Published in Appl. Phys. Lett. 121, 080501 (2022). Rejected for posting in arXiv on 6/28/2023
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Superconducting Materials: Judge and Jury of BCS-Electron-Phonon Theory

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(Dated: June 24, 2022)

By a recent count, there are 32 different classes of superconducting materials [1], only 12 of which are generally believed to be "conventional", i.e. described by the conventional BCS-electron-phonon theory of superconductivity. In this perspective I critically examine the successes and failures of the conventional theory to describe conventional superconductors, and discuss what is understood and not understood about hydrogen-rich materials claimed to be high temperature conventional superconductors under high pressure. I argue that the materials' evidence accumulated to date calls for dethroning the conventional theory of its privileged status, and seriously explore the alternative possibility that a single theory, different from the conventional theory, may describe superconductivity of all materials in a unified way.

I. INTRODUCTION

A paper titled "Failed theories of superconductivity" [2] reviewing the history of the field before the advent of BCS theory in 1957 framed the problem well in stating: "*Formulating the theory of superconductivity was one of the hardest problems in physics of the 20th century.*" Note that it didn't say *"Formulating a theory..."*. Indeed, all the great scientists that attacked the problem before BCS, such as Bloch, Born, Bohr, Heisenberg, Einstein, Landau, Kronig, Brillouin, Slater, Feynman, as well as Bardeen, Cooper and Schrieffer, were attempting to formulate "***the***" theory of superconductivity.

Yet according to the current generally accepted view they were all pursuing an impossible dream. The current view, motivated by the experimental findings in recent years that there are many superconducting materials that do not fit the BCS framework, holds that there is no such thing as "***the***" theory of superconductivity [3]. There is "***a***" theory of superconductivity [4, 5] to describe the superconducting elements and simple compounds [6] (the conventional theory), another theory to describe the high T_c cuprates [7], yet another one for the pnictides [8], another for heavy fermion materials [9], another for Sr_2RuO_4 [10], another for $SrTiO_3$ [11], another for bismuthates [12], another for doped C_{60} [13] another for layered nitrides [14], another for organic materials [15], etc etc [1]. Furthermore, except for being certain about the correctness of the first one [16], it is generally agreed (with the possible exception of their proponents) that it is uncertain which, if any, are the right theories among many proposed candidates to describe each of these numerous other classes of superconductors, and in particular how many different theories are needed to describe all superconducting materials.

These other theories are expected to have their own distinct pairing mechanism, symmetry of the superconducting state, and defining physical characteristics, one or more of these different from BCS. Pairing symmetries "unconventional s" [17], d [18], p [19], s+/- [20], p+ip [21], d+id [22], f [23], g [24], physics involving spin fluctuations [25], resonating valence-bonds [26], Mott insulating state [27], strange metal [28], holographic duality [29], charge density waves [30], d-density waves [31], pair density waves [32], Van Hove singularities [33], stripes [34], loop currents [35], pseudogaps [36], multibands [37], Lifshitz transitions/Fano resonances [38], excitons [39], plasmons [245], low dimensionality [41], polarons [42], bipolarons [43], Jahn-Teller physics [44], Dirac fermions [45], spin-momentum locking [46], anyons [47], topology [48], Majorana fermions [49], Weyl fermions [50, 51], nematicity [52], quantum criticality [53], Hund coupling [54], vestigial order [55], intertwined order [56], Gossamer superconductivity [57], time-reversal symmetry-breaking [58], etc, etc, are all deemed indispensable to describe one or the other of these multitude of different classes of superconductors [1, 3]. A far cry from the one symmetry, s, one mechanism, electron-phonon [4], and one overall framework, Fermi liquid theory, once believed to describe the essential physics of superconductivity for *all* superconducting materials [59].

This enormous proliferation of entities has come about because of the clearly established failure of the conventional theory to describe several classes of superconducting materials, particularly the high T_c cuprates. That opened Pandora's box. It is generally agreed that the electron-phonon mechanism [4] cannot describe the high

FIG. 1. The realization that BCS theory cannot describe all superconducting materials opened Pandora's box.

critical temperatures found in the cuprates. So *if* the conventional theory of superconductivity describes some superconducting materials, there has to be more than one theory of superconductivity. Pandora's box is wide open.

But what if the conventional theory is not the correct explanation of superconductivity for *any* material?

Such a scenario would rekindle the possibility that there is "***the***" theory of superconductivity that the ancient masters were searching for [2] and never found, rather than a large number of different theories on the same footing, as currently believed. True, there may be some classes of superconductors where particular characteristics of the materials are closely intertwined with their superconductivity giving rise to some different physics, yet the underlying mechanism for superconductivity would not be different than for any other material. Pandora's box would shut tight, and the variety of exotic alternatives to explain superconductivity in this or that material shown in Fig. 1 and others would fall by the wayside.

If such a unifying theory exists, with a pairing mechanism that under favorable conditions can give rise to the 140K superconductivity of mercury-barium-calcium copper oxide, would it be very surprising that under less favorable conditions the same pairing mechanism could also give rise to the 7K superconductivity of Pb, the poster-child for the conventional electron-phonon mechanism, the 'exotic' 2.3K superconductivity of CeCoIn5 [60] and 1.6 K superconductivity of UTe_2 [61], as well as the 'magic angle' 1.2K superconductivity of twisted graphene [62]?

In this perspective I argue that the experimental knowledge of superconducting materials accumulated during the last 110 years effectively condemns BCS electron-phonon theory to oblivion, contrary to the current general consensus, *provided* that the hydrides under high pressure are not high temperature superconductors. And that a single mechanism of superconductivity for all superconducting materials is by far the more likely scenario to describe superconductivity in nature, in conformance with Occam's razor.

II. WHAT IS SO COMPELLING ABOUT THE ELECTRON-PHONON MECHANISM?

The electron-phonon mechanism was once compelling because it was believed to be at the root of superconductivity for *all* superconductors. The citation by the Nobel committee that awarded the Nobel Prize to Bardeen, Cooper and Schrieffer in 1972 for the development of the theory of superconductivity reads in part (emphasis mine):

*"...**the** central problem, the question about **the** underlying mechanism for superconductivity, remained a mystery up to the late 50:s. The difference in energy between the superconducting and the normal state in a metal is extremely small in comparison with all typical energies in a metal and therefore many different mechanisms might a priori be possible. A significant step forward was taken around 1950 when it was found theoretically and experimentally that **the** mechanism for superconductivity had to do with the coupling of electrons to the vibrations of the crystal lattice. Starting from this mechanism, Bardeen, Cooper and Schrieffer developed in 1957 a theory of superconductivity, which gave a complete theoretical explanation of the phenomenon."*

And, the BCS paper itself started with the statement in the abstract *"A theory of superconductivity is presented, based on the fact that the interaction between electrons resulting from virtual exchange of phonons is attractive when the energy difference between the electrons states involved is less than the phonon energy, $\hbar\omega$."*

These two statements reveal that (1) the Nobel prize was awarded based on the belief at the time that BCS was ***the*** theory that finally provided an understanding of the phenomenon of superconductivity in nature, and (ii) an essential part of that understanding being that the electron-phonon interaction is ***the*** mechanism that gives rise to superconductivity. Would the BCS Nobel prize had been awarded if it had been known at the time that BCS with the electron-phonon mechanism provides an understanding of only a third of the thirty-plus different classes of superconducting materials that exist [1], as is currently believed?

