



# Article Quantifying Nonadiabaticity in Major Families of Superconductors

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Abstract: The classical Bardeen–Cooper–Schrieffer and Eliashberg theories of the electron–phononmediated superconductivity are based on the Migdal theorem, which is an assumption that the energy of charge carriers,  $k_B T_F$ , significantly exceeds the phononic energy,  $\hbar \omega_D$ , of the crystalline lattice. This assumption, which is also known as adiabatic approximation, implies that the superconductor exhibits fast charge carriers and slow phonons. This picture is valid for pure metals and metallic alloys because these superconductors exhibit  $\frac{\hbar\omega_D}{k_BT_F}$  < 0.01. However, for *n*-type-doped semiconducting SrTiO<sub>3</sub>, this adiabatic approximation is not valid, because this material exhibits  $\frac{\hbar\omega_D}{k_B T_E} \cong 50$ . There is a growing number of newly discovered superconductors which are also beyond the adiabatic approximation. Here, leaving aside pure theoretical aspects of nonadiabatic superconductors, we classified major classes of superconductors (including, elements, A-15 and Heusler alloys, Laves phases, intermetallics, noncentrosymmetric compounds, cuprates, pnictides, highly-compressed hydrides, and two-dimensional superconductors) by the strength of nonadiabaticity (which we defined by the ratio of the Debye temperature to the Fermi temperature,  $\frac{T_{\theta}}{T_{F}}$ ). We found that the majority of analyzed superconductors fall into the  $0.025 \le \frac{T_{\theta}}{T_F} \le 0.4$  band. Based on the analysis, we proposed the classification scheme for the strength of nonadiabatic effects in superconductors and discussed how this classification is linked with other known empirical taxonomies in superconductivity.

**Keywords:** nonadiabatic effects in superconductors; Heusler alloys; Laves phases; magic-angle twisted bilayer graphene; hydrogen-rich superconductors

## 1. Introduction

The majority of experimental works in superconductivity utilize the classical Bardeen– Cooper–Schrieffer (BCS) [1] and Migdal–Eliashberg (ME) [2,3] theories as primary tools to analyze measured data. However, it should be clarified that these theories are valid for superconductors which satisfy the condition designated by the Born–Oppenheimer–Migdal approximation [4]:

$$\frac{\hbar\omega_D}{k_B T_F} = \frac{T_\theta}{T_F} = \frac{88 \text{ K}}{1.1 \times 10^5 \text{ K}} \bigg|_{Ph} = 8 \times 10^{-4} \ll 1$$
(1)

where  $\hbar$  is the reduced Planck constant,  $\omega_D$  is the Debye frequency,  $k_B$  is the Boltzmann constant,  $T_{\theta}$  is the Debye temperature,  $T_F$  is the Fermi temperature, and data for lead were reported by Poole [5]. The Born–Oppenheimer–Migdal approximation allows the separation of electronic and ionic motions in metals, because Equation (1) implies that the conductor exhibits fast charge carriers (for which characteristic energy scale is related to the Fermi temperature,  $T_F$ ) and relatively slow phonons (for which characteristic energy scale is related to the Debye temperature,  $T_{\theta}$ ).



**Citation:** Talantsev, E.F. Quantifying Nonadiabaticity in Major Families of Superconductors. *Nanomaterials* **2023**, *13*, 71. https://doi.org/10.3390/ nano13010071

Academic Editor: Yassine Slimani

Received: 2 December 2022 Revised: 17 December 2022 Accepted: 19 December 2022 Published: 23 December 2022



**Copyright:** © 2022 by the author. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). However, Equation (1) satisfies for many, but not for all superconductors, and the first discovered superconductor for which Equation (1) was found to be violated is *n*-type doped semiconducting  $SrTiO_3$  [6]:

$$\frac{\hbar\omega_D}{k_B T_F} = \frac{T_\theta}{T_F} = \left. \frac{627 \text{ K}}{13 \text{ K}} \right|_{\text{SrTiO}_3} = 48 \gg 1$$
(2)

where data for SrTiO<sub>3</sub> is taken from [7,8]. The theoretical description of the superconductivity in materials, in which the charge carriers and the lattice vibrations exhibit characteristic energy scales similar to Equation (2), is complicated, and the general designation of these superconductors are as nonadiabatic superconductors [9–16]. This theory [9–16] provides a general equation for the superconducting transition temperature,  $T_c$ , in nonadiabatic superconductors [9],  $T_c = 1.124 \times \frac{\epsilon_E}{2} \times a^{-\frac{1}{2}}$ , where  $c_c$  is the Fermi energy and  $\lambda_c$  is the

superconductors [9]:  $T_c = 1.134 \times \frac{\varepsilon_F}{k_B} \times e^{-\frac{1}{\lambda_{nad}}}$ , where  $\varepsilon_F$  is the Fermi energy, and  $\lambda_{nad}$  is the coupling strength constant in nonadiabatic superconductors, which serves a similar role to the electron–phonon coupling strength,  $\lambda_{e-ph}$ , in the BCS [1] and ME [2,3] theories. In addition, one of the primary fundamental theoretical problems is calculating this constant with acceptable accuracy to describe the experiment [4–16].

