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BRIEF REPORT

Upper Limit for Superconducting Transition Temperature in Electron – Phonon Superconductors: Very Strong Coupling

M. V. Sadovskii[1]

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Abstract
We present a brief review of some recent work on the problem of highest achievable temperature of superconducting transition T_c in electron – phonon systems. The discovery of record – breaking values of T_c in quite a number of hydrides under high pressure was an impressive demonstration of capabilities of electron – phonon mechanism of Cooper pairing. This lead to an increased interest on possible limitations of Eliashberg – McMillan theory as the main theory of superconductivity in a system of electrons and phonons. We shall consider some basic conclusions following from this theory and present some remarks on the limit of very strong electron – phonon coupling. We shall discuss possible limitations on the value of the coupling constant related to possible lattice and specific heat instability and conclude that within the stable metallic phase the effective pairing constant may acquire very large values. We discuss some bounds for T_c derived in the strong coupling limit and propose an elementary estimate of an upper limit for T_c, expressed via combination of fundamental physical constants. Finally we also briefly discuss some pessimistic estimates for T_c of metallic hydrogen obtained in "jellium" model

Keywords Eliashberg – McMillan theory · Electron – phonon interaction · Very strong coupling · Transition temperature

1 Introduction

First ideas to enhance superconducting critical temperature T_c were introduced soon after the formulation of BCS theory. In 1964 Little [1] and Ginzburg [2] proposed an idea of an "excitonic" mechanism – the replacement of phonons as a "glue" leading to Cooper pairing by some other Boson – type excitations with higher energies, thus changing the Debye frequency ω_D in the preexponential factor of BCS expression for T_c by some ω_{ex} (with $\omega_{ex} > \omega_D$), which leads to the increase of T_c (probably in some lucky case up to room temperatures). These ideas were further developed in many papers reviewed in the famous book [3]. Unfortunately, up to now this "excitonic" mechanism was never and nowhere realized experimentally. The discoveries of high – temperature in copper oxides (1986) and iron pnictides and chalcogenides (2008) were more or less unrelated to these theoretical proposals and will not be discussed here.

The similar idea of the use of much larger values of ω_D in the case of the usual electron – phonon pairing mechanism was introduced later by Ashcroft [4], who proposed to study the metallic hydrogen (with apparently larger ω_D due to a small mass of hydrogen ion) and different hydrides [5], which can be stable under extremely high pressures. It is important to stress that all of these works used basically the standard weak (or intermediate) coupling approximation of BCS (McMillan) theory.

These proposals were criticized in the notorious paper by Cohen and Anderson [6], where rather elegant arguments were given, seemingly quite convincing, that characteristic scale of T_c values due to electron – phonon or "excitonic" mechanism (based on exchange of Bose – like excitations in metals) can be of the order of about 10 K only. This paper was immediately seriously criticized in Refs. [3, 7], with the conclusion that in reality there are no such limitations. However, the point of view expressed in

Dedicated to the memory of Mikhail Eremets.

✉ M. V. Sadovskii
sadovski@iep.uran.ru

[1] Institute for Electrophysics, Russian Academy of Sciences, Ural Branch, Amundsen str. 106, 620016 Ekaterinburg, Russia

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Cohen – Anderson paper became popular in physics community (Anderson himself till the end of his life adhered to the view expressed in Ref. [6], though Cohen [8] has acknowledged the validity of arguments expressed in Dolgov et al. [7]), so that at the time of discovery of high – temperature superconductivity in cuprates (1986 – 1987), the common belief was that the "usual" electron – phonon mechanism does not allow values of T_c higher, that 30 – 40 K. Because of this after the discovery of superconductivity in cuprates the "great race" has started for new theoretical models and mechanisms of superconductivity, which may explain the high values of T_c. It is most probable, that in these compounds T_c is determined by some kind of non – phonon pairing mechanism (e.g. due to antiferromagnetic fluctuations). Thus the problems of superconductivity in cuprates (as well as in iron pnictides and chalcogenides) are outside the scope of this work, which will discuss only the electron – phonon pairing.

The remarkable discovery by Mikhail Eremets group of superconductivity in H_3S with $T_c \sim$200 K and further rapid development of experimental studies of high – temperature superconductivity in different hydrides [10–13] (to quote only some of the review papers) has opened the new path to almost room – temperatures superconductivity (though at extremely high (megabar) pressures) and stimulated an active theoretical work [14, 15]. There was no doubt from the very beginning, that high – T_c superconductivity in hydrides is due to the usual electron – phonon coupling. So from theoretical point of view probably the most important result of discovery of record T_c values in hydrides under high pressures, in our opinion, is the final (and experimental!) rebuttal of the point of view expressed in Ref. [6], explicitly demonstrating the possibility of achieving high - T_c values (of the order of 10^2 K) with the common electron – phonon mechanism. Most pressing now becomes the question of the upper limit of T_c, which can be achieved due to this pairing mechanism. Below we shall try to discuss this problem once again within the standard approach, based on Eliashberg – McMillan equations, as most successful theory, describing superconductivity in the system of electrons and phonons in metals.

2 Electron – Phonon Interaction and Eliashberg – McMillan theory: Strong Coupling Limit

2.1 Some General Expressions and Definitions

Fröhlich Hamiltonian which is commonly used to describe electron – phonon interaction is written as:

$$H = \sum_{\mathrm{p}} \varepsilon_{\mathrm{p}} a_{\mathrm{p}}^{+} a_{\mathrm{p}} + \sum_{\mathrm{k}} \Omega_{0\mathrm{k}} b_{\mathrm{k}}^{+} b_{\mathrm{k}} + \\ + \frac{1}{\sqrt{N}} \sum_{\mathrm{pk}} g_{\mathrm{k}} a_{\mathbf{p}+\mathbf{k}}^{+} a_{\mathrm{p}} (b_{\mathrm{k}} + b_{-\mathrm{k}}^{+}) \tag{1}$$

where ε_{p} is the conduction electron energy (counted from the Fermi level), $\Omega_{0\mathrm{k}}$ is the "bare" phonon spectrum *in the absence* of electron – phonon interaction (which is actually rather poorly defined in the case of a real metal), and we have introduced the standard notations for creation a_{p}^{+} and annihilation a_{p} operators of electrons and phonons – b_{k}^{+} and b_{k}, N is the number of atoms in crystal.