The experimental evidence that convinced Bardeen [63] and Fröhlich [64] that the interaction responsible for superconductivity is the electron-phonon interaction was measurements of the isotope effect in 1950 [65], that ap-

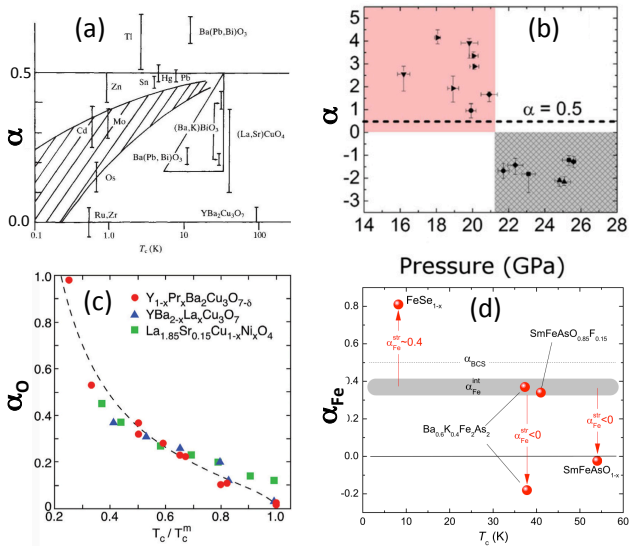

FIG. 2. Isotope coefficient α, predicted to be 0.5 by BCS theory. (a) Various materials, from ref. [66]; (b) Lithium under pressure, from ref. [67]; (c) High T_c cuprates, from ref. [44]; (d) Iron pnictides, from ref. [68].

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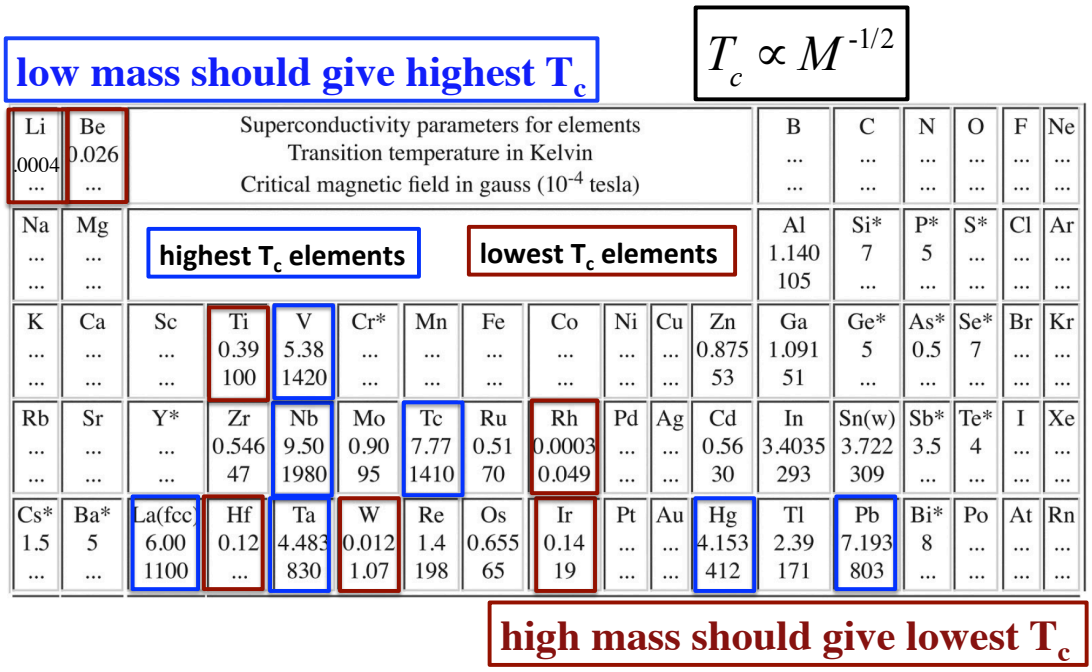

Li .0004 ...	Be 0.026 ...											B	C	N	O	F	Ne ... ...
Na	Mg											Al 1.140 105	Si* 7 ...	P* 5 ...	S*	Cl	Ar ... ...
K	Ca	Sc	Ti 0.39 100	V 5.38 1420	Cr*	Mn	Fe	Co	Ni	Cu	Zn 0.875 53	Ga 1.091 51	Ge* 5 ...	As* 0.5 ...	Se* 7 ...	Br	Kr ... ...
Rb	Sr	Y*	Zr 0.546 47	Nb 9.50 1980	Mo 0.90 95	Tc 7.77 1410	Ru 0.51 70	Rh 0.0003 0.049	Pd	Ag	Cd 0.56 30	In 3.4035 293	Sn(w) 3.722 309	Sb* 3.5 ...	Te* 4 ...	I	Xe ... ...
Cs* 1.5 ...	Ba* 5 ...	La(fcc) 6.00 1100	Hf 0.12 ...	Ta 4.483 830	W 0.012 1.07	Re 1.4 198	Os 0.655 65	Ir 0.14 19	Pt	Au	Hg 4.153 412	Tl 2.39 171	Pb 7.193 803	Bi* 8 ...	Po	At	Rn ... ...

