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Classifying superconductivity in ThH-ThD superhydrides

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Abstract

Satterthwaite and Toepke (1970 *Phys. Rev. Lett.* **25** 741) discovered that Th_4H_{15}-Th_4D_{15} superhydrides exhibit superconductivity and have no isotope effect. The latter is fundamental contradiction with the concept of electron-phonon mediated superconductivity of Bardeen-Cooper-Schrieffer (BCS) theory. Soon after this work, Stritzker and Buckel (1972 *Zeitschrift für Physik A Hadrons and nuclei* 257 1-8) reported that superconductors in PdH_x-PdD_x system exhibit reverse isotope effect. Yussouff *et al* (1995 *Solid State Communications* **94** 549) extended this finding on PdH_x-PdD_x-PdT_x system. Recent interest to hydrogen- and deuterium-rich superconductors is based on the discovery of near-room-temperature superconductivity in highly-compressed H_3S (Drozdov *et al.* 2015 *Nature* **525** 73) and LaH_{10} (Somayazulu *et al* 2019 *Phys. Rev. Lett.* **122** 027001). To date, there is no clarity about isotope effect in H_3S-D_3S system, because thorough examination of available experimental data reported by Drozdov *et al* (2015 *Nature* **525** 73) shows that H_3S-D_3S system perhaps has reverse isotope effect. In attempt to reaffirm/disprove our primary idea that the mechanism for near-room-temperature superconductivity in hydrogen-rich superconductors is not BCS electron-phonon interaction, we analyse the upper critical field data, $B_{c2}(T)$, in Th_4H_{15}-Th_4D_{15} phases (Satterthwaite and Toepke 1970 *Phys. Rev. Lett.* **25** 741) and two recently discovered high-pressure hydrogen-rich phases of ThH_9 and ThH_{10} (Semenok *et al* 2019 *arXiv*:1902.10206). As the result, it is found that all known to date thorium super-hydrides/deuterides are unconventional superconductors which have T_c/T_F ratios within a range of $0.008 < T_c/T_F < 0.120$, where T_c is the superconducting transition temperature and T_F is the Fermi temperature.

I. Introduction

The isotope effect in Bardeen-Cooper-Schrieffer (BCS) theory of superconductivity expressed in the form:

$$T_c \cdot M^{\alpha} = const. \qquad (1)$$

where M is isotope mass, and $\alpha \approx 1/2$ (for weak-coupling limit of BCS theory [1]), is central indispensable feature of electron-phonon mediated superconductivity [1]. This effect was observed in several elemental superconductors, but not in all of them [2]. And, for instance, Geballe *et al* [3] were first who found the absence of the isotope effect in ruthenium (more details can be found elsewhere [2-4]). Later, Satterthwaite and Toepke [5] reported the absence of the isotope effect in Th_4H_{15}-Th_4D_{15} super-hydride/deuteride phases. Soon after [5], Stritzker and Buckel [6] experimentally found that the isotope effect in the palladium-hydrogen-deuterium (PdH_x-PdD_x) system has opposite sign (so called, reverse isotope effect). Yussouff *et al* [7] extended this discovery on palladium-hydrogen-deuterium-tritium system (PdH_x-PdD_x-PdT_x). This reverse isotope effect in PdH_x-PdD_x-PdT_x system is still under wide discussion [8,9]. In regard of considered in this paper ThH-ThD system, detailed studied by Caton and Satterthwaite [10] showed that superconductors in thorium-hydrogen-deuterium (ThH-ThD) system have reverse isotope effect, and Dietrich *et al* [11] showed that transition temperature, T_c, of the Th_4H_{15} phase has linear positive ramp of 0.42 K/GPa versus applied external pressure.

Recent interest to the isotope effect in superconducting compounds based on isotopes of hydrogen is based on experimental discovery of near-room-temperature superconductivity in H_3S-D_3S [12] and LaH_{10} [13]. It should be stressed that to date there is no clarity on the isotope effect in H_3S-D_3S system. Truly, despite a fact that Drozdov *et al* [12] in their Fig. 2(b) showed $R(T)$ curves for H_3S and D_3S which demonstrate much lower T_c for D_3S in compassion with H_3S, through examination of this plot shows that $R(T)$ curves for these two compounds were measured at crucially different pressures of P = 141 GPa and P = 155 GPa. To make direct comparison, $R(T)$ curves for both compounds should be measured at the same pressure. Thoroughly examination of available $R(T)$ curves measured at the same pressure for H_3S and D_3S reveals that H_3S-D_3S system has remarkably large reverse isotope effect.

