and data fit to equation (12) ($k$ and $R_0$ were fixed to zero). Green balls show $T_{c,0.03}$. Red is the fitting curve. 95% confidence bars are shown by a pink shaded area; goodness of fit for all curves is better than 0.998.

Phases are more likely to have reasonably different upper critical fields, $B_{c2}(T)$. Moreover, by applying a magnetic field, $B_{appl}$, the superconducting state in one (or several) phase can be completely suppressed. Thus, at some applied field the superconducting state will remain in one phase, which exhibits the highest upper critical field. As a result, the transport current will be short circuited within the remaining superconducting phase, which will effectively appear as a ‘narrowing’ of the in-field resistive transition, $R(T, B)$. The effect of the extinction of superconducting phases is exhibited at relatively low applied magnetic field. This can be clearly seen in the case of H$_3$S in the supplementary figure 1 of reference [50]. Because secondary phases are eliminated at low applied magnetic fields near $T_c$, at higher applied fields the resistance curve, $R(T, B)$, is solely dependent on a single superconducting phase and the resistive transition appears to be sharper than in the case where several phases contribute to $R(T, B)$. However, at higher applied magnetic field and respectively lower temperatures, conventional resistive transition broadening versus applied magnetic field is expected, because it is related to a single superconducting phase. And this is what we find herein in our report (figure 11), where $R(T, B)$ data measured at 0.1 ≤ $B_{appl}$ ≤ 0.3 in the case of H$_3$S and LaH$_{10}$ were analyzed, and for the single-phase P6$_3$/mmc-CeH$_9$ sample for which measurements were performed in the range of 0.0 ≤ $B_{appl}$ ≤ 0.15. The latter case confirms that the broadening at low applied fields is still valid for hydrogen-rich superconductors if the sample is a single phase.

Our explanation received independent confirmation as a result of a thorough search of the literature. For instance, Wang et al. [73] showed that the transition width, $\Delta T_c$, in SiC-doped polycrystalline MgB$_2$ samples decreases in weak magnetic fields. At some medium fields, $\Delta T_c(B)$ reaches its minimum and starts to linearly increase at higher magnetic fields. This is a compelling example of non-monotonic $\Delta T_c(B)$ in multiphase polycrystalline samples, which were all samples studied in [41]. It should be noted that recently Dogan and Cohen [74] analyzed the $\Delta T_c$ problem in hydrogen-rich superconductors by using a different approach.

It is important to discuss possible limits for the applicability of our basic equations (11) and (12) to describe the resistive transition, $R(T, B)$. From its definition, equation (11) is applicable to materials where charge carriers are predominantly scattered by the lattice vibrations (i.e. phonons). However, for materials where the observed $T_c$ does not solely depend on the electron–phonon interaction (in particular, in cuprates [51, 53, 54], pnictides [75–77] and nickelates [78, 79]), but instead on the thermodynamic fluctuations [58, 80–83], the presence of pseudogap [84, 85], structural disorder [86–89], system dimensionality [90–94] or other mechanisms [95–98] for charge carrier wave function distortions, our equations (11) and (12) might not be a good fitting tool (however, it does not necessarily mean that in some cases these equations will still be a good fitting tool). Based on all the above, alternative $R(T, B)$ models may be developed for other types of materials.
5. Conclusion

In this work, we study the in-field dependence of the reduced resistive transition width, $\Delta T_c / T_c$, in high-entropy alloy (ScZrNb)$_{0.65}$[RhPd]$_{0.35}$ and hydrogen-rich superconductors Im-3m-H$_3$S, C2/m-LaH$_{10}$ and P6$_3$/mmc-CeH$_9$. To perform the analysis, we propose a new function to fit $R(T, B)$ curves and a strict mathematical routine to deduce $\Delta T_c$ from the analysis of $R(T, B)$ curves. As a result, we show that the reduced transition width, $\Delta T_c / T_c$, in (ScZrNb)$_{0.65}$[RhPd]$_{0.35}$, Im-3m-H$_3$S, C2/m-LaH$_{10}$ and P6$_3$/mmc-CeH$_9$ follows a conventional broadening trend with increased applied magnetic field.

Data availability statement

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

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