FIG. 3. (An asterisk denotes an element superconducting only in a crystal modification that is not normally stable). The mass of the ions increases as we go from the upper left to the lower right corners of the periodic table. According to the conventional theory, higher T_c materials, shown in blue in the figure, should tend to have lower mass, hence predominate in the upper left region, and lower T_c materials, shown in brown in the figure, should tend to have higher mass, hence predominate in the lower right region. Such a trend is not apparent in the figure.

peared to show that

$$T_c \propto M^{-\alpha} \tag{1}$$

with M the ionic mass and α the isotope coefficient, predicted to be 0.5 by BCS theory. By sheer accident, several elements measured in 1950 showed an isotope coefficient close to 0.5 [65]. However, Fig. 2 shows isotope coefficients for many materials known today. It can be seen that α is all over the place, both for materials that are generally believed to be electron-phonon superconductors (upper panels) and for materials generally believed to *not be* electron-phonon superconductors (lower panels). There are many other examples not shown in Fig. 2 that call into question the argument that the isotope effect is a valid indicator of electron-phonon superconductivity. In particular, in PdH, when H is replaced by D, with twice the ionic mass, T_c *increases* by 20% [69] rather than decreasing by 30% as BCS would predict. In $SrTiO_3$, substitution of ^{16}O by ^{18}O causes T_c to *increase* by 50% [70] rather than to decrease. Yet the experimental evidence that the T_c of mercury decreases by 0.85% when the isotopic mass increases by 1.85% [65], is regarded as more compelling.

Figure 3 shows the critical temperature of the elements. According to Eq. (1) the highest T_c's should be found in the upper left region of the periodic table, and the lowest T_c's in the lower right region. Nothing of the sort is seen in the periodic table. The same is true for elements under pressure [71], as well as for alloys and compounds. For example, lutetium under high pressure has higher T_c than boron under high pressure, that is 16 times lighter. There is no empirical evidence that compounds with heavier elements have lower T_c's than similar compounds with lighter elements. For example, NbN's T_c is twice as high as VN's T_c, even though Nb is almost twice as heavy as V. TaC's T_c is three times higher than VC's even though Ta is more than three times heavier than V.

Are there reasons to expect that the tendency of T_c to decrease in going from the upper left to the lower right region of the periodic table predicted by Eq. (1) would be offset by some systematic behavior of other quantities that enter into determining T_c in the conventional theory? The generally assumed valid Mc Millan equation for T_c is [72]

$$T_c = \frac{\hbar < \omega >}{1.2 k_B} e^{-1.04(1+\lambda)/(\lambda - \mu^* - 0.62\lambda\mu^*))} \tag{2}$$

with μ^* the "Coulomb pseudopotential" typically assumed to be around 0.1, $< \omega >$ a characteristic phonon frequency proportional to $M^{-1/2}$, and

$$\lambda = g(\epsilon_F)\frac{\alpha^2}{K} \tag{3}$$

with α the electron-phonon interaction (entering the Hamiltonian as $\alpha q c^\dagger c$ with q a lattice displacement and

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$c^\dagger, c$ electron creation and annihilation operators), $g(\epsilon_F)$ the density of states at the Fermi energy, and K describing the stiffness of the lattice (potential energy $\propto Kq^2$). T_c increases as λ increases or μ^* decreases. There is no physical argument suggesting that either $g(\epsilon_F)$ or α should systematically increase, nor that K or μ^* should systematically decrease, as we go from the upper left to the lower right of the periodic table. Therefore, variations in those quantities should give fluctuations on an overall trend for T_c to decrease as the ionic mass increases, if Eq. (1) is valid. Such a trend is *not* observed in Fig 3.

Given this, the fact that the search for high temperature superconductors is currently focused on light elements and particularly hydrogen-rich materials is truly remarkable.

III. BCS'S PREDICTIVE POWER FOR MATERIALS

The inability of BCS-electron-phonon theory to predict either the existence of superconductivity in a material or its T_c before the advent of the hydrides is well known. It is difficult to find even one successful prediction, while wrong predictions abound. For example, early on Allen and Cohen predicted [73] that Li and Mg should be superconducting. Mg is not, Li is, albeit at a temperature vastly lower than originally predicted [73, 74]. Pickett, Klein and Papaconstantopoulos predicted $T_c = 29K$ for MoN [75], much higher than the similar compound NbN 17.3K. In reality, it is much lower, 5.0K. Other early examples of non-realized predictions are given in ref. [6].

The discovery of 39K superconductivity in MgB_2 in 2001 [76] fueled the conviction that light elements favor superconductivity, as predicted by BCS, and its superconductivity was claimed to be fully explained by the conventional theory [77, 78]. Empowered by their success, the authors of [77, 78] made predictions on related materials. Rosner et al [79] and Dewhurst et al [80] predicted superconductivity in hole-doped LiBC, isoelectronic and isostructural to MgB_2, at temperature substantially higher than in MgB_2. None has been found at any temperature [81]. Choi, Louie and Cohen, using *exactly* the same methodology with which they accurately postdicted the 39K superconductivity of MgB_2 [78], predicted superconductivity of CaB_2 in the range 46K to 50K [82]. No superconductivity was found [83]. Quan and Pickett predicted superconductivity for $LiBC_3$ at temperature comparable to MgB_2 [84]. None has been found so far. Undeterred by these theoretical efforts, unpredicted superconductivity at 2K was found experimentally in $CaBi_2$ [85], and successfully explained by the conventional theory *thereafter* [86].

Many more predictions of superconductivity in light-element compounds inspired by MgB_2 have been made in recent years, for materials at ambient pressure. Table I lists most of them. *NONE* of these predictions have been realized so far.

TABLE I. Predictions of superconductivity in light-element compounds inspired by MgB_2