In Fig. 1 we show in $R(T)$ curves for H_3S and D_3S samples measured at P = 155 GPa. Two $R(T)$ curves for H_3S were taken from Fig. 1(a) and Fig. 3(a) of Drozdov *et al* [12]. And the $R(T)$ curve for D_3S was taken from Fig. 2(b). There is clear experimental result that H_3S-D_3S system has reverse isotope effect. Due to $R(T)$ curves recorded at the same pressure for H_3S and D_3S (which we demonstrate in Fig. 1) are only ones available to date, the only conclusion which can be made is that widely accepted point of view that high-temperature superconductivity in H_3S and D_3S is electron-phonon interaction [14] does not have support in isotope effect experiments.

This finding is additional support to our previous proposal that hydrogen-rich compounds (for instance, PdH_x, H_3S, LaH_{10}) are unconventional superconductors [15,16] and the superconductivity in these compounds is not related to electron-phonon interaction.

We should note that in our consideration we do not include:

1. Highly compressed silane SiH_4 (this was the first discovered by Eremets group highly-compressed hydrogen-rich superconductor with $T_c = 17\ K$ (observed at pressure of P = 96-120 GPa) [17].

2. Covalent hydride phosphine, PH_3, is another hydrogen-rich superconductor in which superconductivity with $T_c \simeq 100\ K$ was discovered at $P \gtrsim 200\ GPa$ [18].
3. $PtH_x (x \cong 1)$ which was recently reported to be superconducting at P = 30 GPa [19].
4. $NbTiH_x$ [20] is another hydrogen-rich superconductor which can be potentially considered.

The reason why these interesting materials are not under our consideration, is that for all of these compounds, fundamental experimental data beyond T_c are unknown, and, thus, we were not able to analyse these materials in our consideration herein.

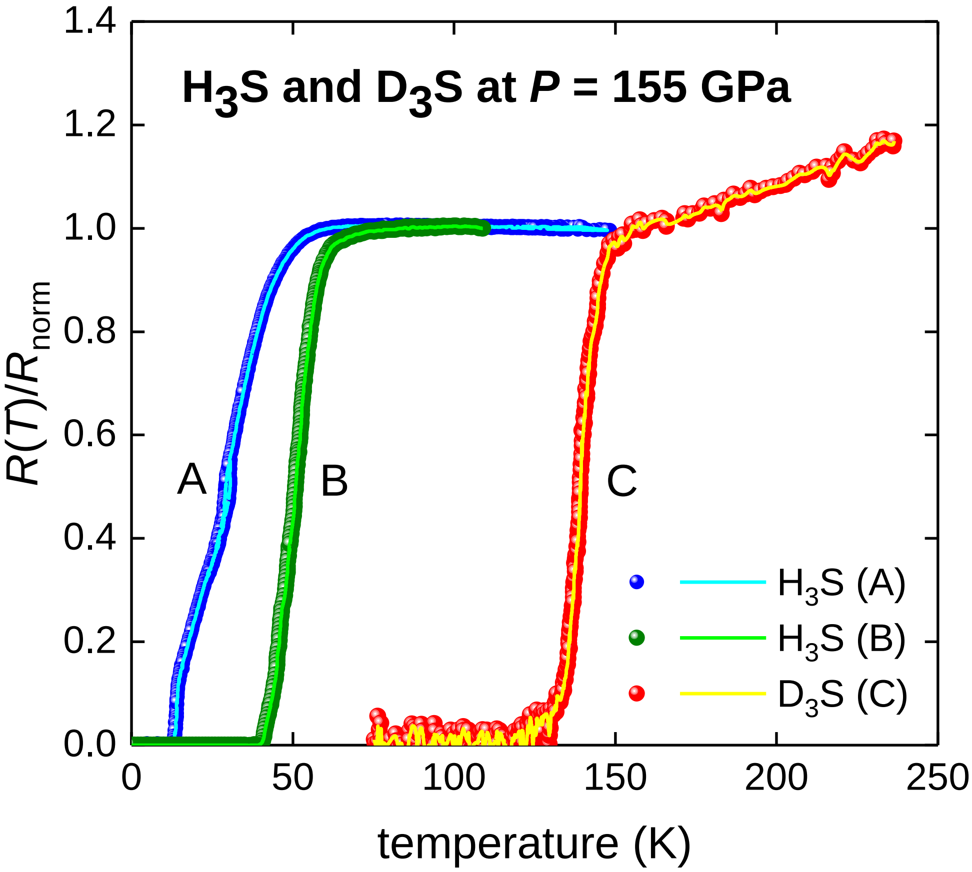

Figure 1. $R(T)$ curves for H_3S (A,B) and D_3S (C) measured at P = 155 GPa. Data for A was taken from Fig. 3(a) [12]; data for B was taken from Fig. 1(a) [12]; data for C was taken from Fig. 2(b) [12].