The matrix element of electron – phonon interaction is usually written as:

$$g_{\mathrm{k}} = -\frac{1}{\sqrt{2M\Omega_{0\mathrm{k}}}} \langle \mathrm{p}|\mathbf{e}(\mathrm{q}) \nabla V_{ei}(\mathrm{r})|\mathbf{p}+\mathbf{q}\rangle \\ \equiv -\frac{1}{\sqrt{2M\Omega_{0\mathrm{k}}}} I(\mathrm{k}) \tag{2}$$

where V_{ei} is electron – ion interaction potential, M is the ion mass, and $\mathbf{e}(\mathbf{q})$ is polarization vector of a phonon with frequency $\Omega_{0\mathrm{q}}$.

McMillan [19] has derived a simple, but very general, expression for the dimensionless electron – phonon coupling in Eliashberg theory. The so called Eliashberg – McMillan function is *defined* [19, 23] as:

$$\alpha^2(\omega)F(\omega) = \\ = \frac{1}{N(0)} \sum_{\mathrm{p}} \sum_{\mathbf{p}'} |g_{\mathbf{pp}'}|^2 \delta(\omega - \Omega_{\mathbf{p}-\mathbf{p}'}) \delta(\varepsilon_{\mathrm{p}}) \delta(\varepsilon_{\mathbf{p}'}) = \\ = \frac{1}{N(0)} \sum_{\mathrm{p}} \sum_{\mathbf{p}'} \frac{1}{2M\Omega_{\mathbf{p}-\mathbf{p}'}} |I(\mathbf{p}-\mathbf{p}')|^2 \delta(\omega - \Omega_{\mathbf{p}-\mathbf{p}'}) \\ \times \delta(\varepsilon_{\mathrm{p}}) \delta(\varepsilon_{\mathbf{p}'}) \tag{3}$$

where $\Omega_{\mathbf{p}-\mathbf{p}'}$ is the phonon frequency and $F(\omega) = \sum_{\mathrm{q}} \delta(\omega - \Omega_{\mathrm{q}})$ is the phonon density of states. As phonons typically scatter electrons in metals only in some narrow region close to the Fermi surface we introduce the matrix element of the gradient of electron –ion potential averaged over Fermi surface:

$$\langle I^2 \rangle = \frac{1}{[N(0)]^2} \sum_{\mathrm{p}} \sum_{\mathbf{p}'} |I(\mathbf{p}-\mathbf{p}')|^2 \delta(\varepsilon_{\mathrm{p}}) \delta(\varepsilon_{\mathbf{p}'}) = \\ = \frac{1}{[N(0)]^2} \sum_{\mathrm{p}} \sum_{\mathbf{p}'} |\langle \mathrm{p}|\nabla V_{ei}(\mathrm{r})|\mathbf{p}'\rangle|^2) \delta(\varepsilon_{\mathrm{p}}) \delta(\varepsilon_{\mathbf{p}'}) = \\ = \langle |\langle \mathrm{p}|\nabla V_{ei}(\mathrm{r})|\mathbf{p}'\rangle|^2 \rangle_{FS} \tag{4}$$

Then we immediately get:

$$\int_0^\infty d\omega\alpha^2(\omega)F(\omega)\omega = \frac{N(0)\langle I^2\rangle}{2M} \tag{5}$$

Dimensionless electron – phonon coupling constant is expressed now via this Fermi – surface average as [19, 23]:

$$\lambda = 2\int_0^\infty \frac{d\omega}{\omega}\alpha^2(\omega)F(\omega) = \\ = \frac{2}{\langle\Omega^2\rangle}\int_0^\infty d\omega\alpha^2(\omega)F(\omega)\omega \tag{6}$$

where the mean square phonon frequency is defined as:

$$\langle\Omega^2\rangle = \frac{\int_0^\infty d\omega\alpha^2(\omega)F(\omega)\omega}{\int_0^\infty \frac{d\omega}{\omega}\alpha^2(\omega)F(\omega)} = \frac{2}{\lambda}\int_0^\infty d\omega\alpha^2(\omega)F(\omega)\omega \tag{7}$$

From this expression we can immediately see that:

$$\lambda = \frac{N(0)\langle I^2\rangle}{M\langle\Omega^2\rangle} \tag{8}$$

This expression gives very useful representation for λ, which is often used in the literature and in practical calculations.

Migdal's theorem [16] allows us to neglect vertex corrections in all calculations of Feynman diagrams related to electron – phonon interaction in typical metals. The actual small parameter of perturbation theory is $\lambda\frac{\Omega_0}{E_F} \ll 1$, where λ is the dimensionless constant of electron – phonon interaction and Ω_0 is characteristic phonon frequency (e.g. of the order of Debye frequency ω_D), while E_F is the Fermi energy of electrons, which in typical metals is of the order of conduction band width. In particular this leads to a common belief, that vertex corrections in this theory can be neglected even in case of $\lambda > 1$ (up to the values of $\lambda \sim \frac{E_F}{\Omega_0} \gg 1$), as inequality $\frac{\Omega_0}{E_F} \ll 1$ holds in typical metals. This fact is the cornerstone of Eliashberg – McMillan theory for superconductors which allows the description of the so called strong coupling superconductivity outside the usual weak coupling limit of BCS theory [17–23].

2.2 *Lower* Bound for T_c in Eliashberg – McMillan Theory and Strong Coupling Limit

Limitations on the value of T_c in Eliashberg – McMillan theory in the limit of very strong coupling can be derived analytically [20, 23]. In the following we shall not consider the role of direct Coulomb repulsion of electrons within the Cooper pair, which is accounted for in the complete Eliashberg – McMillan theory, limiting ourselves only to electron – phonon interaction. The accounting for Coulomb contributions is not especially difficult [19] and reduces at the end to introduction of the usual Coulomb pseudopotential $\mu^\star$, which in typical metals is rather small and not so important in the limit of very strong coupling with phonons, which will be of the main interest for us in the following.