For experimentalists, it is important to have a simple practical routine to establish the strength of nonantiadiabatic effects in newly discovered superconductors. The most obvious parameter, which serves as an experimentally measured value to quantify the strength of nonantiadiabaticity, is the  $\frac{T_{\theta}}{T_{F}}$  ratio. For practical use of this criterion, there is a need for the taxonomy of possible  $\frac{T_{\theta}}{T_{F}}$  values.

To establish the taxonomy, we performed the analysis for a broad a range as possible of superconductors; these range from two- to three-dimensional materials, from elements to compounds of up to five elements, from low- $T_c$  (with  $T_c \sim 0.1$  K) to record high- $T_c$  (with  $T_c = 240$  K) hydrides, and from materials that exhibit a high order of crystalline lattice symmetry to the materials with low symmetry. Namely, we tried to cover all superconductors for which primary characteristic parameters (apart  $T_c$ ,  $T_{\theta}$ , and  $T_F$ ), such as the London penetration depth,  $\lambda(0)$ , the coherence length,  $\xi(0)$ , the amplitude of the superconducting energy gap,  $\Delta(0)$ , and the electron–phonon coupling strength constant,  $\lambda_{e-ph}$ , were established. In the results, we presented the analysis of more than 40 superconductors within the families of main superconductors.

Based on our analysis, we proposed the following classification scheme:

$$\begin{cases} \frac{T_{\theta}}{T_{F}} < 0.025 \rightarrow a diabatic superconductor;\\ 0.025 \lesssim \frac{T_{\theta}}{T_{F}} \lesssim 0.4 \rightarrow m oderately strong nonadiabaticity;\\ 0.4 < \frac{T_{\theta}}{T_{F}} \rightarrow nonadiabatic superconductor. \end{cases}$$
(3)

One of our findings is that for weakly nonadiabatic superconductors (i.e., for materials exhibited  $0.025 \le \frac{T_{\theta}}{T_F} \lesssim 0.4$ ), the predicting power of the BCS-ME theories (for instance, the prediction of the superconducting transition temperature) is reasonably accurate. However, all these superconductors are located outside of the BCS corner in the Uemura plot.

We also showed how the proposed classification scheme is linked to other known empirical scaling laws and taxonomies in superconductivity [13,17–21]; meanwhile, the search for the link of the proposed taxonomy with the recently reported big data [22,23] is under progress.

#### 2. Utilized Models

Proposed taxonomy is based on the knowledge of three fundamental temperatures of the superconductor, which are  $T_c$ ,  $T_{\theta}$ , and  $T_F$ . The superconducting transition temperature,  $T_c$ , is directly measured in either temperature resistance or in magnetization experiments. It is also important to mention the primary experimental techniques and theoretical models utilized to deduce the Debye temperature,  $T_{\theta}$ , and the Fermi temperature,  $T_F$ , in superconductors.

There are two primary techniques to determine the Debye temperature,  $T_{\theta}$ . One technique is to analyze the measured temperature-dependent normal-state specific heat,  $C_p(T)$ , from which the electronic specific heat coefficient,  $\gamma_n$ , and the Debye temperature,  $T_{\theta}$ , are deduced (see, for instance [24–26]):

$$\frac{C_p(T)}{T} = \gamma_n + \beta T^2 + \alpha T^4 \tag{4}$$

where  $\beta$  is the Debye law lattice heat-capacity contribution, and  $\alpha$  is from higher order lattice contributions. The Debye temperature can be calculated:

$$T_{\theta} = \left(\frac{12\pi^4 Rp}{5\beta}\right)^{\frac{1}{3}} \tag{5}$$

where *R* is the molar gas constant, and *p* is the number of atoms per formula unit.

Another technique is to fit normal-state temperature dependent resistance, R(T), to the Bloch–Grüneisen (BG) equation [24–28]:

$$R(T) = \frac{1}{\frac{1}{R_{sat}} + \frac{1}{R_0 + A \times \left(\frac{T}{T_{\theta}}\right)^5 \times \int_0^{T_{\theta}} \frac{x^5}{(e^x - 1)(1 - e^{-x})} \cdot dx}}$$
(6)

where,  $R_{sat}$  is the saturated resistance at high temperatures which is temperature independent,  $R_0$  is the residual resistance at  $T \rightarrow 0$  K, and A is free fitting parameter. Many research groups utilized both techniques (i.e., Equations (4)–(6)) to deduce  $T_{\theta}$  [24–27,29].

