High Pressure RTSC-Hydrides are Extreme Hard Type-II Superconductors

1,2Miodrag L. Kulic
1Institute for Theoretical Physics, Goethe-University Frankfurt am Main, Germany
2Institute of Physics, 11080 Belgrade, Serbia

(Dated: April 27, 2021)

In[1] the authors have called into question the existence of the room-temperature superconductivity (RTSC) in hydrides, as well as the electron-phonon interaction as the pairing mechanism. Basically, in [1] they assume, that these materials are soft type-II superconductors with point-like defects as pinning centers. We show here, that RTSC - hydrides are hard type-II superconductors, with strong pinning centers in form of long columnar defects, with length \(L_{col}\) much larger than the superconducting coherence length \(\xi(T)\) and penetration depth \(\lambda(T)\). It is shown that the elementary pinning energy for the vortex lying along the long columnar defect is given by \(U_{col} \approx U_{point}(L_{col}/\xi)\).

At low temperatures it gives rise to high critical current density \(j_c \sim 10^5 \text{A/cm}^2\), which is of the order of depairing current in hydrides. This theory predicts, that the temperature width of the resistivity broadening (TRB), calculated in the Tinkham theory, depends on \(L_{col}\), i.e. \(\Delta T \sim (B\xi_0/L_{col})^{1/2}\).

Since \((\xi_0/L_{col}) < 10^{-5}\), this width \(\Delta T = T_c(1-t)\) is at least 100 times smaller in RTSC-hydrides, than that predicted in [1] for standard superconductors. However, in the C-S-H superconductor with \(T_c = 287\text{ K}\) the experimental TRB is almost field independent, which probably needs a revision of the Tinkham theory. In an extreme hard type-II superconductor irreversible phenomena are pronounced and the penetration (trapping) of the field in and the magnetization hysteresis are treated in the model of the Bean’s critical state with long columnar pinning centers. For instance, by assuming that the collective pinning is realized in fields \(H > H_I\), one expects a significant shift up of the irreversibility line \(B \sim (1 - t)^2(L_{col}/\xi)\). Due to the intrinsic hardness of the RTSC-hydrides they are promising candidate for producing high critical currents, especially if they would be realized in systems which are in metastable state at low pressure.

I. INTRODUCTION

The first room-temperature superconductor was reported in 2015 in sulphur-hydrides (\(H_3S\)) under high pressure \(P \approx 150\text{ GPa} (=1.5\text{ Mbar})\) [3]. This opened a new frontier in physics and number of other RTSC-hydrides were synthesized. Let us mention some of them with \(T_c \geq 200\text{ K}\), for instance \(LaH_1\nu\text{with} T_c \approx 250\text{ K}\) [4], LaYNb with \(T_c \approx 253\text{ K}\) [5], C-S-H with \(T_c = 287\text{ K}\) at \(P \approx 276\). In that sense, the most promising candidate for the highest \(T_c\) is the metallic hydrogen (\(H\)) which is the lightest atom in nature. One expects, that in the metallic crystal state it could give as high as \(T_c \sim 600\text{ K}\), as it is theoretically predicted by Maksimov and Savrasov in their seminal (but rarely cited!) paper in 2011 [2]. The pairing in the hydrogen-based systems is certainly due to the strong electron-phonon interaction (EPI) via high-frequency \(H\)-phonons. The pronounced isotope effect with \(\alpha \sim 0.3\) (in the pure EPI theory for light atoms, such as \(H\), even without effects of Coulomb \(\mu^*\) term, \(\alpha\) is smaller than the bookkeeping value 0.5 (!)), is a clear evidence for the EPI pairing in RTSC-hydrides - see [7], [8]. More on the pairing mechanism, especially on the role of the EPI, in RTSC-hydrde and other high-temperature superconductors such as cuprates and Fe-based materials will be published elsewhere [8].