Year	material	P(GPa)	T_c (K)	Ref.	**found?**
2002	Li_xBC	10^{-4}	100	[79]	**no**
2004	$B_{13}C_2$	10^{-4}	36	[87]	**no**
2008	B-doped diamond	10^{-4}	55	[88]	**no**
2008	OsN_2	10^{-4}	1	[89]	**no**
2008	$Be_2B_xC_{1-x}$	10^{-4}	5-13	[90]	**no**
2009	CaB_2	10^{-4}	46-50	[82]	**no**
2010	p-doped graphane	10^{-4}	90	[91]	**no**
2012	B_2C single layers	10^{-4}	19.2	[92]	**no**
2012	layered antiperovskite nickel carbides	10^{-4}	5-7	[93]	**no**
2014	BaC	5	4.32	[94]	**no**
2014	$Li_4B_5C_3$	10^{-4}	16.8	[95]	**no**
2015	Li_2B_3C	10^{-4}	54.9	[96]	**no**
2015	$Li_3B_4C_2$	10^{-4}	53.8	[96]	**no**
2015	$LiBSi_{1-x}Al_x$	10^{-4}	11-13	[97]	**no**
2015	CrH	10^{-4}	10.6	[98]	**no**
2016	two-dimensional boron allotropes	10^{-4}	7-27	[99]	**no**
2016	$NaBC$	10^{-4}	35	[100]	**no**
2017	Li-intercalated BN bilayer	10^{-4}	25	[101]	**no**
2017	borophene	10^{-4}	24.7	[102]	**no**
2018	hole-doped black phosphorus	10^{-4}	10	[103]	**no**
2018	$K_{1-x}B_6$	10^{-4}	65.3	[104]	**no**
2019	Li_2B	10^{-4}	0.14	[105]	**no**
2019	XBC (X = Mg, Ca, Sr, Ba)	10^{-4}	4-51	[106]	**no** **no**
2019	BeB_2Ta, Be_2B_2Ta	10^{-4}	7	[107]	**no**
2019	CoO_2	10^{-4}	21-28	[108]	**no**
2019	MgC_2	10^{-4}	15	[109]	**no**
2020	LiB_2	10^{-4}	60	[110]	**no**
2020	B_2Si, BSi, BSi_2	10^{-4}	21-30	[111]	**no**
2020	$MgXB_4$ $(X = Al, Li, Na, K)$	10^{-4}	30.4	[112]	**no**
2020	$LiBC_3$	10^{-4}	40	[84]	**no**
2020	16 B_4C_4 structures	10^{-4}	0-75	[124]	**no**
2020	$BSiC_2$	10^{-4}	73.6	[113]	**no**
2021	SrB_3C_3, BaB_3C_3	10^{-4}	43	[114]	**no**
2021	$c - B_{24}$	10^{-4}	13.8	[115]	**no**
2021	monolayer LiBC	10^{-4}	70	[116]	**no**
2021	2d TiB_4	10^{-4}	1.66	[117]	**no**
2022	monolayer $Mg_2B_4C_2$	10^{-4}	48	[118]	**no**
2022	*Rb*-substituted SrB_3C_3 clathrate	10^{-4}	75	[119]	**no** **no**
2022	hole-doped $MgCN_2$	10^{-4}	14.4	[120]	**no**
2022	$o - B_{16}$ boron allotrope	10^{-4}	14.2	[121]	**no**
2022	monolayer B_3N	10^{-4}	14.1	[122]	**no**

The inability of the conventional theory to predict superconductivity is occasionally acknowledged by the practitioners of the art. For example, Yin, Savrasov and Pickett write after describing their calculation of how the electron-phonon interaction can account for 20K super-

conductivity in Y under pressure [123]: "*there remains a serious shortcoming... What is lacking is even a rudimentary physical picture for what distinguishes Y and Li T_c around 20 K under pressure from other elemental metals which show low, or vanishingly small, values of T_c*". In other words, without knowing the result in advance, the result of such a calculation could be both a T_c of 20 K or a vanishingly small T_c. Similarly, Cohen writes [125] *"predicting is hard especially about the future... calculations after an experimental discovery are easier"*, while at the same time inexplicably claiming [125] that the theory *"is extremely robust and predictive."*

In contrast, Gross and coworkers anounced in 2005 that they had devised a scheme (SCDFT=superconducting density functional theory) to compute superconducting properties (including T_c) of simple conventional superconductors from first principles *"without any experimental input"* and *"free of any adjustable parameter"* [126–128]. Using this approach, they predicted 2K superconductivity in potassium under 23 GPa pressure in 2006 [129], and 120K superconductivity in H_3Se under 100 GPa pressure in 2016 [130]. Neither has been found. They also showed that this approach can predict superconductivity with T_c around 10K in several intermetallic layered materials structurally similar to MgB_2 systems such as SrC_2, $RbSi_2$ and $RbGe_2$ [131], and they point out that the predictions agree with predictions from a different approach [132], giving them both credibility, notwithstanding the fact that no experimental confirmation for any of these compounds exists. They concluded that *"This scheme establishes Eliashberg theory as a method for discovery new superconductors without the need of material-dependent parameters."* [131].

However, *none* of the 192 papers written by Gross after their seminal papers in 2005 [127, 128] contains a single prediction that has been experimentally verified. They do contain many claims that their calculational scheme can accurately explain many experimentally observed features such as gap anisotropy, multiple gaps, isotope effect, etc.

In summary, the pattern denounced by Bernd Matthias more than 50 years ago [133] continues unabated today: *"I can think of no other field in modern physics in which so much has been predicted without producing a single experimental success"*. It is clear that blindfolded monkeys throwing darts at a list of conducting materials would do no worse than BCS-Eliashberg-DFT-SCDFT practitioners in predicting new superconductors. The monkeys would of course do much worse in providing detailed explanations for what has been observed.

Except for a single class of materials: the hydrides under high pressure.

TABLE II. Some predicted superconducting hydrides under pressure before 2015

Year	material	P (GPa)	T_c (K)	Ref.
2006	SiH_4	202	166	[136]
2007	SnH_4	120	80	[145]
2008	GeH_4	120	80	[146]
2009	YH_3	17.7	40	[147]
2010	ScH_3	19	18	[149]
2010	LaH_3	10	20	[149]
2011	AlH_3	70	37	[150]
2011	GaH_3	120	102	[150]
2012	CaH_6	150	220	[151]
2012	KH_6	166	58-70	[152]
2013	BaH_6	100	30-38	[153]
2013	NbH_4	300	38	[154]
2013	MgH_4	100	29-37	[155]
2013	MgH_{12}	140	47-60	[155]
2014	LiH_2	100	31	[156]
2014	BeH_2	250	32	[157]
2014	H_2S	160	80	[158]
2014	$(H_2S)_2H_2$	200	204	[159]

IV. HYDRIDES UNDER PRESSURE

Motivated by Ashcroft's 2004 suggestion [134] that high temperature superconductivity should exist in hydrogen-rich compounds under high pressure, an intensive experimental and theoretical search got underway [135]. In his original paper, Ashcroft suggested compounds of hydrogen and group IV elements (Si, Ge, Sn, Pb), and several followup theoretical studies focused on silane (SiH_4) [136–138]. Dutifully, Eremets et al reported in 2008 that silane becomes superconducting at 17K under 100 GPa [139]. It was later determined that what had been measured by Eremets et al was most likely PtH (platinum hydride) [140], resulting from reaction of hydrogen with Pt wires and foil contained in the diamond anvil cell. Studies of SiH_4 up to 150 GPa revealed that it remains non-metallic and non-superconducting [141]. Nevertheless, as late as 2018 [142, 143] Eremets continued to claim that *"superconductivity at around 17 K in silane (SiH_4) was observed"*. In reality, the existence of superconductivity in silicon-hydrides under pressure remains unconfirmed to date, even though a large number of Si-H compounds under pressure have been predicted to be superconducting with T_c's in the range 16K-166K [144]. In fact, none of the compounds of hydrogen and group IV elements predicted by Ashcroft in 2004 to be superconducting, and confirmed by later theoretical studies, has been experimentally found to be superconducting to date.