From the measured  $T_c$  and the deduced  $T_{\theta}$ , one can derive the electron–phonon coupling constant,  $\lambda_{e-ph}$ , as a root of either the original McMillan equation [30], or its recently revisited form [27]:

$$T_{c} = \left(\frac{1}{1.45}\right) \times T_{\theta} \times e^{-\left(\frac{1.04(1+\lambda_{e-ph})}{\lambda_{e-ph}-\mu^{*}(1+0.62\lambda_{e-ph})}\right)} \times f_{1} \times f_{2}^{*}$$
(7)

$$f_1 = \left(1 + \left(\frac{\lambda_{e-ph}}{2.46(1+3.8\mu^*)}\right)^{3/2}\right)^{1/3}$$
(8)

$$f_2^* = 1 + (0.0241 - 0.0735 \times \mu^*) \times \lambda_{e-ph}^2$$
(9)

where  $\mu^*$  is the Coulomb pseudopotential,  $0.10 \leq \mu^* \leq 0.15$  [27,30].

There are several experimental techniques to derive the Fermi temperature,  $T_F$ , from experimental data. One of these techniques is to measure the temperature dependent Seebeck coefficient, S(T), and fit a measured dataset to the equation [8]:

$$\left|\frac{S(T)}{T}\right| = \frac{\pi^2}{3} \frac{k_B}{e} \frac{1}{T_F}$$
(10)

Another approach is to measure the magnetic quantum oscillations [31], from which the magnitude of charge carrier mass,  $m^* = m_e (1 + \lambda_{e-ph})$  (where  $m_e$  is bare mass of electron), together with the size of the Fermi wave vector,  $k_F$ , can be obtained and plugged into [31]:

$$T_F = \frac{\hbar^2}{2k_B} \frac{k_F^2}{m^*} \tag{11}$$

An alternative approach is based on the extraction of the charge carriers mass,  $m^*$ , and density, n, as two of four parameters from the simultaneous analysis of  $C_p(T)$ , R(T), the muon spin relaxation ( $\mu$ SR), the lower critical field data,  $B_{c1}(T)$ , and the upper critical

field data,  $B_{c2}(T)$  [32], and plugging these parameters into the equation for an isotropic spherical Fermi surface [32]:

$$T_F = \frac{\hbar^2}{2k_B} \frac{1}{m^*} \left(3\pi^2 n_s\right)^{\frac{2}{3}}$$
(12)

where  $n_s$  is bulk charge curriers density at  $T \rightarrow 0$  K. For 3D superconductors,  $n_s$  is given by the equation [33]:

$$n_s(0) = \frac{m^*}{\mu_0 e^2} \frac{1}{\lambda^2(0)} \tag{13}$$

where  $\mu_0$  is the permeability of free space, *l* is the charge carrier mean free path,  $\lambda(0)$  is the ground state London penetration depth, and  $\xi(0)$  is the ground state coherence length.

It should be noted that  $\lambda(0)$  can be also deduced from the ground state lower critical field [28,34]:

$$B_{c1}(0) = \frac{\phi_0}{4\pi} \frac{\ln(1 + \sqrt{2\kappa(0)})}{\lambda^2(0)}$$
(14)

where  $\kappa(0) = \frac{\lambda(0)}{\xi(0)}$  is the ground state Ginzburg–Landau parameter.

For two dimensional (2D) superconductors,  $T_F$  can be determined from  $\mu$ SR measurements and crystallographic data [18]:

$$T_F = \frac{\pi \hbar^2}{k_B} \frac{1}{m^*} n_s \times c_{int}$$
(15)

where *c*<sub>int</sub> is the average distance between superconducting planes.

If measuring techniques are limited to the magnetoresistance measurements, R(T, B) (which was the case in the field of highly-compressed near-room temperature superconductors (NRTS) [35–50], until recent experimental progress by Minkov et al. [51,52]),  $T_F$  can be estimated by the equation [53]:

$$T_F = \frac{\pi^2 m^*}{2k_B \hbar^2} \times \xi^2(0) \times \Delta^2(0)$$
(16)

where  $\Delta(0)$  is the ground state amplitude of the superconducting energy gap, which is varying in a reasonably narrow range  $3.2 \leq \frac{2\Delta(0)}{k_BT_c} \leq 5.0$ , so that the ballpark value for  $T_F$  can be estimated. For instance,  $\xi(0)$  can be deduced from magnetoresistance measurements [54] and the electron–phonon coupling strength constant,  $\lambda_{e-ph}$ , can be assumed to be the average value of values calculated by first-principles calculations [55,56].