Many properties of RTSC-hydrides are unusual, since it turns out, that they are extreme hard type-II superconductors with \(\lambda \gg \xi\), which can not be explained by the theory of soft superconductors. Namely, in soft superconductors vortices are mainly pinned by randomly distributed “point” defects [9], [10], where the elementary pinning force of a “point” defect is due the gain in the condensation energy when the vortex core is sitting on the defect. The mechanism is simple, since in the “point” defect, with the volume \(V_c\), the superconductivity is completely destroyed (with \(\Delta = 0\)) and the condensation energy \(E_{\text{loss}} \sim (H\xi^2/8\pi)\) lost. So, if the part of the vortex (which core of the radius \(\xi\) is in the normal state and the condensation energy is also lost) is sitting on the “point” defect, then the loss of the condensation energy is minimized. However, in order to maximize the pinning force the radius of the defect should be \(\sim \xi\) and \(V_c \sim \xi^3\) (very small defects with \(d \ll \xi\) are inappropriate since \(V_c \sim \xi^2\) is small). This means that the gain of the energy is small due the smallness of the “point” defect, while the Lorenz force acts on the whole vortex line. As the result the critical current of the single “point” defect is small. On the other side, in the presence of large densities of the “point” defects (which are uncorrelated) two facts are against strong pinning. First, vortices can deform in the zig-zag form, thus gaining pinning energy, but the zig-zag vortex line increases its elastic energy. Additionally, each vortex interacts with many (on the average uniformly) distributed defects, thus giving on the average zero pinning force on it. However, there is still pinning due to the local fluctuations of defects density but the pinning force is small, since the maximum pinning force on the volume \(V_c\) is \(F_{\text{pin}} = f_p\sqrt{N}/V_c\). Here, \(N = nV_c\) is the number of the pinning centers in the volume \(V_c\) and \(f_p\) is the elementary pinning force. The critical current density is given by \(j_cB = f_p\sqrt{N}/V_c = f_p(n/V_c)^{1/2}\) [9].
Therefore in order to obtain larger critical current it is desirable to have long (of length L) defects as pinning centers, with the large pinning volume \( V_{\text{d}} = \pi \xi^2 L_{\text{col}} \) and the pinning energy \( U_p = u_p V_{\text{col}} \). In the case of the vortex (with length \( L_v > L_{\text{col}} \)) sitting on such a defect the critical current density is

\[
  j_c = \frac{c}{\Phi_0} \times \frac{u_p V_{\text{col}}}{\xi L_v} = \frac{c}{\Phi_0} \times \left( \pi u_p \xi \right) \frac{L_{\text{col}}}{L_v}. \tag{1}
\]

It is clear, that in order to optimize the critical current density it is desirable to have defects with the same length as the vortex line, i.e. \( L_{\text{col}} \approx L_v \) [13] and with the maximal possible density of the pinning energy \( u_p \).

Let us mention, that in the high-temperature superconductors the long pinning defects were realized in the form of long columnar defects by irradiating \( \text{YBa}_2\text{Cu}_3\text{O}_7 \) single crystals with 580 MeV \( ^{116}\text{Sn}^{30+} \) ions, which produced long tracks with the length \( L_{\text{col}} \approx 20\mu m \) and the diameter 50\( \mu m \). In that case \( j_c \) reached \( 1.5 \times 10^7 \text{A/cm}^2 \) at \( T = 5K \) and \( 10^8 \text{A/cm}^2 \) at \( T = 77K \) [11], which are much higher than in the case of “point” defects. Also, the irreversible lines \( B_{irr}(T) \) lie much higher than in the case of the “point” defects.

In the following we argue, that the vortex pinning by such long defects is intrinsic property of the RTSC-hydrides. First, we introduce the model of vortex pinning by long columnar defects where both, the core and electromagnetic pinning, contribute almost equally to \( u_p \). This is the most optimal situation for pinning. Next, the model is applied to some irreversible properties in the RTSC-hydrides, such as temperature resistivity broadening in magnetic field, critical current in the Bean-model as well as hysteresis in magnetization.