In the years leading up to 2015, a variety of high temperature superconducting hydrides under pressures above 100 GPa (1 Mbar) were theoretically predicted in addition to the group IV ones predicted by Ashcroft, as shown in table II. Experiments at such high pressures are very difficult and only a few experimental groups are able to perform them. Several of these predictions were

Published in Appl. Phys. Lett. 121, 080501 (2022). Rejected for posting in arXiv on 6/28/2023
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TABLE III. Materials claimed to be experimentally confirmed high temperature superconductors under pressure, many predicted (some postdicted) by calculations based on the conventional theory of superconductivity.

Year	material	P (GPa)	T_c (K)	Ref.	structure
2015	H_3S	155	203	[161]	covalent
2015	H_3S	153	197	[162]	
2015	P_3S	200	100	[163]	covalent
2019	LaH_{10}	188	260	[164]	clathrate
2019	LaH_{10}	170	250	[165]	
2020	LaH_{10}	165	250	[166]	
2020	LaH_{10}	180	556	[167]	
2019	YH_9	201	243	[168]	clathrate
2019	YH_6	166	224	[169]	
2021	YH_9	182	262	[170]	
2020	ThH_{10}	170	160	[171]	clathrate
2020	CSH	267	287	[172]	covalent
2021	CSH	89	170	[173]	
2021	CaH_6	180	210	[174]	clathrate
2022	CaH_6	172	215	[175]	
2022	SnH_x	200	70	[176]	covalent
2022	CeH_9	95	115	[177]	clathrate
2022	ZrH_x	220	71	[178]	covalent
2021	$(La,Y)H_{10}$	183	253	[179]	clathrate
2022	$(La,Ce)H_9$	100	176	[180]	clathrate
2022	$(La,Ce)H_9$	110	178	[181]	clathrate
2022	$(La,Nd)H_{10}$	180	148	[182]	clathrate

explored experimentally and no superconductivity was found, and that was frequently blamed on "anharmonicity". Then, the field changed overnight when Eremets and coworkers reported in late 2014 the discovery of superconductivity in sulfur hydride under pressure up to a critical temperature 190K [160], and shortly thereafter up to 203K [161]. Eremets' search for superconductivity in sulfur hydride under pressure had been motivated by the theoretical prediction of ref. [158], and this was widely interpreted as meaning that the conventional theory of superconductivity now had predictive power, or as Allen and Cohen had anticipated 45 years too early [73], *"the theory of the transition temperature had come of age"*.

In the years since 2015, several other hydrides under high pressure were found experimentally to be high temperature superconductors as evidenced from resistance measurements. At the time of this writing, at least 13 different hydrides under pressure have been claimed to be high temperature superconductors, listed in table III, confirmed by at least 21 independent transport experiments (see Refs. in table III). They are believed to be of two different types: covalently bonded [158, 159] and clathrate structures [151] where the metal atom is in a "cage" surrounded by hydrogen atoms. However, because the location of H atoms in the structures cannot be discerned from X-ray experiments, this has not been subject to experimental verification.

Concurrently, the number of predicted superconducting hydrides under pressure has increased exponentially since 2015. Ref. [183] lists 61 predicted binary superconducting hydrides under pressure, 31 of them with critical temperature above 50K. In Ref. [184], the number increases to 240 predicted superconducting binary hydrides, 83 of them with critical temperature above 100K. Ref. [185] lists 31 ternary hydrides predicted to be superconducting, 24 of them with critical temperature at or above 200K. Etc.

TABLE IV. Some predicted superconducting hydrides at pressures below 100 GPa

Year	material	P (GPa)	T_c (K)	Ref.
2021	NaH_6	100	250	[186]
2021	$BeCH_4$	80	30	[187]
2021	$LaBH_8$	50	126	[189]
2022	$LaBeH_8$	50	126	[188]
2021	$Ba-CH_4$	40	30	[190]
2018	UH_7	20	50	[191]
2021	KB_2H_8	12	140	[192]
2022	ErH_2	14.5	80	[193]
2022	$BaSiH_8$	3	71	[194]
2022	$SrSiH_8$	27	126	[194]

Among the materials so far claimed to be experimentally confirmed superconductors listed in Table III, only one is superconducting at pressures below $100GPa$, namely CeH_9, and critical temperatures above 200K require pressures above 150 GPa. Recently the theoretical emphasis has shifted to looking for hydride superconductors at lower pressure, that would be more easily studied experimentally, mostly among ternary hydrides. Table IV lists several examples of predicted superconducting hydrides at pressures below $100GPa$, none of which have been experimentally realized. It is likely that many more theoretical predictions of high temperature superconductivity at lower pressures in ternary hydrides will be made in the near future as the large phase space is being explored through high throughput [186] and other techniques.

But how do we know that any of this is real?

V. ARE HYDRIDES UNDER PRESSURE HIGH TEMPERATURE SUPERCONDUCTORS?

Contrary to what is widely believed currently, I argue that high temperature superconductivity in pressurized hydrides is *not* an established fact.

There have been many reports of "USO" 's, "unidentified superconducting objects", over the years, that were later withdrawn or were never confirmed [195]. Researchers saw unexpected drops in resistance, or certain magnetic signatures, in many materials, and jumped to the conclusion that they were superconductors [196]. As one example, we mention claims of room temperature superconductivity in palladium hydride at ambient pressure [197]. Another recent one was a claim of superconductivity in palladium hydride at ambient pressure with T_c above 50K [198], showing curves for resistance versus

temperature similar to such curves for hydrides under pressure. For materials at ambient pressure it is much easier to determine whether such signals are fact or fiction by followup experiments. For the hydrides under high pressures, where only a few select groups have the ability to do these experiments, and the samples are extremely small and difficult to handle, it is much harder to confirm or rule out such indications.

Most importantly, the key difference between other "USO"s and hydrides is that there is a strong expectation that hydrides under pressure are high temperature superconductors because it is predicted by the conventional theory. The field is entirely driven by theory, as its practitioners readily acknowledge, unlike any other time in the history of superconducting materials research.

Experiments in hydrides under high pressure suffer from a lack of reproducibility. There is no guarantee that when an experiment is repeated, even by the same group, the same compound is synthesized. For example, for La-based superhydrides, starting from the same initial components some researchers obtain a compound with an apparent maximum T_c of $260K$ [164], others with T_c of $250K$ [165], and others with T_c above $550K$ [167]. Ref. [165] reports $T_c's$ of $250K$, $215K$, $110K$ and $70K$ at the same pressure for different samples prepared the same way. Ref. [199] reports a variety of different $T_c's$ in C-S-H for crystals synthesized in the same way explaining that they are "highly sensitive to thermodynamic pathways". In addition, because of the way samples are prepared, with laser heating with a focused beam of diameter much smaller than the size of the sample that the beam is scanning through, the resulting compounds are expected to be highly inhomogeneous and granular in nature. In addition, it is expected that large pressure gradients exist in the samples.

