Nonstandard superconductivity or no superconductivity in hydrides under high pressure

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Over the past six years, superconductivity at high temperatures has been reported in a variety of hydrogen-rich compounds under high pressure. That high temperature superconductivity should exist in these materials is expected according to the conventional theory of superconductivity, as shown by detailed calculations. However, here we argue that experimental observations rule out conventional superconductivity in these materials. Our results indicate that either these materials are unconventional superconductors of a novel kind, which we term "nonstandard superconductors," or alternatively, that they are not superconductors. If the first is true, we point out that the critical current in these materials should be several orders of magnitude larger than in standard superconductors, potentially opening up the way to important technological applications. If the second is the case, which we believe is more likely, we suggest that the signals interpreted as superconductivity are either experimental artifacts or they signal other interesting physics but not superconductivity.

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I. INTRODUCTION

According to the conventional Bardeen-Cooper-Schrieffer (BCS)–Eliashberg theory of superconductivity [1], superconductivity at high temperatures should exist in materials with light ions and large electron-phonon coupling. Ashcroft suggested in 1968 [2] that such conditions would be met in hydrogen turned metallic at high pressures, and in 2004 extended his prediction to include metal hydrides with high hydrogen content [3]. Motivated by these theoretical predictions, substantial experimental and theoretical efforts have been expended in recent years searching for high temperature conventional superconductivity in metal hydrides under high pressure [4–6]. Beginning in 2015 with Eremets *et al.*'s discovery of superconductivity in pressurized hydrogen sulphide [7], several metal hydrides under high pressure have been found to exhibit high temperature superconductivity [7–15], culminating in the recently reported discovery of room temperature superconductivity in a compound with carbon, sulfur, and hydrogen [14] (hereafter called CSH).

Together with these experimental discoveries, sometimes before and sometimes shortly thereafter, a variety of detailed calculations were performed based on BCS-Eliashberg theory that both predicted and apparently confirmed that these materials are high temperature conventional superconductors [16–26]. Sometimes superconductivity was "confirmed" without the need of *any* experimental evidence [27–30]. It should be pointed out, however, that these calculations are applied in a range of electron-phonon coupling constant $\lambda \gtrsim 2$ where numerical calculations [31] and theoretical considerations [32–34] suggest that Eliashberg theory is no longer applicable due to polaron formation. In addition, all these calculations rely on estimating the "Coulomb pseudopotential" μ^* that cannot be calculated independently, and that can suppress superconductivity if it is sufficiently large [1,35]. Furthermore, these calculations do not take into account the possibility of competing instabilities such as charge density wave [36,37] and magnetic phases [38–40]. In this paper we show that, in fact, *these materials cannot be conventional superconductors*, contrary to the experimental and theoretical evidence cited above.

Superconducting materials [41] are generally understood to be either "conventional" or "unconventional" [42]. Unconventional superconductors are materials that do not become superconducting driven by the BCS-electron-phonon interaction, but rather through some different physics, generally believed to be related to strong electron-electron interactions, for example, the high T_c cuprates [43] and iron pnictides [44]. However, even unconventional superconductors exhibit standard properties of superconductivity, such as the Meissner effect, upper and lower critical fields, critical current, and so on [45]. In this paper we call such superconducting materials [41] "standard superconductors" and we introduce a new category of nonconventional superconductors, which we call "*nonstandard superconductors*": materials that do not exhibit the basic properties of superconductors that standard superconductors, both conventional and unconventional, share. We will argue that hydrides under pressure, *if* they are superconductors, belong to this new category of superconductors.

According to the established understanding of superconductivity [45], there are three lengths that play a critical role in all standard superconductors, whether conventional or unconventional: the London penetration depth λ_L, the Pippard coherence length ξ, and the electron mean free path ℓ. Type I superconductors have $\lambda_L < \xi$ and are a limited set of materials, mostly elements in pure form, all believed to be conventional superconductors [35]. Most superconducting materials, both conventional and unconventional, are type II, with $\xi < \lambda_L$. A strongly type II material has $\xi \ll \lambda$. Disorder (short ℓ) increases λ_L and turns a type I material into type II.

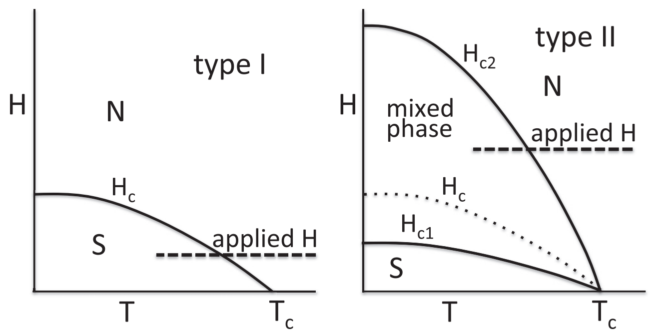

FIG. 1. Schematic phase diagrams of type I and type II superconductors in a magnetic field. N = normal state, S = superconducting state.

Most standard superconductors, and in particular the unconventional cuprates and iron pnictides, are type II materials. The higher the T_c the more strongly type II the material tends to be.

In this paper we argue that hydride superconductors exhibit properties characteristic of *both* type I and type II superconductors, in particular, strongly type II. Standard superconductors can be borderline type I/type II if $\xi \sim \lambda_L$, but they cannot be type I and strongly type II simultaneously. Therefore, we will argue that hydrides under pressure are "nonstandard superconductors" or, alternatively, that they are not superconductors.

A preliminary account of this work can be found in Ref. [46]. While the present paper was in the final stages of completion, a paper by Dogan and Cohen appeared [47] that expanded on our arguments [46] that the behavior of CSH [14] is incompatible with standard superconductivity and independently suggested the possibility of nonstandard superconductivity.

II. TYPE I VERSUS TYPE II SUPERCONDUCTIVITY

As summarized in Sec. II of Ref. [35], the phenomenology of the response of standard superconductors to magnetic fields is common to conventional and unconventional, i.e., standard, superconductors. A microscopic theory is not required to describe this response [45].

Figure 1 shows schematically the phase diagrams of type I and type II superconductors when a magnetic field H is applied. In a type-I superconductor the magnetic response is perfect diamagnetism: the magnetic field is completely expelled (except within a region of thickness λ_L from the surface) provided the field strength is less than a critical value $H_c(T)$, the thermodynamic critical field. Above this field value the material is in the normal state. When the system is cooled in an applied field H, it undergoes a reversible first-order phase transition from the normal to the superconducting state at the temperature $T(H)$ where $H = H_c[T(H)]$. The resistance of the material is finite for $T > T(H)$ and drops to zero discontinuously at $T = T(H)$ in an ideal situation. In the presence of imperfections in the sample the transition will be slightly broadened both in the absence and in the presence of applied magnetic field.