#### 3. Results

In Table 1, we present data for major groups of superconductors, where data sources for  $T_c$ ,  $T_{\theta}$ ,  $T_F$ , and other parameters (for instance,  $\lambda_{e-ph}$ ) are given.

**Table 1.** Superconductors and their parameters used in the work. In all calculations (except some original sources,  $\mu^* = 0.13$ ).

Type/Chemical Composition	λ(0) (nm)	ξ(0) (nm)	$\lambda_{e\text{-}ph}$	Т <sub>с</sub> (К)	Τ <sub>θ</sub> (K)	$\frac{2\Delta(0)}{k_B T_c}$	Т <sub>F</sub> (10 <sup>3</sup> К)	$T_{\theta}/T_{\rm F}$
Pure metals								
Aluminium				1.18 [57,58]	394 [5]		136 [5]	$2.9 imes10^{-3}$
Aluminium	50 [57]	1550 [58]	0.43 [59]	1.18 [57,58]	394 [5]	3.535 [ <b>5</b> 9]	18.9 (Equation (12))	$2.1  imes 10^{-2}$
Tin				3.72 [58]	170 [5]		118 [5]	$1.4  imes 10^{-3}$

Type/Chemical Composition	λ(0) (nm)	ξ(0) (nm)	$\lambda_{e-ph}$	Т <sub>с</sub> (К)	Т <sub>ө</sub> (К)	$\frac{2\Delta(0)}{k_B T_c}$	T <sub>F</sub> (10 <sup>3</sup> K)	$T_{\theta}/T_{\rm F}$
Tin	77 [60]	180 [58]	0.72 [59]	3.72 [58]	170 [5]	3.705 [59]	10.0 (Equation (12))	$1.2  imes 10^{-2}$
Lead				7.20 [58]	88 [5]		110 [5]	$8 imes 10^{-4}$
Lead	64 [60]	87 [58]	1.55 [59]	7.20 [60]	88 [5]	4.497 [59]	11.2 (Equation (12))	$7.8 imes10^{-3}$
Niobium				9.25 [58]	265 [5]		61.8 [5]	$4.3 imes10^{-3}$
Niobium	52 [58]	39 [58]	0.98 [59]	9.25 [58]	265 [5]	3.964 [59]	16.1 (Equation (12))	$1.6  imes 10^{-2}$
Gallium	52 [58]	39 [58]	2.25 [59]	1.09 [5,32,58,59]	325 [5,32,58,59]		120 [5,32,58]	$2.7  imes 10^{-3}$
A15 Alloys								
Nb <sub>3</sub> Sn	124 [61]	3.6 [ <mark>61</mark> ]	1.8 [62]	17.9 [61]	234 [61]	4.2 [62]	4.5 (Equation (12))	$5.2  imes 10^{-2}$
V <sub>3</sub> Si	62 [ <mark>62</mark> ]	3.3 [62]	0.96 [62]	16.4 [63]	297 [64]	3.7 [62]	12.8 (Equation (12))	$2.3  imes 10^{-2}$
Nb <sub>3</sub> Ge	90 [58]	3.0 [58]	1.60 [59]	23.2 [58]	302 [65]	4.364 [59]	7.1 (Equation (12))	$4.3 imes10^{-2}$
Heusler alloys								
ZrNi <sub>2</sub> Ga	350 [ <mark>66</mark> ]	15 [66]	0.551 [ <mark>66</mark> ]	2.85 [66]	300 [66]		1.4 (Equation (12))	$2.2  imes 10^{-1}$
YPd <sub>2</sub> Sn	196 [67]	19 [67]	0.70 [67]	4.7 [67]	210 [67]	4.1 [67]	2.9 (Equation (12))	$7.2  imes 10^{-2}$