From the general system of Eliashberg – McMillan equations directly follows the linearized equation for the gap $\Delta(\omega_n)$ [23], determining T_c:

$$\Delta(\omega_n)Z(\omega_n) = \pi T\sum_{n'}\int_0^\infty \alpha^2(\omega)F(\omega)\times \\ \times D(\omega_n-\omega_{n'};\omega)\frac{\Delta(\omega_{n'})}{|\omega_{n'}|} \tag{9}$$

where $\omega_n = (2n+1)\pi T$ are usual Matsubara frequencies of electrons. The renormalization factor $Z(\omega_n)$ is determined from:

$$1 - Z(\omega_n) = \frac{\pi T}{\omega_n}\sum_{n'}\int_{-\infty}^\infty d\xi\int_0^\infty d\omega\alpha^2(\omega)F(\omega)\times \\ \times D(\omega_n-\omega_{n'};\omega)\frac{\omega_{n'}}{|\omega_n|} \tag{10}$$

and phonon Green's function is expressed as:

$$D(\omega_n-\omega_{n'};\omega) = \frac{2\omega}{(\omega_n-\omega_{n'})^2+\omega^2} \tag{11}$$

Actually, Eq. (9) represents the system of linear equations for $\Delta(\omega_n)$. Let us consider here first the term with $n = 0$. Then, leaving in the sum in Eq. (10) only the contribution from $n' = 0$, we obtain:

$$Z(0) = 1 + \lambda \tag{12}$$

which represents the usual electron mass – renormaliztion factor due to electron – phonon interaction: $m^\star = m(1+\lambda)$. Its substitution into Eq. (9) for $n = 0$ just cancels the similar (corresponding to $n' = 0$) term in the r.h.s., so that the equation for $\Delta(0) = \Delta(\pi T)$ takes the form:

$$\Delta(0) = \\ = \pi T\sum_{n'\neq 0}\int_0^\infty \alpha^2(\omega)F(\omega)\frac{2\omega}{(\pi T-\omega_{n'})^2+\omega^2}\frac{\Delta(\omega_{n'})}{|\omega_{n'}|} \tag{13}$$

All terms in the r.h.s. here are positive. Let us leave only the contribution from $n' = -1$, then after simple algebra with the account of $\Delta(0) = \Delta(\pi T) = \Delta(-\pi T) = \Delta(-1)$ we immediately obtain the *inequality* [20, 23]:

$$1 > \int_0^\infty d\omega\frac{2\alpha^2(\omega)F(\omega)\omega}{(2\pi T)^2+\omega^2} \tag{14}$$

Putting $T = T_c$ in (14) we obtain the *lower* bound for T_c. In particular, in the model with Einstein spectrum of phonons $F(\omega) = \delta(\omega - \Omega_0)$ and this inequality is immediately rewritten as:

$$1 > 2\alpha^2(\Omega_0)\frac{\Omega_0}{(2\pi T)^2 + \Omega_0^2} = \lambda\frac{\Omega_0^2}{(2\pi T)^2 + \Omega_0^2} \tag{15}$$

so that for T_c we get:

$$T_c > \frac{1}{2\pi}\sqrt{\lambda - 1}\Omega_0 \tag{16}$$

which for $\lambda \gg 1$ reduces to:

$$T_c > \frac{1}{2\pi}\sqrt{\lambda}\Omega_0 \approx 0.16\sqrt{\lambda}\Omega_0 \tag{17}$$

For the model of phonon spectrum consisting of discrete set of Einstein phonons:

$$\begin{aligned}\alpha^2(\omega)F(\omega) &= \sum_i \alpha^2(\Omega_i)\delta(\omega - \Omega_i) \\ &= \sum_i \frac{\lambda_i}{2}\Omega_i\delta(\omega - \Omega_i)\end{aligned} \tag{18}$$

In this case from (7) we simply obtain:

$$\langle\Omega^2\rangle = \frac{1}{\lambda}\sum_i \lambda_i\Omega_i^2 \tag{19}$$

where $\lambda = \sum_i \lambda_i$. In this case the inequality (14) reduces to:

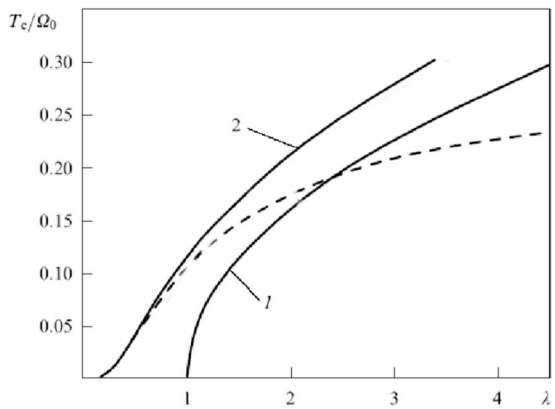

Fig. 1 Temperature of superconducting transition in Einstein model of phonon spectrum in units of T_c/Ω_0 as a function of pairing constant λ [20]: 1 – lower bound (16), 2 – numerically exact solution of the full system of equations [20]. McMillan expression for T_c [19] is shown by dashed line (for the case of μ^*=0)

$$1 > \sum_i \lambda_i\frac{\Omega_i^2}{(2\pi T)^2 + \Omega_i^2} \tag{20}$$

which in the limit of very strong coupling immediately gives the natural generalization of (17):

$$T_c > \frac{1}{2\pi}\sqrt{\lambda\langle\Omega^2\rangle} \tag{21}$$

Numerically exact solution of the full system of Eq. (9), performed in Ref. [20] leads to the final expression for T_c in the strong coupling limit of $\lambda \gg 1$ with replacement of $1/2\pi = 0.16$ in (17) or (21) by 0.182:

$$T_c = 0.182\sqrt{\lambda\langle\Omega^2\rangle} \tag{22}$$

It is obvious, that even the simplest solution (16) is quite sufficient for qualitative estimates of T_c in the limit of very strong coupling. The general situation is illustrated in Fig. 1. From this figure it can be seen, in particular, that asymptotic behavior of T_c for $\lambda \gg 1$ (22) with coefficient 0.182, approximates the values of critical temperature rather well already starting from the values of $\lambda >$1.5-2.0. The remarkable result here is the replacement of exponential dependence of T_c on the coupling constant, typical for the weak coupling BCS or intermediate coupling McMillan approximations, by the square root dependence, leading to monotonous and seemingly unlimited growth of T_c with increasing λ. Then an important question arises – are there any limitations to the growth of λ ?