Instead, in a type-II superconductor the material exhibits perfect diamagnetism only up to a critical field H_{c1} smaller than H_c; with increasing applied field, flux begins to penetrate the material in the form of vortices. This continues to occur up to an upper critical field, H_{c2}, above which the material becomes normal. When cooling a type-II superconductor in an applied field H, the system will undergo a second-order phase transition at the temperature $T(H)$ where $H = H_{c2}[T(H)]$ where it enters the "mixed phase," with vortices in its interior, their density depending on temperature and magnetic field. In this mixed phase, whether or not the system has finite or zero resistance depends on the behavior of the vortices, whether or not they are "pinned." We will discuss this in a later section. Note from Fig. 1 that upon lowering the temperature for given applied H the system necessarily enters the mixed phase first. If the applied field is sufficiently small so that $H < H_{c1}(T=0)$, at a lower temperature the system will enter the perfectly conducting state with zero resistance. Instead, if $H > H_{c1}(T=0)$ the system remains in the mixed phase down to zero temperature.

Whether a material is a type-I or a type-II superconductor depends on the Ginzburg-Landau parameter $\kappa = \lambda_L(T=0)/\xi(T=0)$. Since the penetration depth increases as the mean free path decreases [45] a type-I superconductor can be made into a type-II superconductor through disorder. The thermodynamic critical field $H_c(T)$ is determined by the difference in the free energy per unit volume between normal and superconducting states as

$$f_n(T) - f_s(T) = \frac{H_c(T)^2}{8\pi} \tag{1}$$

both for type I and type II superconductors, and at zero temperature it is given within BCS theory by

$$\frac{H_c(0)^2}{8\pi} = \frac{1}{2}g(\epsilon_F)\Delta(0)^2, \tag{2}$$

where Δ is the energy gap and $g(\epsilon_F)$ the density of states at the Fermi energy. Within Ginzburg-Landau (GL) theory the thermodynamic critical field is given by

$$H_c(T) = \frac{\phi_0}{2\sqrt{2}\pi\lambda_L(T)\xi(T)}, \tag{3}$$

where $\phi_0 = hc/2e$ is the flux quantum. The lower and upper critical fields for type II superconductors are given within GL theory by

$$H_{c1}(T) = \frac{\phi_0}{4\pi\lambda_L(T)^2} ln[\kappa(T)], \tag{4a}$$

with $\kappa(T) = \lambda_L(T)/\xi(T)$,

$$H_{c2}(T) = \frac{\phi_0}{2\pi\xi(T)^2} \tag{4b}$$

and are related to the thermodynamic critical field by

$$H_{c1}H_{c2} = H_c^2 ln(\kappa). \tag{5}$$

A strongly type-II superconductor has $\kappa \gg 1$ and hence $H_{c1} \ll H_c \ll H_{c2}$.

To get simple explicit expressions for the temperature dependence we can use the two-fluid model. The thermodynamic critical field is given by ($t \equiv T/T_c$),

$$H_c(T) = H_c(0)(1 - t^2), \tag{6}$$

and the London penetration depth by

$$\lambda_L^2(T) = \lambda_L^2(0)\frac{1}{1 - t^4}. \tag{7}$$

The upper critical field is then given by

$$H_{c2}(T) = H_{c2}(0)\frac{1 - t^2}{1 + t^2} \tag{8}$$

and the coherence length by

$$\xi(T) = \xi(0)\sqrt{\frac{1 + t^2}{1 - t^2}}. \tag{9}$$

III. ARE HYDRIDE SUPERCONDUCTORS TYPE I OR TYPE II?

We argue that experimental evidence clearly shows that hydride superconductors, particularly those with high T_c, are strongly type II. Ideally, one would want to measure both ξ and λ_L to answer this question. However in none of the experiments reported so far was λ_L inferred from experiment (in Ref. [14] an erroneous value for λ_L not based on experimental evidence was given [46,47]). Experimental values for the coherence length ξ are inferred from the lowering of the transition temperature under an applied magnetic field. Here we argue that those reported results and theoretical considerations necessarily imply that $\lambda_L \gg \xi$, hence that the materials are strongly type II (if they are superconductors).

Consider the carbonaceous sulfur hydride (CSH) recently reported to be a room temperature superconductor. Resistive transitions in applied magnetic field are shown in Fig. 2 of Ref. [14], showing that the transition temperature drops from 287 K for no applied magnetic field to increasingly lower values as the applied magnetic field increases. Let us consider the possibility that CSH might be a type-I superconductor. The temperature dependence of the thermodynamic critical field is given within the two-fluid model by Eq. (5). In an applied field of 9 T, the resistance drops almost discontinuously (in a range $\Delta T < 1K$) from a finite value to essentially zero [14]. This then implies that $H_c(265\,\mathrm{K}) = 9\,\mathrm{T}$. With $T_c = 287\,\mathrm{K}$ Eq. (5) yields $H_c(T = 0) = 61\,\mathrm{T}$. Within BCS theory the energy gap at zero temperature and the critical temperature are related by

$$\frac{2\Delta(0)}{k_B T_c} = 3.53, \tag{10}$$

yielding $\Delta(0) = 44$ meV. From Eq. (2) this yields for the density of states at the Fermi energy

$$g(\epsilon_F) = \frac{9.7\,\mathrm{states}}{\mathrm{eV}\,\mathrm{\AA}^3}. \tag{11}$$

Assuming the density of states is given by the free electron expression with an effective mass m^* yields

$$g(\epsilon_F) = \left(\frac{3}{\pi^4}\right)^{1/3} n^{1/3}\frac{m^*}{\hbar^2} = \frac{0.0411}{} eV\,\mathrm{\AA}^2 n^{1/3}\left(\frac{m^*}{m_e}\right), \tag{12}$$

with n the number density and m_e the bare electron mass. The band effective mass should be close to the bare electron mass, corrected by the mass enhancement resulting from the electron-phonon interaction. Assuming an electron-phonon coupling constant $\lambda \sim 2$ resulting from Eliashberg calculations yields

$$m^* = m_e(1 + \lambda) \sim 3m_e, \tag{13}$$

yielding

$$g(\epsilon_F) = \frac{0.123}{\mathrm{eV}\,\mathrm{\AA}^2} n^{1/3}. \tag{14}$$

The Wigner-Seitz radius (radius of a sphere with volume equal to the volume per conduction electron) is

$$r_s = \left(\frac{3}{4\pi n}\right)^{1/3}, \tag{15}$$

so that in order for the density of states Eq. (13) to give the value Eq. (10) requires

$$r_s = 0.0079\ \mathrm{\AA}, \tag{16}$$

which is much smaller than half of any interatomic distance in CSH or any other hydride under pressure [27]. This establishes that CSH cannot be a type-I superconductor. The same applies to all other reported superconducting hydrides since invariably applied magnetic fields of several Tesla give small shifts in the critical temperature. We note that Talantsev and coworkers have also concluded that sulfur hydride and other hydrides are strongly type II [48,49], and Dogan and Cohen concluded the same for CSH [47].