In recent work, we have pointed out several different instances where experimental evidence that had been put forth as proving superconductivity in hydrides was in fact flawed. Namely:

(1) Unusually sharp drops in the resistance versus temperature of the "room temperature superconductor" CSH that remain sharp when a magnetic field is applied [172] suggest the drops are not due to superconductivity [200, 201].

(2) The fact that the width of the superconducting transition in resistance measurements is independent of applied magnetic field for several other hydrides besides CSH suggests that in those materials the drops are not due to superconductivity either [202].

(3) If some hydride materials show drops in resistance that suggest superconductivity but are not due to superconductivity, the drops must be due to some other physics, that could potentially be the reason for resistance drops seen in all the hydrides believed to be superconducting [203].

(4) Reported magnetic susceptibility data for CSH claimed to show superconductivity [172] were shown to be *inconsistent* with underlying raw data [205–207].

(5) Magnetic susceptibility measurements of sulfur hydride claimed to show superconductivity [209] were shown to be an experimental artifact [210].

(6) Optical reflectance measurements on sulfur hydride claimed to show both that the material is superconducting and that the pairing mechanism is electron-phonon [211] were shown not to be supported by underlying raw data [212].

(7) A nuclear resonant scattering experiment on sulfur hydride claimed to show that the material excludes magnetic fields as expected for a superconductor [213, 214] was shown to be incompatible with another experiment reporting magnetization measurements on the same material [215, 216].

(8) Magnetization measurements reported for sulfur hydride and lanthanum hydride [217] show behavior incompatible with standard superconductivity and give rise to unphysical values of penetration depth and critical fields [218].

Magnetization measurements on only two of the 13 hydrides claimed to be superconducting, H_3S and LaH_{10},

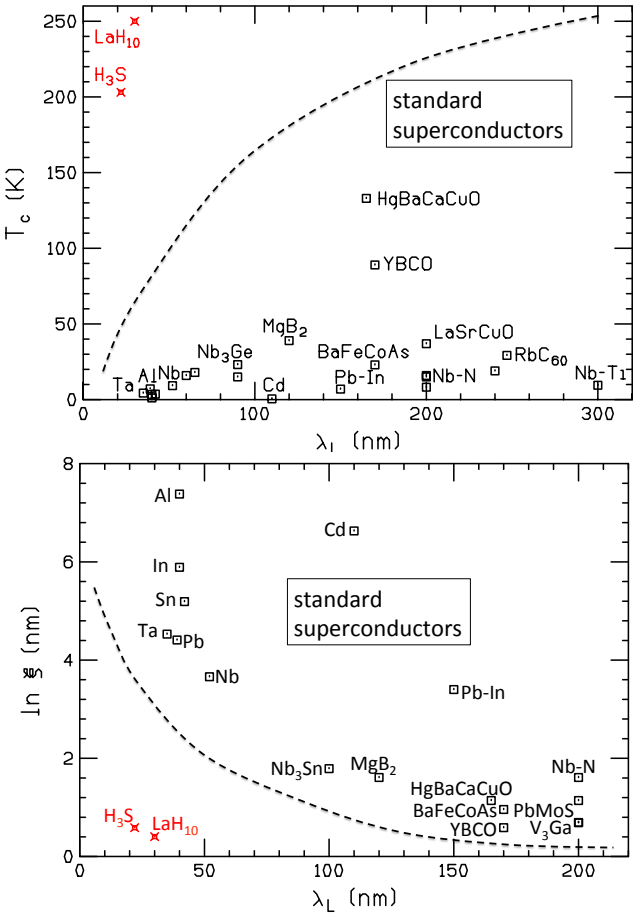

FIG. 4. Top panel: critical temperature vs London penetration depth, and bottom panel: coherence length vs London penetration depth, for standard superconductors (in black) and for hydrides under pressure (in red).

have been reported so far [217]. From those measurements, the London penetration depth λ_L and coherence length ξ were extracted as $\lambda_L = 22nm$, $\xi = 1.8nm$ for H_3S, $\lambda_L = 30nm$, $\xi = 1.5nm$ for LaH_{10} [217]. Fig. 4 shows where these 'superconductors' stand in the context of what we know as 'standard superconductors' [202], both conventional and unconventional. For small values of λ_L coherence lengths are large and T_c's are small for standard superconductors, in stark contrast to the hydrides. In addition, all standard superconductors with small values of λ_L exhibit a very clear Meissner effect, i.e. flux expulsion under field cooling, in contrast to the hydrides that show absolutely nothing under field cooling [217, 219]. All of the above strongly suggests that the hydrides under pressure are not superconductors.

VI. COMPLEMENTARITY BETWEEN THEORETICAL PREDICTION AND EXPERIMENTAL VERIFICATION

There seems to be a peculiar type of complementarity/uncertainty principle at play: theoretical calculations predicting conventional superconductivity in materials, and experiments on those materials, appear to be conjugate variables, like position and momentum in quantum mechanics. The more certain the theoretical prediction is claimed to be, the more uncertain are the experimental facts about the material. The more certain the superconductivity of a material is, the more uncertain (or non-existent) were the theoretical predictions for that material *before* the experiments were performed. Like particle-wave duality, a conventional superconductor can be either predicted with certainty or experimentally realized with certainty, but not both: they are mutually exclusive.

An illustrative example of this complementarity can be found in Ref. [131], where *"genuine predictions"* for superconducting T_c's of *"a set of novel superconducting systems"* are made, namely SrC_2, $RbGe_2$ and $RbSi_2$ at ambient pressure and SH_3 and SeH_3 under high pressure, where it is stated that *"Among these selected materials only SH_3 has been synthesized and confirmed experimentally"*. It should be noted that there is no a priori reason suggesting that it should be easier to predict structures and superconductivity for hydrides under pressure than for any other material.

Notwithstanding cold reality, theorists display enormous confidence that their predictions for superconducting materials and their T_c's are valid and reliable [220]. Some statements in recent theoretical papers predicting new hydrides:

"Conventional superconductivity is well understood, and theoretical tools are available for accurate predictions of the superconducting critical temperature. These predictions depend on knowing the microscopic structure of the material under consideration, which can now be provided by computational first-principles structure predictions." [143] (2020).