3 Possible Limitations for Electron – Phonon Coupling

3.1 Fröhlih Instability

The general expression for phonon Green's function, taking into account the interaction with electrons, is given by Dyson equation shown in Fig. 2. Then such "dressed" phonon Green's function can be written as:

$$D(\mathrm{k}\omega) = \frac{2\Omega_{0\mathrm{k}}}{\omega^2 - \Omega_{\mathrm{k}}^2 + i\delta} \tag{23}$$

where the renormalized phonon spectrum is determined from the equation:

$$\Omega_{\mathrm{k}}^2 = \Omega_{0\mathrm{k}}^2\left[1 + \frac{2|g_{\mathrm{k}}|^2}{\Omega_{0\mathrm{k}}}\Pi(\mathrm{k}, \Omega_{\mathrm{k}})\right] \tag{24}$$

Fig. 2 Dyson equation for the full ("dressed") phonon Green's function

In adiabatic approximation, taking into account Migdal theorem, polarization operator here can be taken as a simple loop. With rather high accuracy in polarization operator here we can put $\omega = 0$, $\mathbf{k} = \mathbf{0}$ so that it reduces to $\Pi(0,0) = -2N(0)$ [23].

Then the phonon spectrum, renormalized by interaction with electrons, is determined by:

$$\Omega_{\mathrm{k}}^2 = \Omega_{0\mathrm{k}}^2 \left[1 + \frac{2|g_{\mathrm{k}}|^2}{\Omega_{0\mathrm{k}}} \Pi(0,0)\right] = \Omega_{0\mathrm{k}}^2 \left[1 - 2\lambda_0^k\right] \tag{25}$$

where we have introduced the usual definition of dimensionless coupling constant of electron – phonon interaction [23]:

$$\lambda_0^k = \frac{2|g_{\mathrm{k}}|^2 N(0)}{\Omega_{0\mathrm{k}}} \tag{26}$$

In this (rough enough) approximation the relatively small damping of phonons due to electron – phonon interaction is just neglected. It can be taken into account by more accurate treatment of the imaginary part of polarization operator which is just zero in the approximation used here.

The spectrum given by Eq. (25) signifies the lattice instability for $\lambda_0^k > 1/2$, when the square of the phonon frequency becomes negative. This instability was correctly interpreted already in the early paper by Fröhlich [24], where it was observed for the first time. Let us rewrite the "dressed" Green's function (23) identically as:

$$D(\mathrm{k}\omega) = \frac{2\Omega_{\mathrm{k}}}{\omega^2 - \Omega_{\mathrm{k}}^2 + i\delta} \frac{\Omega_{0\mathrm{k}}}{\Omega_{\mathrm{k}}} \tag{27}$$

Then it becomes clear that during diagram calculations using from the very beginning this renormalized Green's function of phonons, the physical coupling constant of electron – phonon interaction takes the form (instead of (26)):

$$\lambda^k = \frac{2|g_{\mathrm{k}}|^2 N(0)}{\Omega_{\mathbf{k}}} \frac{\Omega_{0\mathrm{k}}}{\Omega_{\mathrm{k}}} = \frac{2|g_{\mathrm{k}}|^2 N(0)}{\Omega_{0\mathrm{k}}} \frac{\Omega_{0\mathrm{k}}^2}{\Omega_{\mathrm{k}}^2} = \lambda_0^k \frac{\Omega_{0\mathrm{k}}^2}{\Omega_{\mathrm{k}}^2} \tag{28}$$

or, using (25):

$$\lambda^k = \frac{\lambda_0^k}{1 - 2\lambda_0^k} \tag{29}$$

We see, that for $\lambda_0^k \to 1/2$ the *renormalized* coupling constant λ^k monotonously grows and finally diverges. It is this constant that determines the "true" value of electron – phonon interaction (with "dressed" phonons) and there are no limitations for its value at all. This physical picture was discussed in detail in already in Ref. [3].

In the model with single Einstein phonon, which is a reasonable approximation for an optical phonon, we have $\Omega_{\mathrm{k}} = \Omega_0$ and we can forget about dependence of the coupling constant on phonon momentum, so that:

$$\lambda_0 = \frac{2g_0^2 N(0)}{\Omega_0} \tag{30}$$

$$\Omega^2 = \Omega_0^2[1 - 2\lambda_0] \tag{31}$$

$$\lambda = \frac{2g_0^2 N(0)}{\Omega_0} \left(\frac{\Omega_0}{\Omega}\right)^2 = \frac{\lambda_0}{1 - 2\lambda_0} \tag{32}$$

Equation (29) can be reversed and we can write:

$$\lambda_0^k = \frac{\lambda^k}{1 + 2\lambda^k} \tag{33}$$

expressing nonphysical "bare" constant of electron – phonon coupling λ_0^k via the "true" physical coupling constant λ^k. Using this relation in the equation for renormalized phonon spectrum (25), we can write it as:

$$\Omega_{\mathrm{k}}^2 = \Omega_{0\mathrm{k}}^2 \left[1 - \frac{2\lambda^k}{1 + 2\lambda^k}\right] = \Omega_{0\mathrm{k}}^2 \frac{1}{1 + 2\lambda^k} \tag{34}$$

so that in this representation there is no instability of spectrum (lattice), and the growth of λ^k just leads to continuous "softening" of spectrum due to the growth of electron – phonon coupling.

In a model of Einstein phonon all relations simplify and we get:

$$\lambda_0 = \frac{\lambda}{1 + 2\lambda} \tag{35}$$

$$\Omega = \frac{\Omega_0}{\sqrt{1 + 2\lambda}} \tag{36}$$

In Eliashberg – McMillan formalism, where we perform the averaging over the momenta of electrons over the Fermi surface, Eliashberg – McMillan function $\alpha^2(\omega)F(\omega)$, naturally should be determined bu the physical (renormalized) spectrum of phonons. In particular case of Einstein phonon it immediately reduces to (32) and there are no limitations on the value of λ.

In self – consistent derivation of Eliashberg equations we have to use the the phonon Green's function a "dressed"form (23) or (27), which corresponds to the physical (renormalized) phonon spectrum. In this case we *do not have to include* corrections to this function due to electron – phonon interaction, as they are already taken into account in the renormalized phonon spectrum (25).

It should be noted that the value of critical coupling constant obtained above, at which Fröhlich instability of phonon spectrum appears, is obviously directly related to the use of the simplest expression for polarization operator of the gas of free electrons, which was calculated neglecting vertex corrections and self – consistent "dressing" of electron Green's functions entering the loop. Naturally, even in the simplest cases like the problem with Einstein spectrum accounting for these higher corrections, as well as more realistic structure of electron spectrum in a lattice, can somehow change the value of λ_0, corresponding to instability of the "bare" phonon spectrum, so that it will differ from 1/2. In this sense it is better to speak about instability at some "critical" value $\lambda_0^c \sim 1/2$.