HfPd <sub>2</sub> Al	225 [67]	13 [67]	0.68 [67]	3.66 [67]	182 [67]	3.74 [67]	2.4 (Equation (12))	$7.5  imes 10^{-2}$
Noncentrosymme	etric							
Nb <sub>0.5</sub> Os <sub>0.5</sub>	654 [ <mark>68</mark> ]	7.8 [68]	0.53 [68]	3.07 [68]	367 [68]	3.62 [68]	0.60 (Equation (12))	$6.1  imes 10^{-1}$
Re <sub>6</sub> Zr (mSR)	356 [ <b>2</b> 9]	3.7 [29]	0.67 [29]	6.75 [29]	338 [29]	3.72 [29]	1.3	$2.6  imes 10^{-1}$
Re <sub>6</sub> Zr (magnetization)	247 [29]	3.3 [29]	0.67 [29]	6.75 [29]	237 [29]	3.72 [29]	2.1	$1.1  imes 10^{-1}$
Mo <sub>3</sub> Al <sub>2</sub> C	376 [69]	4.2 [69]	0.74 (Equations (7)–(9))	9.2 [69]	339 [69]	4.03 [69]	1.2	$2.8  imes 10^{-1}$
NbIr <sub>2</sub> B <sub>2</sub> [70]	223	4.5	0.74	7.18	274		2.4	$1.1  imes 10^{-1}$
$TaIr_2B_2 [70]$	342	4.7	0.70	5.1	230		1.4	$1.7  imes 10^{-1}$
Re <sub>3</sub> Ta [71]			0.62	4.7	321		0.64	$5.0 imes10^{-1}$
Laves phases								
BaRh <sub>2</sub> [72]	340	8.4	0.80	5.6	178		1.4	$1.3  imes 10^{-1}$
SrRh <sub>2</sub> [72]	229	9.1	0.71	5.4	237		2.3	$1.0  imes 10^{-1}$
SrRh <sub>2</sub> [73]	121	8.6	0.93	5.4	250		5.3	$4.7  imes 10^{-2}$
SrIr <sub>2</sub> [74]	237	7.5	0.84	5.9	180		2.3	$8.2  imes 10^{-2}$
Intermetallics								
MgCNi <sub>3</sub> [75]	248	4.6	0.74 (Equations (7)–(9))	7.6	284		2.1	$1.4  imes 10^{-1}$
RuAl <sub>6</sub> [76]	265	27.7	0.81	1.21	458		1.9	$2.4  imes 10^{-1}$
Perovskite								
SrTiO <sub>3</sub>			0.2 [7]	0.086 [8]	690 [77]		$1.3  imes 10^{-2}$ [8]	$5.3 imes10^1$
Pnictides								
ThFeAsN	375 [78]		1.48 [78]	28.1 [78]	332 [79]		0.47 (Equation (17)) c <sub>int</sub> = 8.5 Å [78]	$7.0 imes10^{-1}$
KCa <sub>2</sub> Fe <sub>4</sub> As <sub>4</sub> F <sub>2</sub>	230 [80]		1.59 [80]	33.4 [80]	366 [80]		1.3 (Equation (17)) $c_{int} = 8.5 \text{ Å} [80]$	$2.9  imes 10^{-1}$
RbCa <sub>2</sub> Fe <sub>4</sub> As <sub>4</sub> F <sub>2</sub>	232 [80]		1.45 [80]	29.2 [80]	332 [80]		1.2 (Equation (17)) $c_{int} = 8.5 \text{ Å} [80]$	$2.8  imes 10^{-1}$
$CsCa_2Fe_4As_4F_2$	244 [80]		1.44 [80]	28.3 [80]	344 [80]		1.1 (Equation (17)) c <sub>int</sub> = 8.5 Å [80]	$3.1  imes 10^{-1}$