In result, in this paper we show that all discovered to date hydrogen-rich superconductors for which experimental data beyond T_c are available, i.e. PdH_x, Th_4H_{15}, Th_4D_{15}, ThH_9, ThH_{10},

H_3S and LaH_{10} are unconventional superconductors which lie in the same band in the Uemura plot as all other unconventional superconductors (particularly heavy fermions, fullerenes, pnictides, and cuprates).

It should be stressed, that in some scenarios, Th_4H_{15} and Th_4D_{15} are located in closed proximity to Bose-Einstein condensate (BEC) line in the Uemura plot.

II. The upper critical field models

Ground state upper critical field, $B_{c2}(0)$, in the Ginzburg-Landau theory [21] is given by:

$$B_{c2}\left(\frac{T}{T_c}=0\right)=\frac{\phi_0}{2\cdot\pi\cdot\xi^2(0)}, \qquad (1)$$

where $\phi_0=2.068\cdot 10^{-15}$ Wb is magnetic flux quantum, and $\xi(0)$ is the ground state coherence length. For real world experiments, when, as a rule, only a part of full $B_{c2}(T)$ temperature dependence can be measured, there are several models were proposed to deduce extrapolated values for $\xi(0)$ from raw $B_{c2}(T)$ data measured at high reduced temperatures.

One of the model, which was proposed by Werthamer, Helfand, and Hohenberg [22,23], is extrapolative expression:

$$B_{c2}(0)=\frac{\phi_0}{2\cdot\pi\cdot\xi^2(0)}=-0.693\cdot T_c\cdot\left(\frac{dB_{c2}(T)}{dT}\right)_{T\sim T_c} \qquad (2)$$

In this paper, we will designate Eq. 2 as WHH model.

Another model, which is based on WHH primary idea [22,23], but one accurately generates full $B_{c2}(T)$ extrapolative curve from experimental data measured at high reduced temperatures, T/T_c, was proposed by Baumgartner *et al* [24]:

$$B_{c2}(T)=\frac{\phi_0}{2\cdot\pi\cdot\xi^2(0)}\cdot\left(\frac{\left(1-\frac{T}{T_c}\right)-0.153\cdot\left(1-\frac{T}{T_c}\right)^2-0.152\cdot\left(1-\frac{T}{T_c}\right)^4}{0.693}\right) \qquad (3)$$

We will designate this model as B-WHH model.

Gor'kov [25] proposed $B_{c2}(T)$ model which we used in our previous papers [15,16]:

$$B_{c2}(T) = \frac{\phi_0}{2 \cdot \pi \cdot \xi^2(0)} \cdot \left(\frac{1.77 - 0.43 \cdot \left(\frac{T}{T_c}\right)^2 + 0.07 \cdot \left(\frac{T}{T_c}\right)^4}{1.77} \right) \cdot \left[1 - \left(\frac{T}{T_c}\right)^2 \right]. \qquad (4)$$

We will designate this model as G-model.

Jones *et al* [26], proposed so called Jones-Hulm-Chandrasekhar (JHC) model:

$$B_{c2}(T) = \frac{\phi_0}{2 \cdot \pi \cdot \xi^2(0)} \cdot \left(\frac{1 - \left(\frac{T}{T_c}\right)^2}{1 + \left(\frac{T}{T_c}\right)^2} \right) \qquad (5)$$