This analysis can be significantly improved within the simplified Holstein model, where the electron – phonon interaction is considered to be local (single site), which allows solving this model using the dynamical mean field theory (DMFT) approach [25], which becomes (numerically) exact in the limit of lattice of infinite dimensions (infinite number of nearest neighbors). Such analysis was performed e.g. in Ref. [26], using as quantum Monte – Carlo (QMC) as impurity solver of DMFT. The usual behavior of Fröhlich theory is nicely reproduced with slightly changed λ_0^c=0.464. Behavior similar to Eq. (36) was obtained also for the renormalized phonon frequency Ω.,

The instability appearing at $\lambda_0 = \lambda_0^c$ in Holstein model with half – filled bare band, was convincingly interpreted in Ref. [27] as transition into the state of *bipolaron insulator*. Until this transition the system remains metallic and is nicely described by Eliashberg theory (with insignificant numerical corrections).

It should be stressed here, that all conclusions on instability of metallic phase were done above in the framework of purely *model* approach and in terms of "bare" parameters of λ_0 and Ω_0, which, as was often noted in the literature, are not so well defined physically. The problem here is that the phonon spectrum in a metal, considered as system of ions and electrons, is usually calculated in adiabatic approximation [28]. This spectrum is relatively weakly renormalized due to nonadiabatic effects, which are small over the parameter $\sqrt{\frac{m}{M}}$ [28, 29]. In this respect, it is drastically different from the "bare" spectra of Fröhlich or Holstein models, which, as we have seen above, is significantly renormalized by electron – phonon interaction. The physical meaning of the "bare" spectrum Ω_0 in these models remains not so clear, in contrast to phonon spectrum in metals, calculated in adiabatic approximation.

3.2 Specific Heat Instability?

Recently Semenok et al. [30] proposed a possible limitation for λ due a certain electronic specific heat instability previously derived in Yuzbashyan and Altshuler [31] within Eliashberg – McMillan theory . It was claimed that electronic specific heat in Eliashberg – McMillan theory becomes negative for the values of $\lambda > \lambda_\star$ in a certain temperature interval, signifying thermodynamic instability of electron – phonon system. For Einstein model it was shown that $\lambda_\star$ =3.69 (for Debye model of phonon spectrum $\lambda_\star$ = 4.72). The use of this value in Allen – Dynes expression for T_c (22), immediately leads to $T_c \approx 0.35\Omega$ as an upper limit for superconducting transition temperature (for Einstein model).

However the authors of Ref. [32] disagreed and explicitly shown that the total specific heat in Eliashberg – McMillan theory remains positive for all parameters of the model, at least until adiabatic approximation is valid. i.e. $\lambda < \frac{E_F}{\Omega_0}$. The essence of argumentation is as follows. The free energy per unit volume in Eliashberg – McMillan theory can be shown [32] to be determined as:

$$F = F_{free} + \frac{T}{2}\sum_{m\mathrm{k}} \log[-D^{-1}(\mathrm{k}, i\omega_m)] \tag{37}$$

where F_{free} is the free energy of free electrons and $D(\mathrm{k}, i\omega_m)$ is the renormalized phonon Green's function (23), (27) in Matsubara representation, calculated in the simplest approximation used above. Actually electron – phonon interaction enters here only through this "dressed" function.

After the detailed calculations an explicit expressions for F and specific heat $C_{ep}(T) = -Td^2F/d^2T$ can be derived [32] of which we quote only the limiting forms for $C_{ep}(T)$ for the case of Einstein phonons, using our notations. For $2\pi T \ll \Omega$ (with Ω given by Eq. (31)):

$$C_{ep} = \frac{2\pi^2}{3}N(0)T\left(1 + \frac{\lambda_0}{1-2\lambda_0}\right) = \frac{2\pi^2}{3}N(0)T(1+\lambda) \tag{38}$$

which is the standard result for low – temperature contribution to specific heat with the account of electronic mass renormalization due to interaction with phonons: $m^\star = m(1+\lambda)$. For high temperatures $2\pi T \gg \Omega$:

$$C_{ep} = \frac{2\pi^2}{3} N(0) \left(T + \frac{6E_F}{\pi^2} - \frac{3}{\pi^2} \lambda_0 \frac{\Omega_0^2}{T} \right) \tag{39}$$

Consider now this last expression. The first term here is just the usual contribution from free electrons, the second is the contribution from free phonons with effective frequency, *renormalized* by the interaction with electrons, and the third term is the contribution from electron – phonon interaction. Without the middle term, the specific heat becomes negative below $T \approx 0.39\sqrt{\lambda_0}\Omega_0 = 0.39\sqrt{\lambda}\Omega$ which is higher that superconducting transition temperature in the strong coupling limit. However, with the account of the middle term the full $C_{ep}(T)$ is never negative.

The authors of Ref. [31] argued that the negative contribution from the third term in (39) indicates the normal state instability below a certain T, despite the total $C_{ep}(T)$ remains positive, as the contribution from free phonons has nothing to do with electrons. However, both positive and negative contributions here come from the free energy term $\frac{T}{2}\sum_{m\mathrm{k}} \log[-D^{-1}(\mathrm{k}, i\omega_m)]$, so that both terms should be treated equally. Thus, the electron – phonon coupling generates the negative contribution to $C_{ep}(T)$ and simultaneously gives the rise to *much larger* positive T– independent contribution. The detailed numerical calculations of specific heat for all temperatures and different values of parameters of the model, performed in Ref. [32], has shown that specific heat remains always positive. In our opinion this clearly shows, that there is no specific heat instability discussed in Refs. [30, 31], at least within the standard formalism of equilibrium statistical mechanics.

However, the situation may be more complicated. It was shown in Ref. [33] using the standard kinetic equation approach, that thermal equilibrium between electrons and phonons can actually become unstable for large values of electron – phonon coupling constant. It was claimed that the negative values of electronic specific heat only are sufficient for such instability, leading to the difference between the temperatures of electrons and phonons. These results stress the importance of further studies of possible specific heat instabily in electron – phonon system at large couplings.

4 *Upper* Bound for T_c in the Very Strong Coupling Limit

As we have seen above in the limit of very strong coupling $\lambda \gg 1$ solution of Eliashberg – McMillan equations gives the following expression for T_c:

$$T_c = 0.18\sqrt{\lambda\langle\Omega^2\rangle} \tag{40}$$

It may seem now, that there is no limit for T_c growth due to electron – phonon pairing mechanism in the limit of very strong coupling. The only more or less obvious limit is related to the limits of adiabatic approximation, which is usually considered to be the cornerstone of Eliashberg theory.