Table 1. Cont.

Type/Chemical Composition	λ(0) (nm)	ξ(0) (nm)	$\lambda_{e-ph}$	Т <sub>с</sub> (К)	Т <sub>ө</sub> (К)	$\frac{2\Delta(0)}{k_BT_c}$	<i>T</i> <sub>F</sub> (10 <sup>3</sup> K)	$T_{\theta}/T_{\rm F}$
Cuprates								
YBa <sub>2</sub> Cu <sub>3</sub> O <sub>7</sub> [81]	115 [81,82]	2.5 [81]	1.5 [83]	93.2 [81]	437 [7]		3.4 (Equation (17)) c <sub>int</sub> = 5.8 Å [83]	$1.2  imes 10^{-1}$
(Y,Dy)Ba <sub>2</sub> Cu <sub>3</sub> O <sub>7</sub> [84]	128 [84,85]	2.5 [81]	1.5 [83]	90.4 [84,85]	437 [7]	4.24 [84,85]	2.9 (Equation (17)) c <sub>int</sub> = 5.8 Å [83]	$1.5  imes 10^{-1}$
Bi <sub>2</sub> Sr <sub>2</sub> CaCu <sub>2</sub> O <sub>8</sub> [86]	196 [85]	1.2 [85]	4.7 [7]	82.7 [85]	240 [7]	3.9 [85]	1.2 (Equation (17)) c <sub>int</sub> = 6 Å [83]	$2.0  imes 10^{-1}$
Tl <sub>2</sub> Ba <sub>2</sub> CaCu <sub>2</sub> O <sub>8</sub> [87]	179 [85]	1.2 [85]		103 [85]	425 [88]	4.3 [85]	1.5 (Equation (17)) c <sub>int</sub> = 6 Å [83]	$2.9  imes 10^{-1}$
HgBa <sub>2</sub> CaCu <sub>2</sub> O <sub>8</sub> [89]	188 [85]	1.6 [85]		120 [85]	525 [88]	3.3 [85]	1.3 (Equation (17)) c <sub>int</sub> = 6 Å [83]	$3.9  imes 10^{-1}$
$\begin{array}{c} \text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}\\ [90] \end{array}$	175 [85]	1.0 [85]	4.5	85 [85]	319 [7]	4.5 [85]	1.5 (Equation (17)) c <sub>int</sub> = 6 Å [83]	$2.1  imes 10^{-1}$
Bismuthates								
$Ba_{1-x}K_{x}BiO_{3}$ (x = 0.4) [91,92]			1.10 [93]	23 [92]	210 [94]	3.8-4.1 [93]	1.5 [92]	$1.4  imes 10^{-1}$
$Ba_{1-x}K_{x}BiO_{3}$ (x = 0.5) [91,92]			1.10 [93]	14 [92]	210 [94]	3.8-4.0 [93]	1.1 [92]	$1.9  imes 10^{-1}$
2D superconductors								
MATBG [95]	2180 [96]		$rac{m^*}{m_e} = 0.2$ [96]	1.2 [96]	1864 [97]	4.4 [96]	$16.5 \times 10^{-3}$ (Equation (15)) $c_{int} = 1 \text{ nm}$	$1.1  imes 10^2$
MATBG [95]		61.4 [96]	[96]	1.2 [96]	1864 [97]	4.4 [96]	$28.6 \times 10^{-3}$ (Equation (16))	$6.5  imes 10^1$
Li-doped graphene, LiC <sub>6</sub> [98]			0.61 [99]	5.9 [98]	2240 [99]		15.5 [99]	$1.45  imes 10^{-1}$
IrTe <sub>2</sub> [100] (sample thickness is 21 nm)	600 [100]	75 [100]		1.6 [100]		5.46 [100]	$0.118 (Equation (15))$ $c_{int} = 0.54 \text{ nm}$	
Ionic Salt								
CsI ( <i>P</i> = 206 GPa) [101]			0.445 [102]	1.1 [101]	339 [102]		$\begin{array}{c} (20\pm 4)\times 10^{-2} \\ [102] \end{array}$	$17 \pm 4$ [102]
NRTS hydrides								
$H_3S$ ( <i>P</i> = 155 GPa) [35]	37 [52]	1.9 [54]	2.2 [103]	197 [103]	1427 [103]		21.6 (Equation (12)) and [104]	$6.6  imes 10^{-2}$
$H_3S$ ( <i>P</i> = 155 GPa) [35]		1.9 [54]	1.76 [55,56]	197 [103]	1427 [103]	3.55 [53]	$10 \pm 3$ (Equation (16)) and [104]	$\begin{array}{c} (1.4\pm 0.3)\times \\ 10^{-1} \end{array}$
LaH <sub>10</sub> ( $P = 150$ GPa) [36]	30 [51]	1.5 [51]	2.77 [27]	240 [27]	1310 [27]		27.0 (Equation (12))	$2.7 \times 10^{-2}$
$\begin{array}{c} \text{La}_{1-x}\text{Nd}_x\text{H}_{10} \\ (x = 0.15) \\ (P = 180 \text{ GPa}) \\ \hline [48] \end{array}$		2.3 [105]	1.65 [105]	122 [105]	1156 [105]	4.0 [105]	4.4 [105] (Equation (16))	$2.6 \times 10^{-1}$
Compressed oxygen								
ζ-O <sub>2</sub> ( <i>P</i> = 115 GPa) [106]		42 [107]	0.42 [107]	0.64 [107]	306 [107]		$3.5 \times 10^{-2}$ [107] (Equation (16))	8.7

Table 1. Cont.

In Figure 1, we show the  $T_c$  vs. $T_F$  dataset in a log–log plot, which is the traditional data representation in the well-known Uemura plot [18].





**Figure 1.** Uemura plot ( $T_c$  vs.  $T_F$ ) for primary superconducting families. References on original data ( $T_c$  and  $T_F$ ) can be found in Table 1.

In Figure 2, we represent the same superconducting materials, but here we display the  $\lambda_{e-ph}$  vs.  $\frac{T_{\theta}}{T_F}$  dataset in a semi-log plot. To our best knowledge, the  $\lambda_{e-ph}$  vs.  $\frac{T_{\theta}}{T_F}$  plot was first plotted by Pietronero et al. [13] in linear–linear scales. However, because the  $\frac{T_{\theta}}{T_F}$  ratio for main families of superconductors is varied within four orders of magnitude (Table 1), and  $0.4 \leq \lambda_{e-ph} \leq 3.0$ , it is more suitable to use the semi-log plot (Figure 2).



electron-phonon coupling strength,  $\lambda_{e-ph}$ 

**Figure 2.** Plot of  $\frac{T_{\theta}}{T_{F}}$  vs.  $\lambda_{e-ph}$  for primary superconducting families. This type of plot proposed by Pietronero et al. [13]. References for original data  $(T_{\theta}, \lambda_{e-ph}, T_{F})$  can be found in Table 1.