We conclude then that hydrides under pressure, if they are superconductors, are type-II superconductors. The reduction in T_c with applied magnetic field then reflects the upper critical field H_{c2} and not the thermodynamic critical field H_c. Under this assumption, for the case of CSH, extrapolation to zero temperature using either GL theory or other theoretical approaches yields for the zero temperature upper critical field values in the range $H_{c2}(0) = 50\,\mathrm{T}$ to $85\,\mathrm{T}$ [14]. Assuming the value obtained from GL theory $H_{c2}(T = 0) = 62\,\mathrm{T}$ yields from Eq. (4b) a zero temperature coherence length $\xi(T = 0) = 23\,\mathrm{\AA}$. Similar values for ξ are obtained for the other superconducting hydrides.

For type-I superconductors, coherence lengths are at minimum several hundred Å. Coherence lengths of order 20 Å indicate the materials are strongly type II. For example, for the conventional superconductor MgB_2, $\xi \sim 50\,\mathrm{\AA}$ and $\lambda_L \sim 1400\,\mathrm{\AA}$. For YBCO, $\xi \sim 30$ Å and $\lambda_L \sim 1500$ Å. For a rough estimate of λ_L here, we use the BCS formula for the coherence length [45]

$$\xi = \frac{\hbar v_F}{\Delta} = \frac{\hbar k_F}{m^*\Delta} \tag{17}$$

together with the relation

$$k_F = (3\pi^2 n_s)^{1/3} \tag{18}$$

and the London penetration depth given by [45]

$$\lambda_L = \sqrt{m^* c^2/4\pi n_s e^2} \tag{19}$$

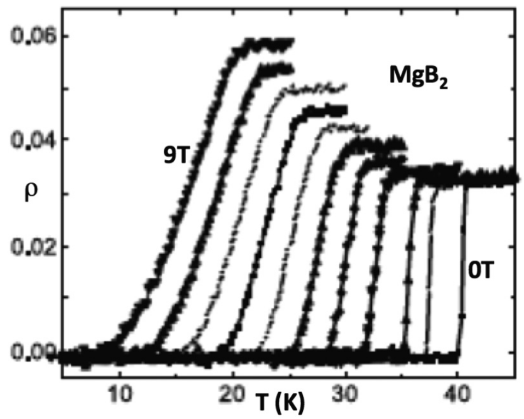

FIG. 2. Resistive transition for MgB_2 in a magnetic field ranging from 0 to 9T, from Ref. [50].

to yield

$$\lambda_L = \sqrt{\frac{3(\hbar c)^6}{4\pi^2 e^2 \Delta^3 (m_e c^2)^2 \xi_0^3}} \left(\frac{m_e}{m^*}\right)$$

$$= 1935\Big(\frac{1}{\Delta(eV)\xi(\text{Å})}\Big)^3 \left(\frac{m_e}{m^*}\right). \qquad (20)$$

In particular for CSH with $\xi = 23$ Å and $\Delta(0) = 0.0437$ eV,

$$\lambda_L = 1906\,\text{Å}\left(\frac{m_e}{m^*}\right) \qquad (21)$$

so that $\lambda_L \gg \xi$ for any reasonable value of m_e/m^*.

IV. RESISTIVE BROADENING IN STANDARD CONVENTIONAL AND UNCONVENTIONAL SUPERCONDUCTORS

In standard conventional and unconventional type-II superconductors it is empirically observed that the resistive transition is broadened in an applied magnetic field, the more so the larger the field. It is also observed that the broadening is larger for materials with higher T_c, which are also strongly

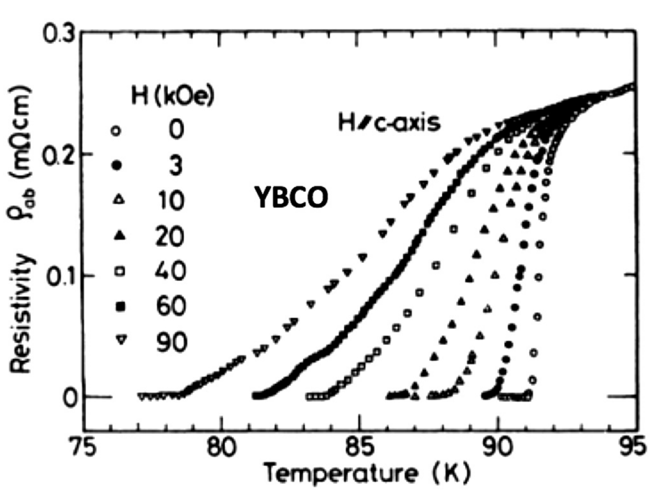

FIG. 3. Resistive transition for YBCO in a magnetic field ranging from 0 to 9 T, from Ref. [51].

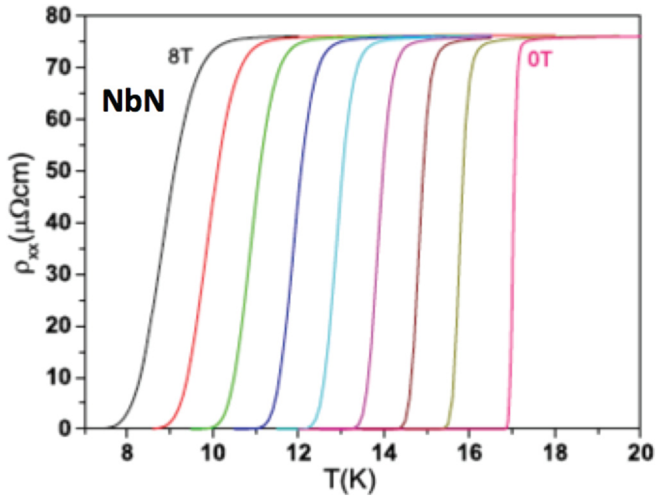

FIG. 4. Resistive transition for NbN in a magnetic field ranging from 0 to 8 T, from Ref. [54].

type II ($\lambda_L \gg \xi$). Typical data for broadening of the resistive transition in standard superconductors are shown in Figs. 2–5 for Mg B_2, NbN, and Mo_xRe_{1-x} (conventional) and YBCO (unconventional), respectively.