In the model with Einstein spectrum of phonons we simply have: $\langle\Omega^2\rangle^{1/2} = \Omega$, where Ω is assumed to be the renormalized phonon frequency. Then (40) reduces to:

$$T_c = 0.18\sqrt{\lambda}\Omega \tag{41}$$

so that seemingly for $\lambda \gg 1$ we can, in principle, obtain even $T_c > \Omega$. However, if we remember the renormalization of phonon spectrum and take into account Eq. (36), we immediately obtain from Eq. (41):

$$T_c = 0.18\sqrt{\lambda}\Omega = 0.18\Omega_0\sqrt{\frac{\lambda}{1+2\lambda}} \tag{42}$$

which in the limit of $\lambda \gg 1$ gives

$$T_c^{max} \approx 0.13\Omega_0, \tag{43}$$

because of significant softening of phonon spectrum. At the same time, as noted above, the physical meaning of "bare" frequency Ω_0 in a metal is rather poorly defined, and in particular it can not be determined from any experiments.

If we just forget about "bare" spectrum of phonons and consider parameters Ω and λ *independent*, we can obtain from Eq. (41) very high values of T_c. A certain , though rather artificial model, leading precisely to this kind of behavior was recently introduced in Ref. [34]. It considered the interaction of N–component electrons with $N \times N$ –component system of Einstein phonons in the limit of $N \to \infty$. It was shown that in this model the renormalization of phonon spectrum due to interaction with conduction electrons is suppressed, so that in the limit of very strong coupling with $1 \ll \lambda \ll N$ we always obtain Allen – Dynes estimate (41) with $\Omega = \Omega_0$.

However, the problem here is, that in real situation we never can consider Ω and λ as independent parameters simply because of the general relations (6) and (7), which express λ and $\langle\Omega^2\rangle$ via integrals of Eliashberg – McMillan function $\alpha^2(\omega)F(\omega)$. In fact, we may rewrite the expression for T_c in the region of very strong coupling as:

$$T_c = 0.18\sqrt{\lambda\langle\Omega^2\rangle} = 0.25\left[\int_0^\infty d\omega\alpha^2(\omega)F(\omega)\omega\right]^{1/2} \tag{44}$$

We see, that this expression for T_c is completely determined by the integral of $\alpha^2(\omega)F(\omega)$ over the phonon spectrum,

while there is no explicit dependence of T_c on λ and $\langle\Omega^2\rangle$ separately.

Experimental discovery of high – temperature superconductivity in hydrides under high (megabar) pressures [9] stimulated the search for the ways to achieve superconductivity at room temperature [10–13]. At the moment the common view [14, 15] is that the high – temperature superconductivity in hydrides can be described in the framework of the standard Eliashberg – McMillan theory. Within this theory many attempts were undertaken to estimate the maximal achievable superconducting transition temperature and the discussion of some of these attempts can be found in the reviews [14, 15, 23].

In Ref. [35] a another simple inequality for T_c was proposed, limiting its value by the square A under $\alpha^2(\omega)F(\omega)$:

$$T_c \leq 0.2309 \int_0^\infty d\omega \alpha^2(\omega)F(\omega) \equiv 0.2309A \tag{45}$$

For the case of Einstein spectrum of phonons this can be rewritten as:

$$T_c \leq 0.115\lambda\Omega_0 \tag{46}$$

This inequality is relatively often used in the literature.

In Ref. [37] a semi empirical limit for T_c was proposed for conventional semiconductors, which can be written in a very simple form:

$$k_B T_c \leq A_{max}\Theta_D = A_{max}\hbar\Omega_D \tag{47}$$

where $A_{max} \approx 0.10$, and $\Theta_D = \hbar\Omega_D$ is Debye temperature, which may be determined e.g. from standard measurements of specific heat. This inequality obviously correlates with T_c^{max}, obtained above in the limit of $\lambda \to \infty$ in Eq. (42), if we identify Ω_0 with Ω_D. It is seen from Fig. 3 this limitation is satisfied for most of conventional superconductors [37].

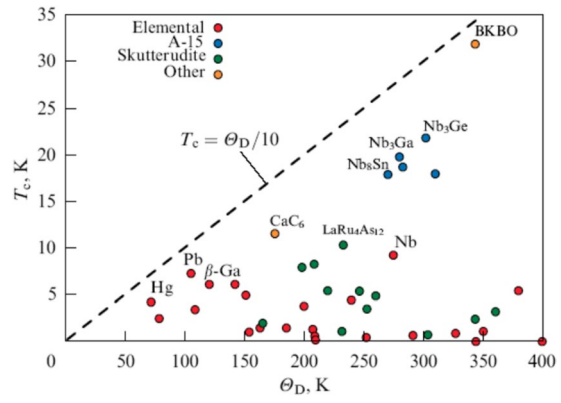

Fig. 3 Experimental values of the temperature of superconducting transition for conventional superconductors dependence on their Debye temperature Θ_D [37]

In the recent paper [36] a new upper limit for T_c was proposed, expressed via certain combination of fundamental constants. Below we show that with minor modifications such T_c limit follows directly from Eliashberg – McMillan theory. The relation of λ and $\langle\Omega^2\rangle$ is clearly expressed by McMillan's formula for λ (8). Equation (8) gives very useful representation for the coupling constant λ, which is routinely used in the literature and in practical (first – principles) calculations [15, 39].

Using Eq. (8) in Eq. (40) we immediately obtain:

$$T_c^\star = 0.18\sqrt{\frac{N(0)\langle I^2\rangle}{M}} \tag{48}$$

where $\langle I^2\rangle$ was defined in Eq. (4). Both λ and $\langle\Omega^2\rangle$ just drop out from the expression for $T_c^\star$, which is now expressed via Fermi surface averaged matrix element of the gradient of electron – ion potential, ion mass and electronic density of states at the Fermi level.

As was already noted, all parameters entering this expression can be rather simply obtained during the first – principles calculations of T_c for specific materials (compounds) [15, 39]. Let us also stress that the value of $T_c^\star$ defined in Eq. (48), calculated for any specific material does not have any direct relation to real value of T_c, but just defines precisely the upper limit of T_c, which "would be achieved" in the limit of strong enough electron – phonon coupling. Below we shall present some rough heuristic (dimensional) estimates of its value [38].