Finally, in Figure 3, we represented the same superconducting materials, but here we displayed the  $T_c$  vs.  $\frac{T_{\theta}}{T_F}$  dataset in a log–log plot. This type of plot was chosen because as  $T_c$ , as  $\frac{T_{\theta}}{T_r}$  are varied within several orders of magnitude.



**Figure 3.** Plot  $\frac{T_{\theta}}{T_F}$  vs.  $T_c$  for primary superconducting families. References on original data ( $T_{\theta}$ ,  $T_c$ ,  $T_F$ ) can be found in Table 1.

#### 4. Discussion

The family of near-room temperature superconductors (NRTS) is represented in Table 1 and Figure 1 by H<sub>3</sub>S (P = 155 GPa), SnH<sub>12</sub> (P = 190 Gpa), and La<sub>1-x</sub>Nd<sub>x</sub>H<sub>10</sub> (x = 0.09, P = 180 Gpa). Two independent approaches were used to perform calculations in H<sub>3</sub>S:

- 1.  $T_F$  was calculated based on Equations (12) and (13). In these calculations  $\lambda(0) = 37$  nm (extracted from the analysis of DC magnetization experiments reported by Minkov et al. [51,52]) was used.
- 2.  $T_F$  was calculated based Equations (6)–(9) and (18), in which utilized  $\xi(0)$  values were extracted [53] from magnetoresistance measurements reported by Mozaffari et al. [54]).

It should be noted that, in both approaches, the electron–phonon coupling strength constant,  $\lambda_{e-ph}$ , was assumed to be  $\lambda_{e-ph} = 1.76$ , which is the average value of values calculated by first-principles calculations [55,56], and values extracted from experimental R(T) data [27].

It can be seen in Table 1 and Figure 1 that the calculated  $T_F$  values for H<sub>3</sub>S, by two alternative approaches, are in a very good agreement with each other. To demonstrate the acceptable level of variation in  $T_F$  values for the same material, in Table 1 and Figure 1 we present the results of the calculations for pure metals, where  $T_F$  was calculated by the two approaches mentioned above and the use of experimental data reported by different research groups.

 $T_F$  in HTS cuprates were calculated by the Equations (13) and (15), which do not require the knowledge of the electron–phonon coupling constants,  $\lambda_{e-ph}$ . This is despite Ledbetter et al. [7] reporting the so-called effective electron–phonon coupling strength,  $\lambda_{e-ph,eff}$ , from which the effective mass can be deduced,  $m^* = (1 + \lambda_{e-ph,eff}) \times m_e$ .

In addition, it should be noted that for YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>, Uemura [83] reported the relation [83]:

$$\frac{n^*}{n_e} = 2.5 \tag{17}$$

from which  $\lambda_{e-ph} = 1.5$  can be derived. Calculated values are in a reasonable agreement with experimental  $\frac{m^*}{m_e}$  values reported by several research groups [108–110] in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-x</sub>.

However, because the phenomenology of the electron–phonon mediated superconductivity cannot describe the superconducting state in cuprates, and the  $T_{\theta}$  for cuprates were taken as experimental values (see, for instance, report by Ledbetter et al. [7,88]), all cuprate superconductors are shown in Figures 1 and 3 and are not shown in Figure 2.

It should be mentioned that the result of the  $T_F$  calculation in MATBG (Table 1),  $T_F = 16.5$  K, which was primarily based on the London penetration depth,  $\lambda(0) = 1860$  nm, was deduced in Ref. [96] from the self-field critical current density,  $J_c(sf, T)$ , by the approach proposed by us [84]:

$$J_c(sf,T) = \frac{\phi_0}{4\pi\mu_0} \frac{ln\left(1 + \sqrt{2}\kappa(T)\right)}{\lambda^3(T)}$$
(18)

The remarkable agreement of the deduced value,  $T_F = 16.5$  K, and the value reported in the original work on MATBG by Cao et al. [95],  $T_F = 17$  K, which was calculated based on normal state charge carriers density in MATBG, independently validates our primary idea [84] about the fundamental nature of the self-field critical current in weaklinks samples [84,85,111]. This concept was recently proven by Paturi and Huhtinen [112], who utilized the fact that the London penetration depth,  $\lambda(0)$ , in real samples, depends on the mean free-path of charge carriers, *l*:

$$\lambda(0) = \lambda_{clean\ limit}(0)\sqrt{1 + \frac{\xi(0)}{l}}$$
(19)

where  $\lambda(0)$  is the effective penetration depth, and  $\lambda_{clean \ limit}(0)$  is the penetration depth in samples, exhibiting a very long mean free-path,  $l \gg \xi(0)$ . Paturi and Huhtinen [112] varied l in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-x</sub> films and showed that the change in  $J_c(st, T)$  satisfies Equations (18) and (19).