As discussed in Sec. II, when a type-II superconductor is cooled in a magnetic field H, it first enters the mixed state, with vortices in its interior. A vortex has a normal core of radius ξ and carries magnetic flux ϕ_0, the flux quantum. Right at $T_c(H)$ (defined as $H = H_{c2}[T_c(H)]$) the number of vortices per unit area is $1/(2\pi\xi^2)$, so that the vortex cores of radius ξ, where the system is normal, almost overlap. As the temperature is further lowered, with constant applied H, the density of the vortices stays roughly constant but their core size shrinks as more electrons condense into the superconducting state.

Figures 2–5 show that as the system enters this mixed state, the resistivity does not drop discontinuously to zero but rather gradually. So there is dissipation as current flows through the mixed state. As the temperature is further lowered, the resistivity drops to zero over a temperature range ΔT that is an increasing function of the applied magnetic field H. This is the expected behavior in all standard type-II

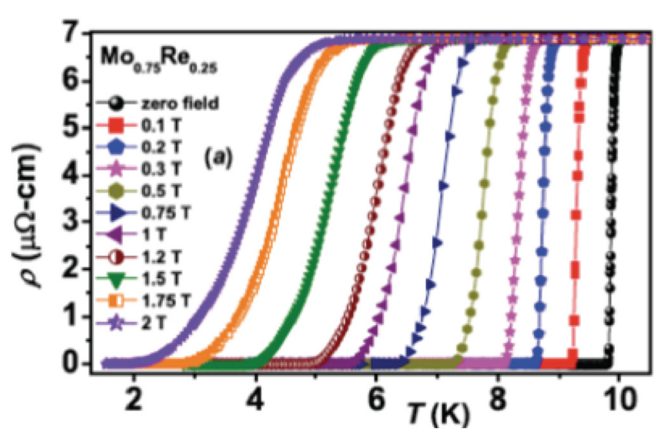

FIG. 5. Resistive transition for Mo–Re alloy in a magnetic field ranging from 0 to 2 T, from Ref. [52].

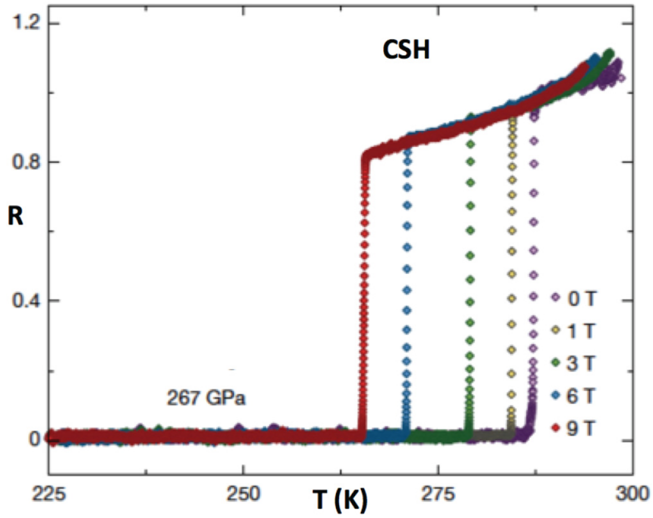

FIG. 6. Resistive transition for CSH in a magnetic field ranging from 0 to 9 T, from Ref. [14].

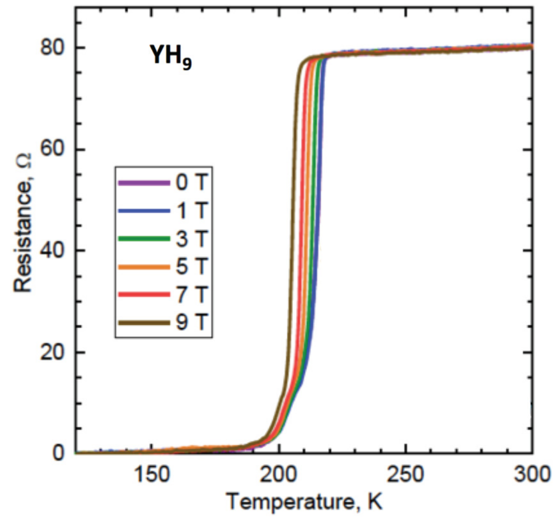

FIG. 7. Resistive transition for YH_9 in a magnetic field ranging from 0 to 9 T, from Ref. [12].

superconductors, which can be understood theoretically using concepts developed more than 50 years ago that apply to both conventional and unconventional superconductors [45]. We review this theory in a following section.

V. EXPECTED VERSUS OBSERVED BROADENING OF THE RESISTIVE TRANSITION IN HYDRIDES

In hydride superconductors, the resistive transition in a field looks qualitatively different to what it looks like in standard superconductors, as shown in Figs. 6–10.

The most glaring difference is seen for the case of the "room temperature superconductor" CSH [14], Fig. 6. The transition is already unusually sharp in the absence of applied magnetic field: T_c drops to zero over a range of temperature not larger than $\Delta T = 0.5\,\mathrm{K}$, hence a fractional broadening $\Delta T/T = 0.0018$. Such sharp transitions are not seen in standard superconductors except for exceptionally pure type-I superconductors. In all the examples shown in Figs. 2–5, the width of the transition at $H = 0$ is at least $\Delta T/T > 0.02$, i.e., an order of magnitude larger. In addition, the transition in CSH remains equally sharp upon application of a magnetic field as high as $9T$. Given the estimated critical field $H_{c2} = 62T$ for this case, $H = 9T$ is 15% of H_{c2}.

For the other examples of hydride superconductors shown (Figs. 7, 8, 9, 10) the transition in the absence of a field is broader, more in line with standard superconductors, but the behavior under application of magnetic field is equally anomalous. Either the transition width does not change with field, or it even narrows, as seen in Fig. 7 for YH_9.

In Fig. 11 we contrast the qualitatively different behavior of standard and nonstandard superconductors, showing several representative examples. The horizontal axis gives H, the applied field, divided by the reported upper critical field for each case. The vertical axis gives the ratio of the broadening of the transition ΔT divided by the temperature T_c where the resistance starts to drop for each applied magnetic field.

For all standard superconductors shown in Fig. 11 top panel, the transition broadens under application of a magnetic field, as expected from the theory discussed in the next section. We looked at many more examples of standard superconductors and the qualitative behavior is always the same. Note that the upward slope is larger for the higher T_c cases, YBCO (unconventional) and MgB_2 (conventional), as expected from the standard theory discussed in the next section. This leads to the expectation that the slope should be even larger for the high T_c hydrides under pressure. Nothing of the sort is seen in the lower panel of Fig. 11.