In the following we assume to be dealing with three – dimensional metal with cubic symmetry with an elementary cell with lattice constant a and just one conduction electron per atom. Then we can estimate the density of states at the Fermi level as for free electrons: $N(0) = \frac{mp_F}{2\pi^2\hbar^3}a^3$, where $p_F \sim \hbar/a$ is the Fermi momentum, m is the mass of free (band) electron. Electron – ion potential (single – charged ion, e is electron charge) can be estimated as:

$$V_{ei} \sim \frac{e^2}{a} \sim e^2 p_F/\hbar \tag{49}$$

so that its gradient is:

$$\nabla V_{ei} \sim \frac{e^2}{a^2} \sim e^2 p_F^2/\hbar^2 \tag{50}$$

Then we easily obtain the following estimate of (4):

$$I^2 \sim \left(\frac{e^2}{a^2}\right)^2 \sim (e^2 p_F^2/\hbar^2)^2 \tag{51}$$

which is probably too optimistic, as we neglected all the fine details, which were analyzed e.g. in Pickett [39]. Here we also dropped different numerical factors of the order of unity, which are obviously not so important for our order of magnitude estimates. Now we obtain an estimate for $T_c^\star$ from Eq. (48) as:

$$T_c^\star \sim 0.2\sqrt{\frac{m}{M}}\frac{e^2}{\hbar v_F}E_F \tag{52}$$

where $E_F = p_F^2/2m$ is Fermi energy, $v_F = p_F/m$ is electron velocity at the Fermi surface. The value of $\frac{e^2}{\hbar v_F}$, as is well known, represents the dimensionless coupling for Coulomb interaction and for typical metals it is of the order of or greater than unity. The factor of $\sqrt{\frac{m}{M}}$ determines isotopic effect.

Let us measure length in units of Bohr radius a_B introducing the standard dimensionless parameter r_s by the relation $a^3 = \frac{4\pi}{3}(r_s a_B)^3$. Then we have:

$$a \sim r_s a_B = r_s\frac{\hbar^2}{me^2} = r_s\frac{\hbar}{mc\alpha} \tag{53}$$

where we have introduced the fine structure constant $\alpha = \frac{e^2}{\hbar c}$. Correspondingly the Fermi momentum is given by:

$$p_F \sim \frac{\hbar}{r_s a_B} = \frac{me^2}{\hbar r_s} = \frac{mc}{\hbar r_s}\alpha \tag{54}$$

Then $T_c^\star$ (48) can be rewritten as:

$$T_c^\star \sim \frac{0.2}{r_s}\sqrt{\frac{m}{M}}\alpha^2 mc^2/2 = \frac{0.2}{r_s}\sqrt{\frac{m}{M}}\frac{me^4}{2\hbar^2} = \frac{0.2}{r_s}\sqrt{\frac{m}{M}}Ry \tag{55}$$

where $Ry = me^4/2\hbar^2 \approx 13.6$ eV is the Rydberg constant. Here we have obtained the same combination of fundamental (atomic) constants, which was actually suggested in Ref. [36], by some quite different reasoning, as determining the upper limit of superconducting critical temperature. However, our expression contains an extra factor of r_s^{-1}, which necessarily reflects the specifics of a material under consideration (density of conduction electrons), so that the value of $T_c^\star$ is in no sense universal. Apparently in Ref. [36] it was somehow implicitly assumed the value of $r_s = 1$.

For metallic hydrogen *M* is equal to proton mass and we have $\sqrt{\frac{m}{m_p}} \sim 0.02$, so that for $r_s = 1$ we get an estimate of $T_c^\star \sim 650$ K. This is in nice agreement with the result of $T_c = 600$ K, obtained in Ref. [40, 41] solving Eliashberg equations for FCC lattice of metallic hydrogen with $r_s = 1$, taking into account the calculated softening of the phonon spectrum, leading to realizations of very strong coupling ($\lambda = 6.1$).

5 Metallic Hydrogen and the "jellium" Model: Insufficiency of the Weak – Coupling

In the recent paper [42] an elegant study of superconductivity of metallic hydrogen was performed within the "jellium" model [3, 43]. In this model it is rather easy to calculate all the relevant parameters to calculate T_c (cf. section Methods of Ref. [42]) in weak – coupling BCS approach or for the general case of Eliashberg – McMillan theory .

The upper boundary for the phonon frequency in this model is just given by the ion plasma frequency (we again consider the single – charged ions):

$$\omega_0 = \sqrt{\frac{4\pi ne^2}{M}} = 2Ry\sqrt{\frac{3m}{M}}\frac{1}{r_s^{3/2}} \tag{56}$$

where *n* is conduction electron density. Note that the combination of fundamental constants here is the same as in Eq. (55), but r_s – dependence is different.

Electron – phonon and Coulomb dimensionless coupling constants are given by van der Marel and Berthod [42]:

$$\lambda = \mu = \frac{1}{2\pi p_F a_0/\hbar}\ln(1+\pi p_F a_0/\hbar) = \\ = \frac{r_s}{2\pi(\frac{9\pi}{4})^{1/3}}\ln\left[1+\pi\left(\frac{9\pi}{4}\right)^{1/3}r_s^{-1}\right] \tag{57}$$

Coulomb pseudopotential $\mu^\star$ is given by the usual expression:

$$\mu^\star = \frac{\mu}{1+\mu\ln\frac{E_F}{\omega_0}} \tag{58}$$

where

$$\frac{E_F}{\omega_0} = \frac{1}{2}\frac{\left(\frac{9\pi}{4}\right)^{2/3}}{\sqrt{\frac{3m}{M}}}r_s^{-1/2} \tag{59}$$

In principle the "jellium" model is more or less valid if we assume that perturbation theory in Coulomb interaction can be applied, that is only for $r_s < 1$. However, we shall use all these expressions for rather wide region of r_s as it was done in van der Marel and Berthod [42].

Now consider metallic hydrogen and take $M = m_p$. Then the characteristic phonon frequency ω_0 given by Eq. (56) is pretty large and changes between circa 12000 K and 2000 K as r_s changes between typical values of 1.0 and 3.5 (as in usual metals). As this frequency enters as a preexponential factor in BCS – like expressions for T_c we can expect for it rather large values [2–4].