II. PINNING OF VORTICES BY LONG COLUMNAR DEFECTS

In Fig.1 are shown schematically columnar defects - blue coloured cylinders, which strongly pin vortex lines in RTSC-hydrides, giving rise to large magnetization hysteresis and critical current density in \( H^3S \) see [12]. For simplicity we first analyze isotropic systems [13]. It turns out that for the optimal pinning superconducting is destroyed inside the defect, i.e. \( \Delta = 0 \) and the radius of the columnar defect should be small \( \xi < R \ll \lambda \) [13]. In this case both mechanism of pinning, the core and electromagnetic one, are operative and with almost the same energy [13], \( U_p = U_{\text{core}} + U_{\text{em}} \approx 2U_{\text{cor}} \), where \( U_p \approx 2\pi \xi^2 L_{\text{col}} (H^2/8\pi) = 2L_{\text{col}} \Phi_0^2/(8\pi \lambda)^2 \). Since the pinning force (per unit vortex length \( L_v \)) \( f_p = U_p/\xi L_v \) in the critical state is balanced with the Lorentz force (per unit length) \( f_L = j_c \Phi_0/c \) then the critical current density is given by [13]

\[
  j_c = \frac{c\Phi_0}{32\pi^2 \lambda^2 \xi} \left( \frac{L_{\text{col}}}{L_v} \right). \tag{2}
\]

It is obvious that the critical current is maximized when the length of columnar defect is equal to the vortex length, i.e. \( L_{\text{col}} = L_v \). In the case of anisotropic superconductors when the axis of the columnar defect and the vortex are parallel to the crystal axis \( \gamma \) and the current flows along the \( \beta \)-axis (but \( \alpha \neq \beta \neq \gamma \)) it holds [13]

\[
  j_c^{\alpha,\beta} = \frac{c\Phi_0}{32\pi^2 \lambda_\alpha \lambda_\beta \xi_\alpha} \left( \frac{L_{\text{col}}}{L_v} \right). \tag{3}
\]

If one assumes that in RTSC-hydrides at \( T \ll T_c \) one has \( \lambda \sim 10^3\AA \) and \( \xi_0 \sim 20\AA \), then \( 10^7 (A/cm^2) < j_c < 10^8 A/cm^2 \), which is of the order of depairing current density.

III. TEMPERATURE BROADENING OF RESISTIVITY IN MAGNETIC FIELD

The papers by Hirsh and Marsiglio [1, 14] are stimulating in understanding the physics of superconductivity in the RTSC-hydrides and therefore deserve tribute, in spite of some controversing claims. Namely, they were first to pay attention to some very pronounced differences in properties of RTSC-hydrides and standard superconductors, by including HTSC-cuprates and Fe-based superconductors. For instance they point, that the temperature broadening of resistivity (TBR) \( \Delta T \) in several RTSC-hydrides is much smaller than in standard superconductors by at least two order of magnitude. If one interprets this fact in the framework of soft superconductors it is necessary to invoke unrealistically high critical current density of the order \( 10^{11} A/cm^2 \). Here, we argue that this effect can be explained by assuming that RTSC-hydrides are hard type-II superconductors with many intrinsic long pinning defects. Let us briefly introduce the reader into the subject, which is based on the Tinkham theory for TBR [17].

Practically in all superconductors there are some defects which pin vortices allowing a dissipationless current to flow even in the vortex state. However, when the pinning energy is small there is a possibility that vortices under temperature fluctuations jump from one center to another thus giving rise to vortex motion and dissipation of energy. This jumps are activation-like and proportional to the escape (from the pinning center) probability \( \exp(-U_p/T) \) - this is so called flux creep [15]. Since for small defects \( U_p \sim \xi^3 \) then this energy barrier is small in superconductors with small \( \xi \) what is the origin of pronounced dissipation in HTSC-cuprates due to small \( \xi \). In that sense, a long pinning defect with \( U_p \sim L_{\text{col}}\xi^2 \) make this barrier much higher, thus suppressing the dissipation effects.

In magnetic fields much higher than the lower critical field \( H_{c1} \ll H < H_{c2} \) one has \( B \approx H \) and the vortex distance is given by \( a \approx l(\Phi_0/B) \ll \lambda \) and the so called collective pinning of vortices should work (at least it was useful for HTSC-cuprates) - where the bundle of vortices are pinned. In that case, one has \( U_p \sim L_{\text{col}}a^2 \) (instead of...
FIG. 1: Schematic view of the experiment of the flux trapping and penetrating in the H$_3$S sample [12]. $D = 30\mu m; P = 5\mu m$. The non-superconducting Sn foil (yellow) for detection of the penetrated magnetic field: $d = 20\mu m; p = 2.6\mu m$. Long columnar defects (blue cylinders) strongly pin and trapp vortices making huge magnetization hysteresis and critical current density $j_c \sim \Delta M$.