Materials, in which  $\lambda(0)$  was deduced by the mean temperature dependent self-field critical current density,  $J_c(sf, T)$  (Equation (18)), have designation " $J_c(sf, T)$ " in Figures 1–3.

The MATBG does not show in Figure 2, because the derivation of  $\lambda_{e-ph}$  cannot be performed by the used phenomenology:  $m^* = (1 + \lambda_{e-ph}) \times m_e$ , because  $\frac{m^*}{m_e} = 0.2$  [96]; however, this material is shown in Figures 1 and 3, because  $\lambda_{e-ph}$  is not required for these plots.

Returning back to hydrides, we need to note that Durajski [56] performed firstprinciples and studied the strength of the nonadiabatic effects in highly-compressed sulfur hydride and phosphorus hydride. Calculations show that the strength of the nonadiabatic effects can be quantified as moderately weak in comparison with the classical nonadiabatic superconductor SrTiO<sub>3</sub>. This is in a good agreement with our result (see Figure 3 and Table 1), that all deduced  $\frac{T_{\theta}}{T_{r}}$  values for NRTS are within the range of:

$$0.03 \le \frac{T_{\theta}}{T_F} \le 0.3 \tag{20}$$

Moreover, the classical nonadiabatic superconductor  $\text{SrTiO}_3$  falls into the intermediate zone between unconventional and BCS superconductors; this is because this material exhibits  $\frac{T_c}{T_F} = 0.0066$ , and by this criterion,  $\text{SrTiO}_3$  is similar to the Laves phase materials, intermetallics, A-15 alloys, and Heusler alloys, which cannot be considered to be a correct manifestation of primary uniqueness for this nonadiabatic material.

More unexpectedly, a two dimensional LiC<sub>6</sub> (which is a lithium-doped graphene) superconductor falls into the BCS metals zone in the Uemura plot (Figure 1), despite the fact that this material exhibits reasonable strength in the nonadiabatic effects,  $\frac{T_c}{T_F} = 0.15$  [99].

However, in Figures 2 and 3, the outstanding separations of all nonadiabatic superconductors from their adiabatic and moderate nonadiabatic counterparts are clearly manifested.

By looking at the data in Figures 2 and 3, it is easy to recognize that 3/4 (32 of 42) of the analyzed superconductors fall into a reasonably narrow band:

$$0.025 \le \frac{T_{\theta}}{T_F} \le 0.4 \tag{21}$$

Based on this, we proposed that the values in Equation (21) were used as empirical limits for the adiabatic superconductors ( $\frac{T_{\theta}}{T_F} \leq 0.025$ ), moderate nonadiabatic superconductors ( $0.025 \leq \frac{T_{\theta}}{T_F} \leq 0.4$ ), and strong nonadiabatic superconductors ( $\frac{T_{\theta}}{T_F} \geq 0.4$ ).

It also follows from our analysis that all strong nonadiabatic superconductors exhibit low superconducting transition temperatures,  $T_c \leq 1.2$  K (Figure 3).

#### 5. Conclusions

In this work, we proposed a new classification scheme to quantify the effects of nonadiabaticity in superconductors. By performing the analysis of experimental data for more than 40 superconductors, which represent the primary families of superconductors, we found that  $\frac{3}{4}$  of all analyzed superconductors fall into a narrow  $0.025 \leq \frac{T_{\theta}}{T_F} \leq 0.4$  band. Based on this, we proposed the taxonomy for the strength of the nonadiabatic effects in superconductors.

**Funding:** This research was funded by the Ministry of Science and Higher Education of the Russian Federation, grant number No. AAAA-A18-118020190104-3 (theme "Pressure"). The research funding from the Ministry of Science and Higher Education of the Russian Federation (Ural Federal University Program of Development within the Priority-2030 Program) is gratefully acknowledged. The APC was funded by MDPI.

Data Availability Statement: Not applicable.

Acknowledgments: The author thanks Luciano Pietronero (Universita' di Roma) for comments about the limitations of the applicability of Migdal–Eliashberg (ME) theory of the electron–phonon mediated superconductivity. The author also thanks Dmitry V. Semenok (Skolkovo Institute of Science and Technology) and Dominik Szczesniak (Jan Dlugosz University in Czestochowa) for reading and commenting on the paper.

Conflicts of Interest: The author declares no conflict of interest.

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