Instead, we see in the lower panel of Fig. 11 that there is no upward trend in the broadening of the transition with increased magnetic field. Quite the contrary, the broadening

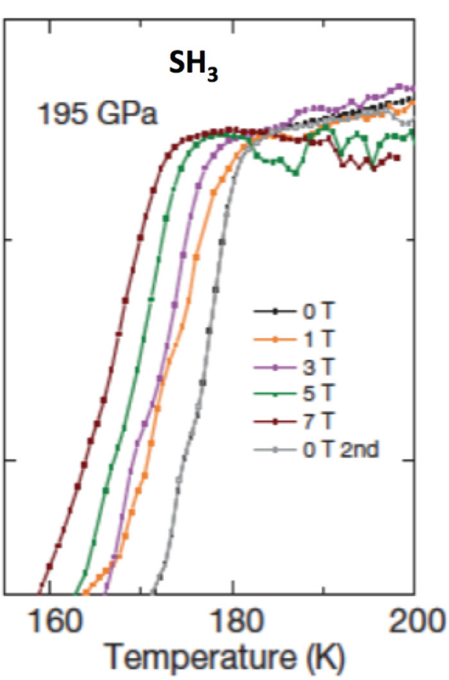

FIG. 8. Resistive transition for SH_3 in a magnetic field ranging from 0 to 7 T, Ref. [7].

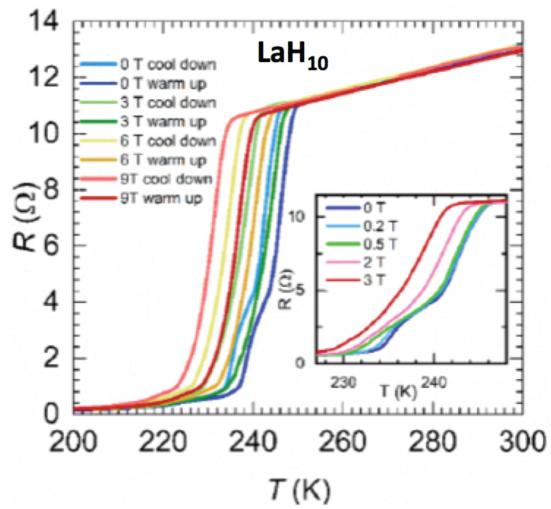

FIG. 9. Resistive transition for LaH_{10} in a magnetic field ranging from 0 to 9 T, from Ref. [9].

either stays the same as for $H = 0$ or even decreases with applied field, as the data for YH_9 show. Note also that the broadening for CSH, both without magnetic field and with an applied magnetic field of up to 15% of the upper critical field, is two orders of magnitude smaller than any of the broadening seen in the top panel for standard superconductors. The data in Fig. 11 clearly show that high temperature hydride superconductors are qualitatively different from standard superconductors as far as the behavior of the resistance versus temperature in an applied magnetic field is concerned. This cannot be accounted for within the standard theory of superconductivity, as discussed in the next section.

VI. RESISTIVE BROADENING UNDER AN APPLIED MAGNETIC FIELD

As mentioned above, when a type-II superconductor is cooled in the presence of an applied magnetic field H, it undergoes a second-order phase transition to a mixed state at temperature $T_c(H)$ that satisfies $H_{c2}[T_c(H)] = H$. Immediately below this temperature, the normal vortex cores in this mixed phase occupy approximately half of the volume of the material according to Eq. (4b). When current flows, vortices are expected to flow driven by the Lorentz force, which is equivalent to the flow of normal current, giving rise to a finite voltage drop across the electrodes and Joule heat dissipation. Hence there is nonzero resistance, called flux-flow resistance. As the temperature is lowered further for fixed magnetic field, the density of vortices stays constant but their core size shrinks, the condensation energy increases and a larger fraction of the material enters the superconducting state. At some lower temperature it is observed that the resistance drops to zero or to immeasurably small values, which is attributed to the vortices becoming pinned at impurities

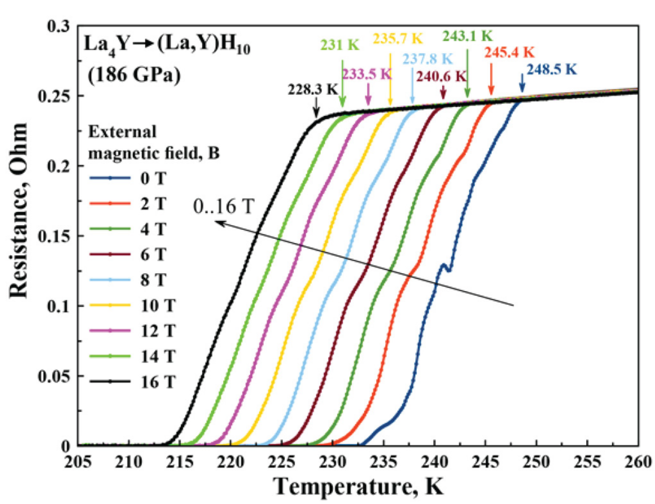

FIG. 10. Resistive transition for $(La,Y)H_{10}$ in a magnetic field ranging from 0 to 16 T, from Ref. [15].