"first-principles calculations can provide accurate estimates for T_c" [221] (2022)

"State-of-the-art -ab-initio methods have reached predictive accuracy for conventional (phonon-mediated) superconductors" [222] (2020)

"it is now possible to predict the crystal structure using the quantum mechanical methods...Ab initio calculation power has been verified" [223] (2019).

"the normal- and superconducting-state properties of actual materials can now be computed to a high degree of accuracy based on the sole knowledge of their chemical composition and crystal structure" [224] (2019).

"The Eliashberg theory of superconductivity accounts for the fundamental physics of conventional superconductors, including the retardation of the interaction and the Coulomb pseudopotential, to predict the critical temperature T_c" [225] (2022).

"Migdal- Eliashberg theory, combined with first-principles calculations of the electron-phonon coupling, permits to make quantitative predictions of superconducting properties" [226] (2020).

Contrary to the above-cited quotes, I argue that, so far at least, violation of the complementarity/uncertaintly principle enunciated here has been just as elusive as attempting to determine which slit an electron in the double slit experiment went through before landing on a bright fringe.

VII. CAN THE CONVENTIONAL THEORY OF SUPERCONDUCTIVITY BE WRONG?

Let me clarify what I mean by the theory being wrong: I mean, the theory does not describe the physics of superconductivity of any real superconducting material. That is of course not incompatible with the fact that some aspects of the conventional theory are undoubtedly correct and describe real properties of real superconductors. The conventional theory predicts, in agreement with observations, that:

(1) Superconductors have macroscopic phase coherence.

(2) Pairs of electrons (or holes) play a key role in superconductivity.

(3) There is an energy gap in (most) superconductors between the ground state and excited states.

(4) The ground state of a superconductor in the presence of a small external magnetic field excludes the magnetic field from its interior.

(5) The ground state of a rotating superconductor in the absence of applied external fields has a uniform magnetic field in its interior.

However, those predictions are unrelated to the assumption that the electron-phonon interaction drives superconductivity according to the conventional theory, which is what guides the search for conventional superconducting materials focused on light elements. A Hamil-

tonian with an instantaneous local attractive interaction [227] (e.g. an attractive Hubbard model) also predicts (1)-(5), as does a retarded interaction of non-phononic origin.

The "evidence" that the electron-phonon interaction drives conventional superconductivity is (a) the isotope effect, (b) the fact that for some materials there is structure in tunneling spectra that appears to match structure in the phonon spectra [228], and (c) the claim by theorists that Eliashberg theory and the electron-phonon interaction can account for the critical temperatures of all "conventional superconductors". I have discussed here and elsewhere [229] reasons for why this evidence is questionable.

So let us assume that the electron-phonon interaction is not responsible for superconductivity, but some other attractive interaction is. Can the rest of the conventional theory still be correct, with a different pairing interaction?

The Hamiltonian that describes superconductivity has to have the necessary physical ingredients to describe the Meissner effect, the *process* by which a magnetic field is expelled from the interior of a material cooled from the normal into the superconducting state. And similarly, it has to have the physical ingredients necessary to describe the fact that when a rotating normal metal is cooled into the superconducting state, electrons near the surface spontaneously slow down. I have argued elsewhere that the BCS Hamiltonian and its variants don't have the physical ingredients necessary to describe that physics, and that the essential missing physics is electron-hole asymmetry [230]. This leads to a unified theory of superconductivity for all materials [231] and to guidelines for the search for higher temperature superconducting materials that are qualitatively different from those given by the conventional theory. In a nutshell, that high T_c will occur in systems where holes propagate through negatively charged anions in close proximity, as in MgB_2, the cuprates, and the pnictides, completely independent of whether the ions are light or heavy. The theory predicts that superconductivity cannot exist in materials that don't have hole carriers [232], consistent with the empirical observation that hole carriers favor superconductivity [233, 234].

VIII. THROWING DOWN THE GAUNTLET ON ELECTRON-PHONON SUPERCONDUCTIVITY

For 65 years it has been claimed, with increasing levels of confidence, that the conventional BCS-electron-phonon theory of superconductivity can predict T_c's for real materials. The claim has not been proven, because:

- Essentially all successful 'predictions' were made for materials for which T_c had already been measured
- Many materials for which T_c was predicted turned out not to be possible to make. This is despite the fact that for several years now it has been claimed that first principles calculations can reliably predict which materials will form or not [238–241].
- When a material could be made and did not exhibit the T_c predicted (which is always, except for the hydrides under pressure) it was claimed that the sample 'made a mistake', meaning it had properties that had not been anticipated, e.g. the phonons were anharmonic, spin fluctuations, etc.

This real state of affairs is not accurately represented in the literature. For example, Ref. [183] states about the superconducting T_c of elements: *"The accuracy reached by computational methods for conventional superconductors is demonstrated by the periodic table of superconductivity for elemental solids...A large body of literature shows that, if computational-theoretical methods are used to estimate the value of T_c for these 53 superconducting elements, the deviations with respect to the experimental values are small, usually less than* 20% *of T_c"*. It omits to say that those calculations were *all* performed *after* the experimental value of T_c was known, and even so they often don't work. For example, already in 1967 Carbotte and Dynes computed the transition temperature of Al and found it to be $T_c = 1.17$ K, in remarkable agreement with the experimental value $T_c = 1.18$ K [235]. Two years later another calculation for Al [73] yielded $T_c = 3.0K$, differing from the experimental value and the earlier theoretical prediction by 254%. Eight years later an improved calculation yielded $T_c = 0$ for Al [242]. Fast forward to 2020, where calculations using SCDFT including plasmonic effects yielded values for the T_c of Al ranging all the way from 0.3K to 7.6K depending on the approximation scheme used [243]. More generally, the T_c's of several elements were recently calculated [244] from first principles using SCDFT [127], no adjustable parameters, including plasmons [245], with spin fluctuations (SF) [246], without SF, with the spin-orbit interaction (SOI) [247] and without the SOI. The agreement with experiment was not better than it was in 1967, nor better than what it was in 2005 without plasmons, SF and SOI corrections [128], and results still disagreed qualitatively with observations in e.g. Cd, Zn, V, Pt and Au [244].

The fact is, the *only* cases where it can rightfully be said that the conventional theory has proven it's claimed "predictive power" in several instances is hydrides under high pressure [151, 158, 159, 236, 237]. But this is only so assuming the predicted hydrides currently claimed to be superconductors are indeed superconductors. That is far from established, as discussed in Sect. V.