III. Th_4H_{15}-Th_4D_{15} superconductors in Uemura plot

We start our consideration with the first discovered superhydride/superdeuteride superconductors, i.e., Th_4H_{15} and Th_4D_{15} [5]. From the author's knowledge, available to date experimental data for the upper critical field, $B_{c2}(T)$, for Th_4H_{15} and Th_4D_{15} are limited by values reported by Satterthwaite and Toepke [5], who measured that both Th_4H_{15} and Th_4D_{15} compounds have ground state upper critical field:

$$B_{c2}(T \sim 0) = 2.5 - 3.0\ T. \qquad (6)$$

From these values, the ground state coherence length, $\xi(0)$, for Th_4H_{15} and Th_4D_{15} phases, can be derived as following:

$$\xi(0) = 11.0\ \pm 0.5\ nm \qquad (7)$$

Miller *et al.* [27] for both phases reported the BCS ratio within a range:

$$\alpha = \frac{2 \cdot \Delta(0)}{k_B \cdot T_c} = 3.42 - 3.47. \qquad (8)$$

By utilizing superconducting transition temperature for Th_4H_{15} and Th_4D_{15} phases [5]:

$$T_c = 8.20\ \pm 0.15\ K \qquad (9)$$

one can deduce ground state superconducting energy gap:

$$\Delta(0) = 1.22\ \pm 0.03\ meV \qquad (10)$$

and by using well-known BCS expression [1]:

$$\xi(0) = \frac{\hbar \cdot v_F}{\pi \cdot \Delta(0)} \qquad (11)$$

where $\hbar = h/2\pi$ is reduced Planck constant, one can calculate the Fermi velocity, v_F, in Th_4H_{15} and Th_4D_{15} phases:

$$v_F = \pi \cdot \frac{\xi(0) \cdot \Delta(0)}{\hbar} = (6.4 \pm 0.2) \cdot 10^4 \ m/s \tag{12}$$

To place Th_4H_{15} and Th_4D_{15} phases in the Uemura plot [28,29], we need to make assumption about the effective charge carrier mass, m^*_{eff}, to calculate the Fermi temperature, T_F:

$$T_F = \frac{\varepsilon_F}{k_B} = \frac{m^*_{eff} \cdot v_F^2}{2 \cdot k_B} \tag{13}$$

Due to there is no any available experimental m^*_{eff} values for Th_4H_{15} and Th_4D_{15} phases, we can use for the lower bound of m^*_{eff} the value for another ambient pressure hydrogen-rich superconductor, PdH_x [30]:

$$m^*_{eff} = 0.49 \cdot m_e \tag{14}$$

and one calculates the Fermi temperature, T_F:

$$T_F = \frac{\varepsilon_F}{k_B} = \frac{m^*_{eff} \cdot v_F^2}{2 \cdot k_B} = 67 \pm 4 \ K \tag{15}$$

and the ratio:

$$\frac{T_c}{T_F} = 0.12 \pm 0.01 \tag{16}$$

For the upper bond of m^*_{eff} value we used the highest value reported for highly compressed hydrides, $m^*_{eff} = 3.0 \cdot m_e$ [31], and the lower bound for the T_c/T_F value can be calculated as:

$$\frac{T_c}{T_F} = 0.020 \pm 0.002 \tag{17}$$

Thus, performed analysis shows (Fig. 2) that in the scenario of $m^*_{eff} = 0.49 \cdot m_e$, Th_4H_{15} and Th_4D_{15} phases are located in the Uemura plot in the closed proximity to Bose-Einstein condensate (BEC) superfluid line together with ^{4}He and ^{40}K, and thus these two phases cannot be obey BCS theory. For the scenario of $m^*_{eff} = 3.0 \cdot m_e$, Th_4H_{15} and Th_4D_{15} phases are still within unconventional superconductor band in the Uemura plot (Fig. 2), where all unconventional superconductors (i.e. heavy fermions, fullerens, pnictides and cuprates) are located.