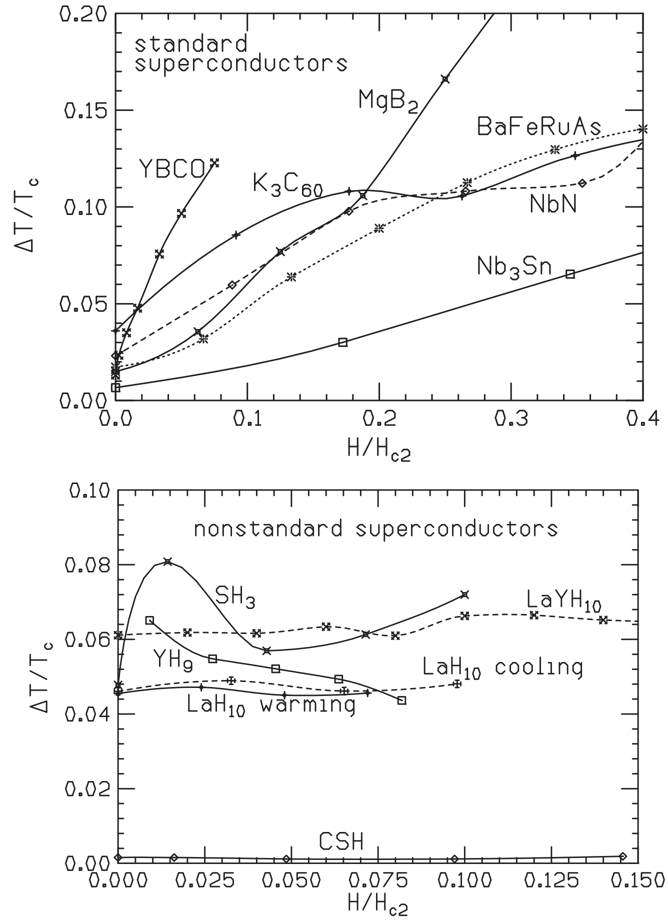

FIG. 11. Upper panel: YBCO single crystal, $H_{c2} = 120\,\mathrm{T}$, $T_c^{max} = 91\,\mathrm{K}$ [51]; MgB_2 polycrystalline, $H_{c2} = 16\,\mathrm{T}$, $T_c^{max} = 40\,\mathrm{K}$ [50]; K_3C_{60} single crystal, $H_{c2} = 17.5\,\mathrm{T}$, $T_c^{max} = 20\,\mathrm{K}$ [53]; NbN thin film epitaxial, $H_{c2} = 11.3\,\mathrm{T}$, $T_c^{max} = 17\,\mathrm{K}$ [54]; Nb_3Sn polycrystalline, $H_{c2} = 29\,\mathrm{T}$, $T_c^{max} = 18\,\mathrm{K}$ [55]; $BaFe_{2-x}Ru_xAs_2$ $x = 0.71$, single crystal, $H_{c2} = 30\,\mathrm{T}$, $T_c^{max} = 20\,\mathrm{K}$ [56]. Lower panel: SH_3, $P = 155$ GPa, $H_{c2} = 70\,\mathrm{T}$, $T_c^{max} = 180\,\mathrm{K}$ [7]; YH_9, $P = 185$ GPa, $H_{c2} = 110\,\mathrm{T}$, $T_c^{max} = 218\,\mathrm{K}$ [12]; LaH_{10} cooling, $P = 150$ GPa, $H_{c2} = 92\,\mathrm{T}$, $T_c^{max} = 245\,\mathrm{K}$ [9]; LaH_{10} warming, $P = 150$ GPa, $H_{c2} = 125\,\mathrm{T}$, $T_c^{max} = 249\,\mathrm{K}$ [9]; CSH, $P = 267$ GPa, $H_{c2} = 61.8\,\mathrm{T}$, $T_c^{max} = 288\,\mathrm{K}$ [14], $(La,Y)H_{10}$, $P = 186$ GPa, $H_{c2} = 100\,\mathrm{T}$, $T_c^{max} = 248.5\,\mathrm{K}$ [15].

or imperfections in the sample, preventing their motion. The strength of the pinning potential needed to pin the vortices also depends on the interaction between vortices: if the interaction is sufficiently large that they become rigidly connected to each other forming a "vortex glass" [57], it becomes easier to pin the entire vortex assembly than if the vortices are pinned independently.

Why should we expect the broadening of the transition to be larger for higher T_c materials, and to increase with increased magnetic field? We can understand it as follows. As discussed by Anderson [58] and Anderson and Kim [59], let us call U_0 the characteristic energy of pinning. Thermally activated depinning will be proportional to $e^{-U_0/(k_BT)}$, so for given pinning energy it will be exponentially larger at higher temperature. For a given vortex to remain stationary, the pinning threshold energy is given approximately by [58]

$$U_0 \sim \frac{H_c}{8\pi}\xi^3, \tag{22}$$

with H_c the thermodynamic critical field. This indicates that the required pinning energy becomes smaller for shorter coherence length; this is the case for more strongly type-II superconductors.

In addition, Yeshurun and Malozemoff argued [60] that when the flux lattice spacing (distance between nearest neighbor vortices) $a_0 \sim (\phi_0/H)^{1/2}$, with H the applied magnetic field, becomes significantly smaller than the London penetration depth, a cross-over to collective pinning should occur where

$$U_0 \sim \frac{H_c^2}{8\pi}a_0^2\xi. \tag{23}$$

Within GL theory Eqs. (6) and (9) give $H_c \sim H_{c0}(1-t)$, $\xi \sim \xi_0(1-t)^{-1/2}$, with $t = T/T_c$ the reduced temperature, and hence Eq. (23) yields

$$U_0 = C\frac{H_{c0}^2}{8\pi}\phi_0\xi_0\frac{(1-t)^{3/2}}{H}, \tag{24}$$

where C is a numerical factor. The pinning energy decreases with increased magnetic field H, which gives larger broadening for larger H.

This realization provided some theoretical support for the work of Müller *et al.* [61], who recognized almost immediately upon discovering cuprate superconductivity that an "irreversibility line" exists in the H–T plane. This line delineates the reversible region from the region where magnetization shows hysteresis. They also found that the irreversibility line had a temperature dependence, $H_{\rm irr} \propto (T_c - T)^{3/2}$. Tinkham [62] then made use of this temperature dependence attributed to the activation energy for flux motion [60] to understand the resistance in this regime, following the Ambegaokar-Halperin [63] theory of the resistance due to thermally activated phase slippage. From Eq. (24), Tinkham argued that the flux pinning barrier height U_0 normalized to k_BT would be given by an expression

$$\gamma_0 \equiv \frac{U_0}{k_BT} = C'\frac{J_{c0}(0)}{T_c}\frac{(1-t)^{3/2}}{H}, \tag{25}$$

where C' is a constant and $J_{c0}(0)$ is the critical current density for $T = 0$ and $H = 0$. Insertion into the Ambegaokar-Halperin expression [63] results in the Tinkham formula

$$\frac{R}{R_n} = \frac{1}{I_0^2[A(1-t)^{3/2}/(2H/H_{c2})]}, \tag{26}$$

where A is a dimensionless constant, R_n is the normal state resistance, and $I_0(x)$ is the modified Bessel function of the first kind. As Tinkham illustrated [62] with the data of Ref. [51] (also shown in our Fig. 3), this formula reproduces these curves in considerable detail with one fitting parameter, $A = 10$. As noted by Tinkham [62], his fitted value of this constant is the correct order of magnitude expected for the YBCO, given the measured value of $J_{c0}(0)$ and the extrapolated value of H_{c2}. This is also true for the other materials (see below) shown in the upper panel of Fig. 11. For our purposes, the important outcome is that for a type-II superconductor, the width of the transition, ΔT, naturally increases with increasing applied field. For the cuprates, following the authors of Ref. [62], one finds