FIG. 2: Low temperature electric resistance in magnetic field: H=0T, 1T, 3T, 6T and 9T (increasing from right to left) at 267 GPa - from [8]. Temperature broadening of resistivity is extremely small due to strong pinning by columnar defects.

$U_p \sim L_{col} \xi^2$ for the single vortex pinning. The Thinkam developed the TRB theory which is based on the Ambegaokar and Halperin theory for the thermaly activated phase motion in Josephson junctions [18]. As the result one obtains the formula for the resistance in superconductors for small currents [17]

$$R/R_N = [I_0(\gamma_0/2)]^{-2}, \gamma_0 = U_p/T. \tag{4}$$

$R_N$ is the resistance of the normal state at $T_c$. The modified Bessel function $I_0$ gives the asymptotics for $R/R_N \sim \gamma_0^3$ for $\gamma_0 < 1$ and $R/R_N \sim \gamma_0 \exp(-\gamma_0)$ for $\gamma_0 \gg 1$ [17], where $\gamma_0$ is the dimensionless pinning barrier with respect to temperature. In the model with long columnar pinning centers and near $T_c$ one has

$$\gamma_0^{col} = \beta_K \frac{L_{col}}{\xi_0} \frac{(1 - t)^2}{b} \left(2\pi \xi_0^2 \frac{j_c(0)\Phi_0}{cT_c}\right), \tag{5}$$

where $b = B/H_{c3}(0)$ and $\beta_K \approx 1$. In the case of the “point” defects one has

$$\gamma_0^st \approx \frac{(1 - t)^{3/2}}{b} \left(2\pi \xi_0^2 \frac{j_c(0)\Phi_0}{cT_c}\right), \tag{6}$$

where $\gamma_0^st$ is the prediction of the standard Thinkam theory. From the above equations it is seen that relative barrier in the long columnar case is larger by factor $L_{col}/\xi(T)$, than the one for the “point” defects. Let us compare the prediction of the standard theory [17] with the new one. Let us assume, that we analyze the case when the resistance is measured at the 10% level, i.e. $R/R_N = 0.1$ for the field $B \approx 1T$ and $H_{c3}(0) \approx 100T$, i.e. $b = 10^{-2}$. This means that $\gamma_0^{col} = \gamma_0^st \approx \gamma_0$. Since, $I_0(\gamma_0/2) = 3$ one has $\gamma_0 \approx 5$. We assume the following realistic parameters for the RTSC-hydrides: $\xi_0 \approx 20\AA$, and $j_c \approx 10^8 A/cm^2$ (in cgs units $j_c \approx 3 \times 10^7 esu/cm^2$, $T_c \approx 200K$, $\Phi_0 \approx 2 \times 10^{-7} G \times cm^2$, $L_{col} \approx 20\mu m$. As the result we obtain for $(\Delta T)/T_c \approx 2 \times 10^{-2}$ and the ratio

$$\frac{(\Delta T)^{col}}{(\Delta T)^{st}} \sim 5 \times 10^{-2}. \tag{7}$$

This result means, that by assuming long columnar pinning defects the temperature broadening of the resistance is much smaller than in standard superconductors. This is encouraging result having in mind that the Thinkam theory might be not quite appropriate for all RTSC-hydrides. This is demonstrated in Fig.2 where the line broadening does not depend on the magnetic field. It might be, that the collective pinning theory with $a \sim (\Phi/B)^{1/2}$ is not guaranted in systems with long and strong pinning defects. This is a matter for further investigation.

IV. DISCUSSION AND CONCLUSIONS

That the RTSC-hydrides are hard type-II superconductors with strongly pinned vortices in magnetic field is clearly shown in an inventive experiment by Troyan et al. [12]. In the setup shown schematically in Fig.1 they have intended to study the Meissner effect in $H_3S$ hydride. It was expected that in the magnetic field $H = 0, 68T$, which is much larger than the first critical field $H_{c1}$, that the vortices penetrate into the sample, what should be detected by the NRS technique in the non-superconducting Sn foil (yellow in Fig.1). However, even in such a large field the vortices do not show up in the Sn foil. This means that they are trapped in the $H_3S$ sample, where the magnetization is highly inhomogeneous and hysteretic. This is strong evidence
that the pinning in $H_3S$ is huge. By using the Bean critical state model and the fact that vortices penetrate into the sample only the distance $(D - d)/2$ and that they are pinned along some long defects with the length $P = 50 \mu m$, one obtains that the critical current density is of the order $j_c \sim (10^7 - 10^8) A/cm^2$. To this point, recently has appeared very interesting arXiv preprint by Hirsh and Marsiglio [14], were they have realized that their interpretation of the Troyan’s paper as the Meissner effect was inadequate. Namely, if it were true this would mean that the critical current density should be enorm $\sim 10^{11} A/cm^2$, what is impossible. Since there is a pronounced magnetization hysteresis they propose the existence of strong pinning in the system. They predict much smaller but realistic critical current density $\sim 10^7 A/cm^2$.