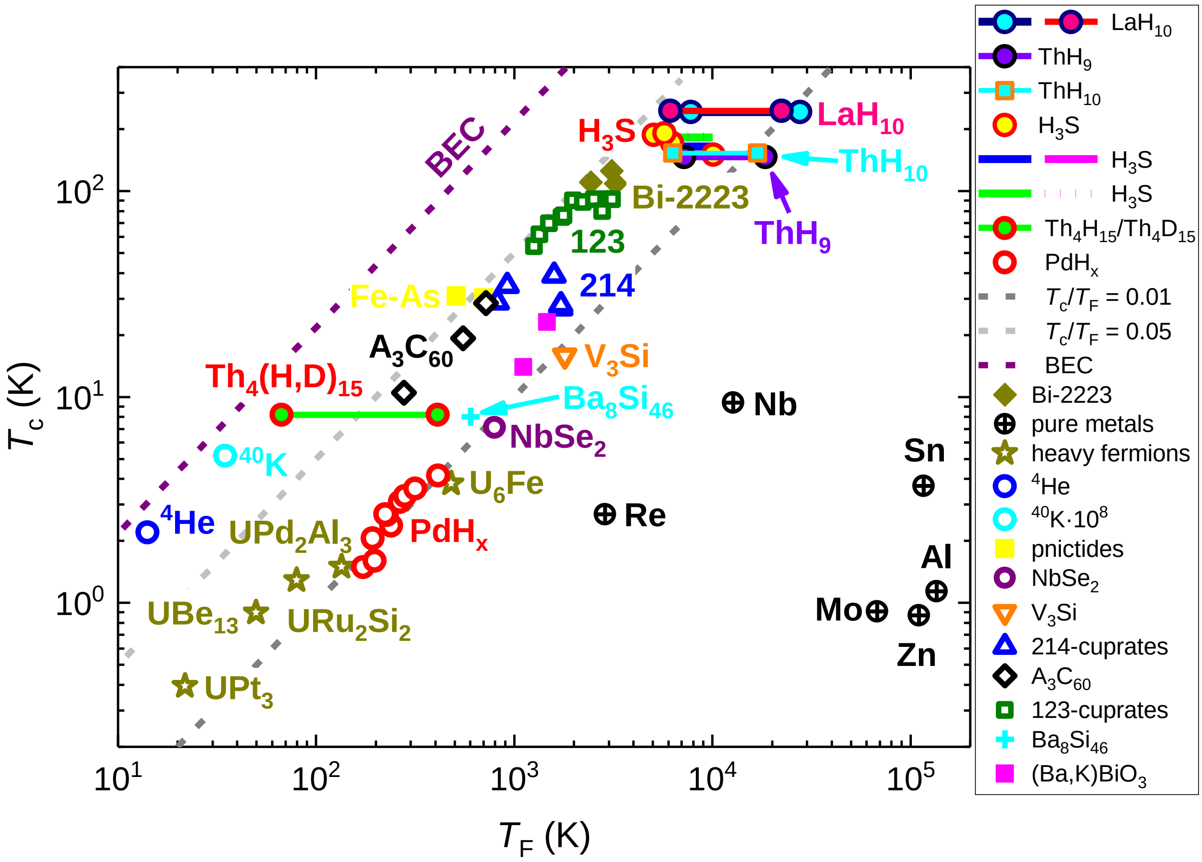

Figure 2. A plot of T_c versus T_F obtained for most representative superconducting families including PdH_x, Th_4H_{15}/Th_4D_{15}, ThH_9, ThH_{10}, H_3S, and LaH_{10}. Data was taken from Uemura [29], Ye *et al.* [32], Qian *et al.* [33], Hashimoto *et al.* [34] and Refs. 15,16.

IV. ThH_9 (P = 170 GPa) in Uemura plot

Semenok *et al* [35] reported on the discovery of high-temperature superconducting phase of ThH_9 at P = 170 GPa which exhibits *P6₃/mmc* crystallographic symmetry and superconducting transition temperature of T_c = 146 K. Semenok *et al* [35] also performed first principles calculations and deduced the effective mass in this superconductor:

$$m_{eff}^* = 2.73 \cdot m_e \tag{18}$$

which is remarkably close to the effective mass of $m_{eff}^* = 2.76 \cdot m_e$ in compressed H_3S [36].

Semenok *et al* [35] also proposed that ThH_9 has BCS ratio:

$$\alpha = \frac{2 \cdot \Delta(0)}{k_B \cdot T_c} = 4.74 - 4.89. \quad (19)$$

It should be noted, that as we already mentioned in our previous papers [15,16,37,38], that first principles calculations [31,35,36,39-41] always provide α-values near 5, which is the range of very strong-coupling limit for *s*-wave symmetry (it should be noted that other superconducting gap symmetries have weak-coupling limits of $\alpha \sim 5$ [42-44]).

However, it is needed to be mentioned, that first there are several new alternative approaches were developed to explain near-room-temperature superconductivity in compressed hydrides (we can mention, Hirsch and Marsiglio [45], Souza and Marsiglio [46], Harshman and Fiory [47], Kaplan and Imry [48]). For instance, Kaplan and Imry [48] showed that for the case of highly compressed H_3S their model gives α within weak-coupling BCS limit:

$$\alpha = \frac{2 \cdot \Delta(0)}{k_B \cdot T_c} = 3.53 \quad (20)$$

This α value is in a good agreement with ones deduced from experimental $B_{c2}(T)$ [15] and the self-field critical current density, $J_c(sf,T)$, data [,37,49]. Assuming that all hydrogen-rich superconductors have similar origin for the superconductivity, the value of a = 3.53 was used in our calculations as the lowest boundary value for α.