$$\Delta T \propto H^{2/3}. \tag{27}$$

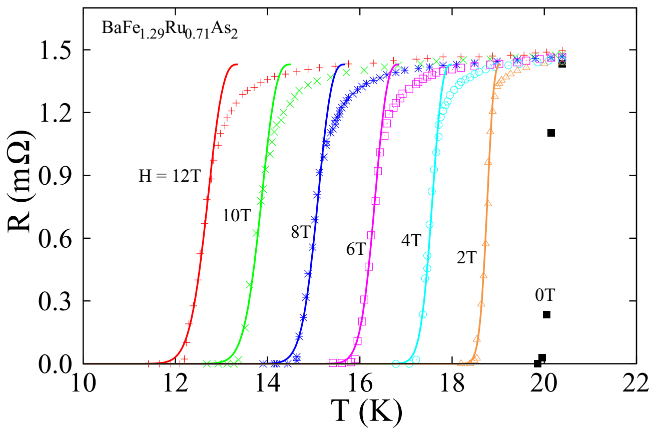

FIG. 12. Resistance vs temperature for $BaFe_{1.29}Ru_{0.71}As_2$ for a variety of applied magnetic fields, as indicated. Experimental data, represented by the points for the various fields, is taken from Ref. [56]. Our theoretical fits are given by curves. We normalized the fits to the normal state resistance at $T_c = 20.3$ K, and used the single fitting parameter, $A = 83.3$, as discussed in the text. We used $T_c(H) = 19.05, 18.0, 16.85, 15.68, 14.513.36, 12.0$ K for $H = 2, 4, 6, 8, 10, 12$ T, respectively. These form a linear dependence of $H_{c2}(T)$ and extrapolate to $H_{c2}(0) = 25$ T (WHH theory [64]) or 35 T (two-fluid model).

The broadening that occurs in the Iye data [51] is visually obvious. We showed other systems that clearly exhibit similar broadening. Even where it is not so obvious, for example, in the pnictides [56] [see their Fig. 1(a)], the Tinkham formula works very well in reproducing the broadening trend illustrated in Fig. 12. We generalized this formula slightly by using $t \equiv T/T_c(H)$. An H-dependent T_c was not required in the case of YBCO. The result is shown in Fig. 12, again with a single fitting parameter, $A = 83.3$ in this case. We actually used an optimal fit for $T_c(H)$ in obtaining the curves shown, but when plotting the resulting H_{c2} versus T_c, we obtain a straight line near $T_{c0} \equiv T_c(H = 0)$, as expected in the two-fluid model (or any other more sophisticated model). We could have started

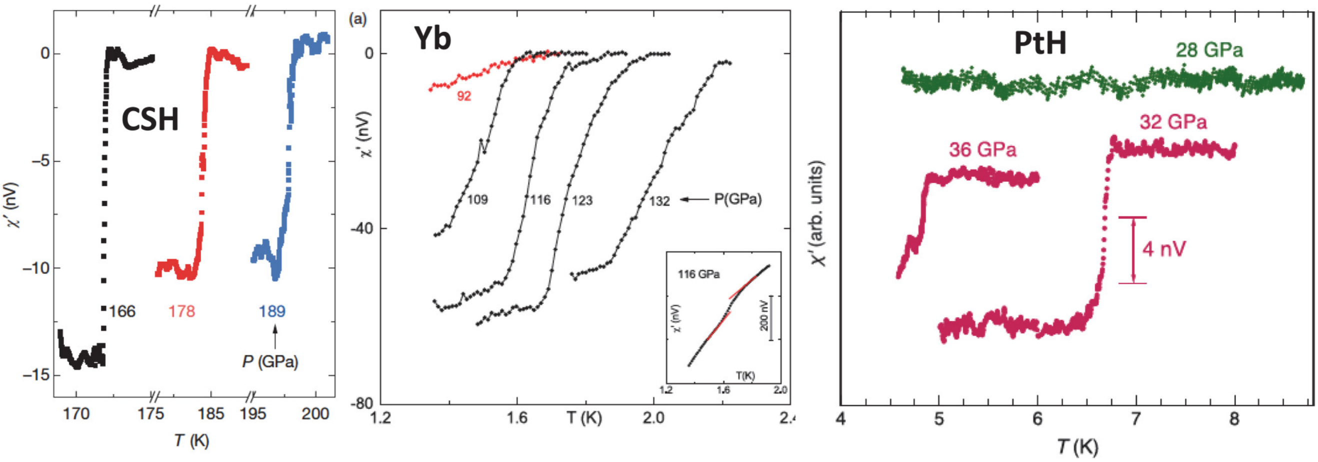

FIG. 13. Ac magnetic susceptibility data for materials under pressure. Left panel: CSH, from Ref. [14]; middle panel: Yb, from Ref. [69]; right panel: PtH, from Ref. [70]. The width of the transition is $\Delta T/T_c \sim 0.03$ for PtH ($P = 32$ GPa), $\Delta T/T_c \sim 0.11$ for Yb ($P = 116$ G Pa) and $\Delta T/T_c \sim 0.001$ for CSH ($P = 166$ GPa).

with this curve, dictated by the experiment, so, for this reason we do not regard these as fitting parameters. In fact, we consider this self-consistency a further check of the theory. Note that a significant amount of broadening occurs, and the Tinkham formula captures the majority of this broadening. As is clear from Fig. 12 there is an additional "rounding" of the resistivity that the Tinkham formula does not capture, but nonetheless increases with increasing applied magnetic field. We do not pursue the origin of this additional rounding here.

The previous expression assumes that the intrinsic width, with no applied magnetic field, H_0, is zero, but in general this is not the case for a variety of possible reasons such as disorder in the sample. To take into account this intrinsic width we substitute in Tinkham's formula Eq. (26) $1 - t = \Delta T/T_c$ by $\Delta T/T_c - \Delta T_0/T_c$, where ΔT_0 is the intrinsic broadening for $H = 0$. The behavior seen in all the examples shown in Fig. 11 upper panel can then be fitted semiquantitatively with the parameter A in Eq. (25) ranging from 10 for YBCO to 257 for Nb_3Sn ($A = 42.4, 45.1, 38.9$ for MgB_2, NbN, K_3C_{60}, respectively). Instead, the behavior shown in the lower panel of Fig. 11 for CSH cannot be fitted to the Tinkham formula Eq. (25) unless $A > 100\,000$. Given the values of T_c and H_{c2} for CSH, then, following Tinkham's analysis for YBCO [62], this implies a zero temperature critical current density in excess of 10^{11} A/cm^2. The occurrence of such a large critical current density should be easily tested by experiment. For YH_9 the fitting fails completely since the width of the transition decreases instead of increasing with magnetic field. We conclude that if hydrides under pressure are truly superconductors, they do not obey the same physical principles that all other type-II superconductors, whether conventional or unconventional, obey.