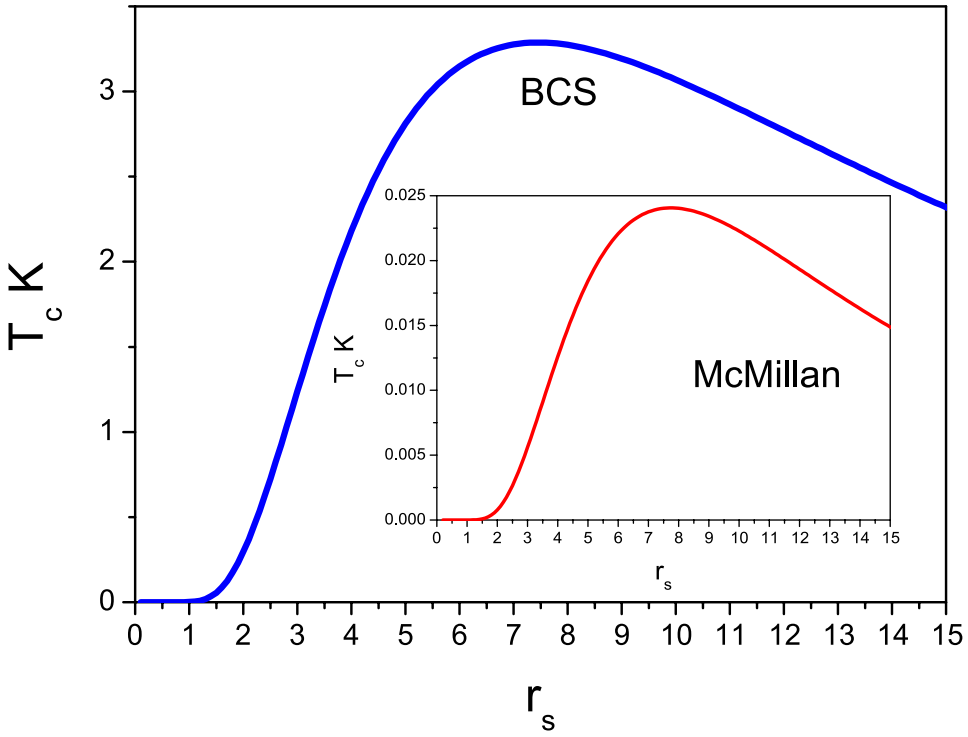

Fig. 4 Superconducting transition temperature of metallic hydrogen in "jellium" model calculated from the weak – coupling BCS expression. At the insert: the same calculated from McMillan formula for intermediate coupling

However, these expectations are not realized when we calculate coupling constants. Direct calculations show that $\lambda = \mu < 1/2$ for all values of r_s and change between 0.17 – 0.3 for most relevant values of r_s =1.0 – 3.5, signifying the weak coupling regime. For the same interval of r_s the Coulomb pseudopotential $\mu^\star$ changes between 0.1 and 0.15, as is usually assumed.

Then calculating T_c for the wide range of r_s, using Eqs. (56), (57), (58), (59) in the standard weak – coupling BCS expression ($\gamma = 1.78$ is Euler constant):

$$T_c = \frac{2\gamma}{\pi}\omega_0 \exp\left(-\frac{1}{\lambda - \mu^\star}\right) \tag{60}$$

we obtain the results shown in Fig. 4. We can see the characteristic maximum of T_c at $r_s \sim 7$, but the value of T_c at this maximum only slightly exceeds 3K.

Even more pessimistic results are obtained if we calculate T_c taking into account intermediate coupling corrections and using McMillan formula [42]:

$$T_c = \frac{\omega_0}{1.45} \exp\left[-\frac{1.04(1+\lambda)}{\lambda - \mu^\star(1+0.62\lambda)}\right] \tag{61}$$

which are shown at the insert in Fig. 4. Here we again observe the characteristic maximum of T_c, but it is less than 25 mK only! We must stress the great sensitivity of these results to rather small change of the values of main parameters, even such as the relatively weak r_s – dependence of the Coulomb pseudopotential as given by Eq. (58).

However, we clearly see that in the weak (or intermediate) coupling approximations the "jellium" model can not produce high values of T_c for metallic hydrogen [42].

Somewhat better results were obtained in Ref. [42] by numerical solution of BCS integral gap equation with the explicit use of the screened "jellium" – like interaction kernel. Still, only the values of T_c not exceeding 30K were found at $r_s \sim 3$, which is also much lower than the optimistic estimates of Refs. [2–4]. Actually the great difference obtained in Ref. [42] between the results of direct solution of the integral gap equation and more or less obvious use of the standard BCS – like of McMillan approximations is somehow obscure. We can only note, that the authors of Ref. [42] used the "jellium" model interaction as direct input into BCS gap (integral) equation, though it is known since the famous KMK paper [44], that it should be actually replaced by some effective (smoothed) integral kernel. This remains to be done for the correct analysis and we just leave the use of KMK formalism in "jellium" model for the future work.

As to real metallic hydrogen, we must stress that the "jellium" model is only an oversimplified approximation, strictly valid for $r_s < 1$ and neglecting all the crystal structure effects in solid metallic hydrogen. At present there is no consensus on the stable crystal structures of metallic

hydrogen at all. It is clear from our discussion above that the strong coupling regime can be actually achieved, e.g. due to some lattice (phonon spectrum) softening effects, as was demonstrated in Refs. [40, 41]

6 Conclusion

Eliashberg theory remains the main theory, which completely explains the values of the critical temperature in superconductors with electron – phonon mechanism of pairing. This theory is also applicable in the region of strong electron – phonon coupling, limited only by the applicability of adiabatic approximation, based on Migdal theorem, which is valid in the vast majority of metals, including the new superhydrides with record values of T_c. The values of (renormalized, physical) pairing coupling constant λ can surely exceed unity until the system possess the metallic ground state.

A number of simple expressions can be derived to estimate the maximal value of T_c in terms of experimentally measurable or calculable parameters like characteristic (average) values of phonon frequencies and pairing coupling constant. Actually this maximal value of T_c is just determined by some "game" of atomic constants. These estimates of the maximal possible T_c are rather optimistic, and the perspective for the experimental search for its higher values, e.g. in hydrides, seems still very interesting. However, all the present day data on superhydrides strongly indicate that all these systems are actually very close to the strong coupling region of Eliashberg theory, which indicate that the maximal values of T_c for metals are probably more or less already achieved in these experiments. In this respect the metallic hydrogen remains in our opinion probably the most promising candidate system [40, 41].

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Author Contributions Michael Sadovskii authored the whole paper as a single author.

Data Availability No datasets were generated or analysed during the current study.

Declarations

Competing interests The authors declare no competing interests.

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