Semenok *et al* [35] measured $B_{c2}(T)$ data for ThH_9 phase at P = 170 GPa, which we fit to Eqs. 2-5 in Fig. 3. It can be seen that deduced T_c/T_F ratios are within usual range of unconventional superconductors band (Fig. 2).

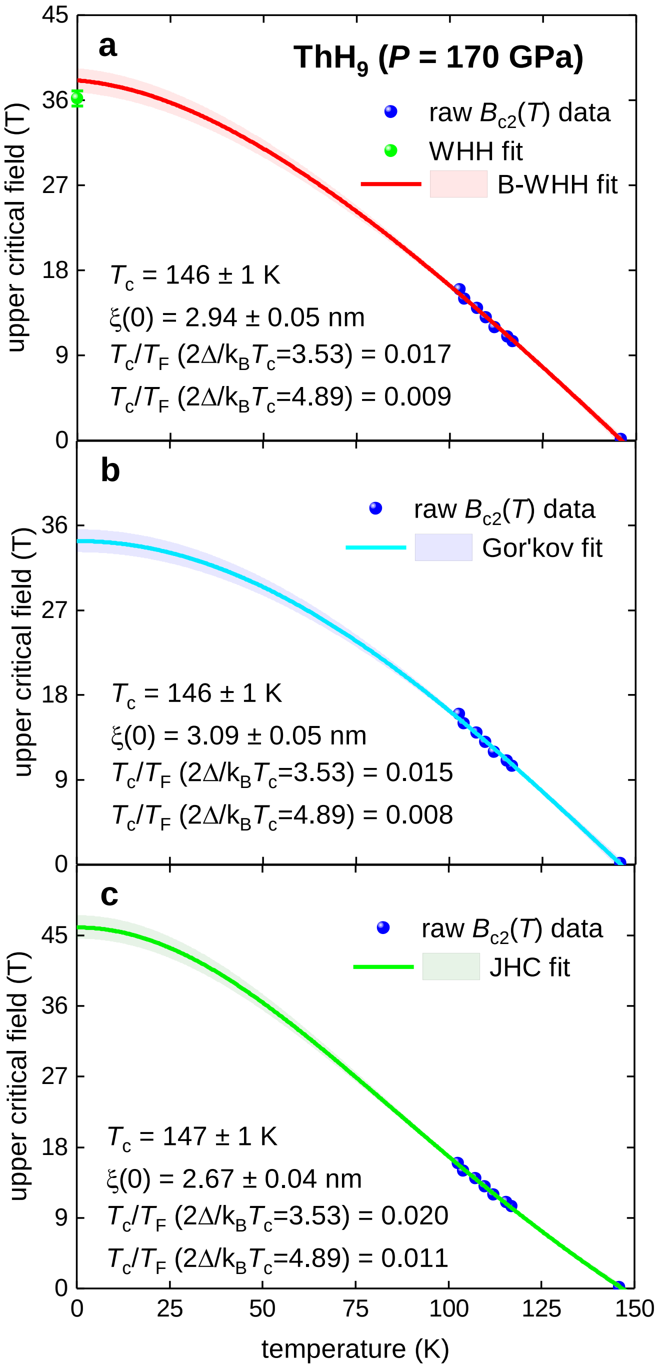

Figure 3. Superconducting upper critical field, $B_{c2}(T)$, data and fits to four different model (Eqs. 2-5) for ThH_9 superhydride compressed at pressure P = 170 GPa (raw data are from Ref. 35). (a) fit to WHH and B-WHH models, for latter the fit quality is $R = 0.998$; (b) fit to Gor'kov model, $R = 0.998$; (c) fit to JHC model, $R = 0.9988$. 95% confidence bars are shown.

V. ThH_{10} (P = 174 GPa) in Uemura plot

Semenok *et al* [35] also reported on the discovery of another high-temperature superconducting phase of ThH_{10} at P = 174 GPa, which exhibits $Fm\bar{3}m$ crystallographic symmetry and superconducting transition temperature of T_c = 159 K. In Fig. 4 we show raw upper critical field, $B_{c2}(T)$, data for this phase [35] and data fit to Eqs. 2-5.