VII. MAGNETIC MEASUREMENTS

In addition to transport measurements, some of the experimental papers on high temperature hydride superconductors also report results of magnetic measurements that appear to support the claim of superconductivity. Let us consider those measurements. The authors of Ref. [14] showed results for ac magnetic susceptibility measurements in CSH that indicated a sharp drop at certain temperatures, that the authors claimed was consistent with onset of superconductivity. First, it should be pointed out that the magnetic measurements are not done during the same run where the transport data are collected due to experimental constraints, and as a consequence it is not possible to verify whether the signals indicating superconductivity show up at the same temperatures in transport and magnetic data. In fact, the authors of Ref. [14] commented that the susceptibility signal occured at somewhat higher temperature than the transport signal, which they attributed to uncertainties in the estimated pressures due to pressure gradients. Furthermore, the transitions shown in the susceptibility data were also anomalously sharp. For example, the width of the transition for 166 GPa is merely $\Delta T = 0.2K$, hence $\Delta T/T_c = 0.001$. Typical ac susceptibility measurements for materials under pressure in diamond anvil cells show much broader transitions [65–70]. We show some examples in Fig. 13. Given that the magnetic penetration depth is much larger than the coherence length in CSH and other hydrides, and that half of the sample remains in the normal state at the onset of superconductivity, one would expect a much broader transition in the magnetic susceptibility than seen in the left panel of Fig. 13. The same is true for ac magnetic susceptibility data for other hydrides [71]. Other reasons for why the ac magnetic susceptibility measurements on CSH may not indicate superconductivity were discussed in Ref. [38].

In Ref. [72], it was claimed that the Meissner effect was detected in sulfur hydride through a novel nuclear resonant scattering experiment, and hence that the existence of superconductivity was unequivocally confirmed [72,73]. However, we challenged that claim in Ref. [74]. We showed that a standard superconductor, whether conventional or unconventional, would not show the behavior reported in Ref. [72]. More specifically, that to show that behavior the critical current would have to be enormous, qualitatively larger than for standard superconductors, similarly to what we found in this paper from consideration of the width of the resistive transition. We concluded in Ref. [74] that either the observations reported in Ref. [72] were experimental artifacts and hence provided no evidence for superconductivity, or alternatively, that they provided further evidence that hydride superconductors are nonstandard superconductors, fundamentally different from standard superconductors.

VIII. DISCUSSION

In this paper we consider the question of whether superconductivity in hydrides under high pressure follows the expected behavior of other superconductors. We find that the answer is negative.

Our finding is remarkable because hydride superconductors are considered to be textbook examples of conventional BCS-Eliashberg superconductors driven by the electron-phonon interaction, which unlike unconventional superconductivity, has been thought to be well understood for the last 60 years. All the theoretical analysis of these materials conclude that their T_c is perfectly well explained by standard superconductivity theory [16–28]. How is it possible then that the behavior of resistivity in the presence of a magnetic field does not follow the standard behavior seen in type-II superconductors [45]? How is it possible that magnetic measurements also show anomalous behavior incompatible with standard superconductivity?

The only conceivable explanation within the standard theory for the absence of broadening of the resistive transitions in a magnetic field would be that for some reason there is an enormous pinning potential that confines the vortices immediately as the system enters the superconducting state, much larger than in standard superconductors. If that was the case, it would also be true that the critical current should be enormous in these superconductors, opening up the way for important technological applications. Unfortunately, we know of no physical reason why these materials should have such anomalously large pinning potentials. Furthermore, even if that was the case it would not explain the extreme sharpness of the transition in magnetic susceptibility measurements, which is not affected by the strength of the pinning potential.

We conclude then that there are two possibilities.

(1) These materials constitute a new category, "nonstandard superconductors," exhibiting physics that is qualitatively different from that of standard conventional and unconventional superconductors. Perhaps these materials are simultaneously type-I and strongly type-II superconductors, exhibiting a new kind of "complementarity." Recall that for standard type-I superconductors the transition to superconductivity in a field is first order, while for type II it is second order. Perhaps even the classification of type I versus type II does not apply to them. Perhaps the coherence length and the London penetration depth are not the relevant lengths. Their electrodynamic behavior needs to be understood in a new theoretical framework, unlike the one formulated for standard superconductors. If this is the case it is very remarkable that the conventional framework of BCS-Eliashberg theory would still apply to them with no modification, as found in the theoretical analyses [16–28].

(2) These materials are not superconductors. Their measured properties that were interpreted as reflecting superconductivity were instead reflecting either (a) experimental artifacts or (b) different physics or (c) a combination of both. Let us consider these possibilities.

The possibility that experimental artifacts may be responsible for all or some of the signals mistakenly interpreted as superconductivity cannot be ruled out *a priori* in any experiment. Examples in the scientific literature where claims of superconductivity were subsequently withdrawn abound. The strong desire to find superconductivity in a compound may cause careful researchers to misinterpret the origin of observations. We feel that research in high pressure hydride superconductivity may have reached the stage where theoretical bias is unduly guiding the interpretation of experimental findings. [2–6]

With respect to different physics, quite generally the sharpness of the signals observed suggests that if they indicate phase transitions they are first rather than second-order transitions. We suggested in Ref. [46] that the abrupt increases in resistance found upon heating may result from metallic paths being destroyed as the temperature is increased. This could result from atomic rearrangements due to a local first-order phase transition between phases that are close in free energy but have very different electrical conductivities. This was also recently suggested by Dogan and Cohen [47]. The fact that the transitions shift to lower temperature upon application of a magnetic field could result from the fact that as the magnetic field is applied, Joule heating of the sample produced by eddy currents may randomly rearrange atomic positions. Cases where such processes may result in increase rather than decrease of "critical temperature" may be disregarded as spurious because they do not conform to the expected behavior predicted by theory.

Alternatively, we suggested in Ref. [38] that other different physics could be the onset of weak ferromagnetic order. This is supported by the raw data in susceptibility measurements reported in Ref. [14], and would be in agreement with alternative theoretical expectations [39]. The authors of Refs. [40] and [47] also suggested that magnetic effects may account for some of the observations, and they also suggested alternative possibilities not discussed here.

In conclusion, more experimental and theoretical work is needed to establish what is going on in these materials.

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