So here I would like to propose that the predictive power of the conventional theory be tested in the following way. Table V shows critical temperatures for binary compounds, all of which crystallize in the NaCl structure. There are 14 elements on the left column and 9 elements on the upper row. That gives 126 combinations, of which

TABLE V. Binary compounds with the NaCl structure, from refs. [248–250]

	N	C	B	P	Sb	O	S	Se	Te
Nb	17.3	12				1.39			
V	8.5	3.2				<0.3			
Ta	6.5	10.3							
Ti	5.49	3.42				2.0			
Zr	10.7	<0.3	3.4	4.8			3.3		
Hf	8.83	<1.20	3.1	4					
Sc	<1.38	<1.38					4.2	3.7	
Y	<1.4	<1.38			<1.02		1.9	2.5	2.05
La	1.35			<1.68	<1.02		0.87	1.02	1.48
Cr	<1.28								
Mo	5.0	14.3							
W	<1.38	10.0							
Re		3.4							
Th				0.22			0.49	1.72	

at least 45 exist at ambient pressure, those with non-zero entries in the table. It is possible that even more exist already or can be made in the future. Each of those 45 compounds has a lattice constant a that has presumably been measured, and a measured value of T_c (or an upper bound) that is given in the table.

It should be straightforward for practicioners of first-principle calculations within DFT-Eliashberg or SCDFT theory to construct a computer program that will take as input the atomic number of an element in the column and that of an element in the row, and the measured lattice constant of the compound, and calculate a value for T_c. The program should obviously *not* have any if-statements that tell it to do different things depending on what *both* elements are. To make it very clear: the program of course could say *"if the column element is Ti, do this, if it is Sc, do that"*. But it cannot say *"if the column element is Ti and the row element is N do this, if the column element is Ti and the row element is C do this instead"*.

Such a computer program can be used to check whether the values of T_c predicted by that program bear resemblance to the values given in table V, or whether they do not. It will not allow for "superflexibility" [251]. The program should also be made publicly available, so anybody can run it themselves, as well as read the source code and verify that it complies with the ground rules given in the previous paragraph. This would allow for a meaningful evaluation of the predictive power of the conventional theory where not only the 'gatekeepers' [229] get to participate.

IX. CONCLUSIONS

At the time of this writing (June 2022), the paper [161] "Conventional superconductivity at 203 kelvin at high pressures in the sulfur hydride system" has been cited 1,877 times, and an entire field of research has sprung up from the firm conviction in the scientific community that sulfur hydride both is a superconductor and is governed by the conventional BCS-Eliashberg theory of superconductivity. Enormous resources have been and continue to be devoted world-wide to the search for new superconducting materials using guidelines provided by the conventional theory.

If the conventional theory does not describe real superconductors these resources are wasted. I suggest that experimentalists should stop relying on the conventional theory to interpret and guide their experiments searching for ever higher $T_c's$, as well as stop highlighting differences in their experimental findings with BCS predictions to bolster their case for having found a new interesting "unconventional" superconductor, and instead analyze critically what experiments tell them without invoking the conventional theory to interpret the significance of their findings. And in particular, if a material shows a drop in resistance, it does not mean it is superconductivity just because the conventional theory says so!

In searching the literature for writing this perspective, I was open to the possibility of finding some examples of recent predictions other than the hydrides that were validated by posterior experiments, even approximately so. I found none. For example, table I in this paper lists 40 predictions of new superconducting materials inspired by MgB_2 during the last 20 years. Not a single one of these predictions has been realized. Excluding the hydrides, the conventional theory has never been able to *predict* superconducting $T_c's$, not even qualitatively, to this day.

All of the above suggests, *under the assumption* that the hydrides under high pressure will in the end be proven to *not* be high temperature superconductors, that high temperature superconductivity in pressurized hydrides is the conventional theory's swan song:

The silver Swan, who living had no Note,
when Death approached, unlocked her silent throat.
Leaning her breast against the reedy shore,
thus sang her first and last, and sang no more. [252]

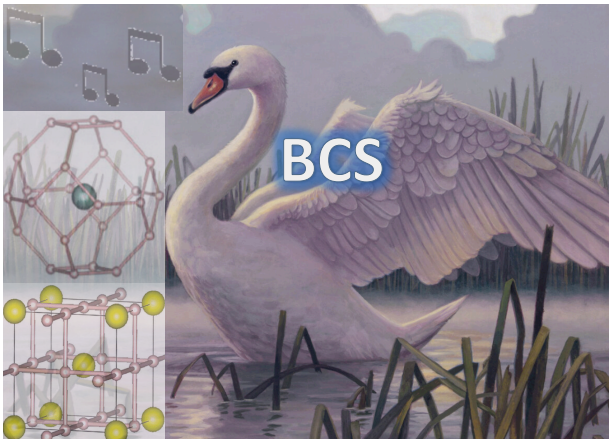

FIG. 5. Hydrogen-rich materials under high pressure: BCS theory's swan song?

which will be followed (hopefully sooner rather than later) by the demise of the conventional theory, once it is established that the experimental observations interpreted as indicating superconductivity in pressurized hydrides were, in fact, not indicating superconductivity.

Thereafter, theorists resuming the search for a unified theory of superconductivity that the reverence to BCS theory blocked for so many years, will hopefully start making positive rather than negative contributions to the quest for higher temperature superconducting materials, and room temperature superconductivity will have a sporting chance to turn from dream to reality.

ACKNOWLEDGMENTS

I am grateful to Frank Marsiglio for long-standing collaboration in the problem of superconductivity, and to D. van der Marel for recent collaboration. I would also like to acknowledge helpful discussions and information received on various aspects of hydride superconductivity with/from D. Semenok, S. Shylin, M. Eremets, S. Budko, V. Minkov, T. Timusk, P. Roy, S. Friedemann, A. Grockowiak, S. Tozer, X. Huang, T. Cui, J. J. Hamlin, V. Struzhkin, J. Cheng, R. Hemley, M. Nuñez-Regueiro, J. Flores Livas, E. Zurek, M. L. Cohen, W. Pickett, I. Felner, E. Talantsev, I. Bozovic, H. R. Ott, T. Forgan, M. Debessai, J. Schilling, K. Shimizu, S. K. Sinha, M. B. Maple and the late N. W. Ashcroft.

Note added: While this paper was being reviewed, experimental results claiming to show the existence of trapped magnetic flux in H_3S were reported [253]. If indeed the material traps flux, it would be a proof that it is a superconductor [254]. However, we have argued [255] that the interpretation [253] of the experimental results is flawed, and that when interpreted correctly the experimental results show that the material does not trap flux.

Data availability statement: The data that support the findings of this study are available from the author upon reasonable request.

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