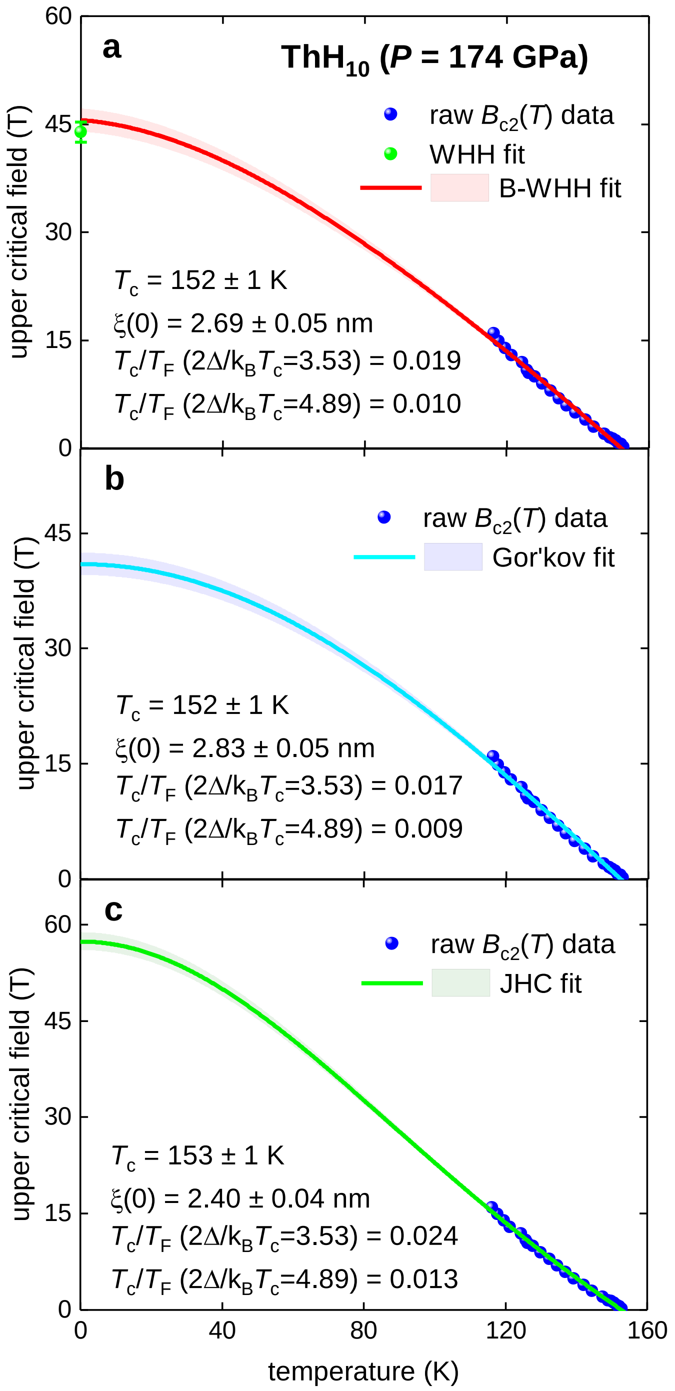

Figure 4. Superconducting upper critical field, $B_{c2}(T)$, data and fits to four different model (Eqs. 2-5) for ThH$_{10}$ superhydride compressed at pressure P = 174 GPa (raw data are from Ref. 35). (a) fit to WHH and B-WHH models, for latter the fit quality is R = 0.992; (b) fit to Gor'kov model, R = 0.992; (c) fit to JHC model, R = 0.997. 95% confidence bars are shown.

As expected, in the Uemura plot highly-compressed ThH_{10} superconductor is located within unconventional superconductors band (Fig. 2).

VI. Conclusions

In this paper we analyse experimental $B_{c2}(T)$ data for several thorium based superhydrides and Th_4D_{15} superdeuteride and come to conclusion that all discovered to date thorium hydrogen- and deuterium-rich superconductors for which fundamental superconducting parameters beyond T_c have measured, i.e., Th_4H_{15}, Th_4D_{15}, ThH_9 and ThH_{10}, are unconventional superconductors.

In addition, we stress that the isotope effect in H_3S-D_3S system should be further studied, because available to date experimental data are not sufficient to make solid conclusion.

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