In this paper we assume the existence of long pinning defects in the columnar form, and the pinning of vortices take place on long defects. As the result of the strong pinning in RTSC-hydrides, the temperature broadening of the resistance in magnetic field is drastically smaller than the standard theory predicts. This reduction is due to the small factor $\xi_0/L_{col}$. These columnar defects cause also large magnetization hysteresis $\Delta M$ - see [12] and accordingly high critical current density $J \sim \Delta M$. The irreversible line of the magnetic has the form $H \sim (1 - t)^2 L/\xi_0$, i.e. it is drastically increased compared to the situation in HTSC-cuprates.

There is an intriguing question - what is the physical origin for these long defects in RTSC-hydrides? Answering on this question will probably open a new physics in hard type-II superconductors.

Acknowledgments

The author would like to thank Professor Dirk Rischke for his support, both scientific and moral, which has lasted a very long time. I am thankful to Dr Igor Kulič for discussions and support.

[1] J. E. Hirisch. F. Marsiglio, Phys. Rev. B 103, 134505 (2021); arXiv: 2101.01701
[2] E. G. Maksimov, D. Yu. Savrasov, Solid State Comm. 119, 569 (2011)
[3] A. Drozdov, M. Eremets, I. Troyan, V. Ksenofontov, S. Shylin, Nature 525, 73 (2015)
[4] M. Somayazulu, M. Ahart, A. K. Mishra, Z. M. Geballe, M. Baldini, Y. Meng, V. V. Struzhkin, and R. J. Hemley, Phys. Rev. Lett., 122:027001, (2019); A. P. Drozdov, P. P. Kong, V. S. Minkov, S. P. Besedin, M. A. Kuzovnikov, S. Mozaffari, L. Balicas, F. F. Balakirev, D. E. Graf, V. B. Prakapenka, E. Greenberg, D. A. Knyazev, M. Tkacz, and M. I. Eremets, Nature, 569:528, (2019)
[5] D. V. Semenok et al., arXiv:2012.04787 (2020)
[6] E. Snider et al., Nature 586, 373 (2020)
[7] L. P. Gorkov, V. Kresin, Rev. Mod. Phys. 90,011001-1 (2018)
[8] M. L. Kulič, review in preparation
[9] A. I. Larkin, Yu. N. Ovchinnikov, J. Low. Temp. Phys. 34, 409 (1979)
[10] Supercon. Sci. Technol. 10, A11-A28 (1997)
[11] L. Civale et al., Phys. Rev. Lett. 67. 648 (1991)
[12] I. Troyan et al., Science 351, 133 (2016)
[13] M. L. Kulič, A. Krämer, K. D. Schotte, Solid State Commun. 82, 541 (1992); A. Krämer et al. 48, 9673 (1993; A. Krämer et al. 50, 9484 (1993)
[14] J. arXiv: 2104.03925v2.
[15] P. W. Anderson, Phys. Rev. Lett. 9, 309 (1962)
[16] E. F. Talantsev et all., Annal. Phys.(Berlin) 529, 1600390 (2016); Matter. Res. Express, 6, 106002 (2019); J. E. Tallon, E. F. Talantsev, J. Supercond Nov Mag, 31, 619 (2018)
[17] M. Tinkham, Phys. Rev. Lett. 61, 1658 (1988)
[18] V. Ambegaokar, V. I. Halperin, Phys. Rev. Lett. 22, 1364 (1969)
[19] Flores-Livas et al., Phys. Rev. B. 93, 020508
[20] M. Einaga et al., Nat. Phys. 12, 835 (2016)