Temperature of superconducting transition for very strong coupling in antiadiabatic limit of Eliashberg equations

M. V. Sadovskii¹⁾,

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

It is shown that the famous Allen – Dynes asymptotic limit for superconducting transition temperature in very strong coupling region $T_c > \frac{1}{2\pi}\sqrt{\lambda}\Omega_0$ (where $\lambda \gg 1$ – is Eliashberg – McMillan electron – phonon coupling constant and Ω_0 – the characteristic frequency of phonons) in antiadiabatic limit of Eliashberg equations $\Omega_0/D \gg 1$ ($D \sim E_F$ is conduction band half-width and E_F is Fermi energy) is replaced by $T_c > (2\pi^4)^{-1/3} (\lambda D \Omega_0^2)^{1/3}$, with the upper limit for T_c given by $T_c < \frac{2}{\pi^2} \lambda D$.

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1. INTRODUCTION

The discovery of superconductivity [1] with critical temperature up to $T_c = 203$ in pressure interval of 100-250 GPa (in diamond anvils) in H₃S system initiated the flow of articles with experimental studies of high – temperature superconductivity of hydrides in megabar region (cf. reviews [2, 3]). Theoretical analysis immediately confirmed that these record – breaking values of T_c are ensured by traditional electron – phonon interaction in the limit of strong – enough electron – phonon coupling [4, 5]. More so, the detailed calculations performed for quite a number of hydrides of transition metals under pressure [4] lead to prediction of pretty large number of such systems with record values of T_c . In some cases these predictions were almost immediately confirmed by experiment, in particular the record values of $T_c = 160-260 \text{ K}$ were achieved in in LaH₁₀ [6, 7], ThH₁₀ $[8], YH_6 [9], (La, Y)H_{6,10} [10]$. At last, some time ago the psychological barrier was overpassed, when in Ref. [11] superconductivity was obtained with $T_c = 287.7 \pm 1.2$ K (i.e. near +15 degrees of Celsius) in C-H-S system at pressure of 267 ± 10 GPa.

The principal achievement of these works was, before everything else, the demonstration of absence of any significant limitations for T_c , within the traditional picture of electron – phonon mechanism of Cooper pairing, contrary to a common opinion that T_c due to it can not exceed 30-40 K. Correspondingly, even more demanding now is the problem of the upper limit of T_c values, which can be achieved with this mechanism of pairing.

Since BCS theory appeared it became obvious the the increase of T_c can be achieved either by the increase of the frequency of phonons, responsible for Cooper pairing, or by the increase of the effective interaction of these phonons with electrons. These problems were

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thoroughly studied by different authors. The most developed approach to description of superconductivity in electron - phonon system is Eliashberg - McMillan theory [5, 12, 13]. It is well known that this theory is entirely based on the applicability of adiabatic approximation and Migdal theorem [15], which allows to neglect vertex corrections while calculating the effects of electron – phonon interactions in typical metals. The actual small parameter of perturbation theory in these calculations is $\lambda \frac{\Omega_0}{E_F} \ll 1$, where λ is the dimensionless coupling constant of electron – phonon interactions, Ω_0 is characteristic frequency of phonons and E_F is Fermi energy of electrons. In particular, this means that vertex corrections in this theory can be neglected even in case of $\lambda > 1,$ as we always have an inequality $\frac{\Omega_0}{E_F} \ll 1$ valid for typical metals.

In recent papers [16, 17, 18] we have shown that in case of strong nonadiabaticity, when $\Omega_0 \gg E_F$, a new small parameter appears in the theory $\lambda_D \sim \lambda \frac{E_F}{\Omega_0} \sim \lambda \frac{D}{\Omega_0} \ll 1$ (*D* is electronic band half-width), so that corrections to electronic spectrum become irrelevant. Vertex corrections can also be neglected, as it was shown in an earlier Ref. [19]. In general case the renormalization of electronic spectrum (effective mass of an electron) is determined by a new dimensionless constant $\tilde{\lambda}$, which reduces to the usual λ in the adiabatic limit, while in strong antiadiabatic limit it tends to λ_D . At the same time, the temperature of superconducting transition T_c in antiadiabatic limit is determined by Eliasnberg – McMillan pairing constant λ , generalized by the account of finite phonon frequencies.

For the case of interaction with a single optical (Einstein) phonon in Ref. [16] we have obtained the single expression for T_c , which is valid both in adiabatic and

¹⁾E-mail: sadovski@iep.uran.ru

antiadiabatic regimes and smoothly interpolating in between: $\tilde{}$

$$T_c \sim \frac{D}{1 + \frac{D}{\Omega_0}} \exp\left(-\frac{1+\lambda}{\lambda}\right)$$
 (1)

where $\tilde{\lambda} = \lambda \frac{D}{\Omega_0 + D}$ is smoothly changing from λ for $\Omega_0 \ll D \sim E_F$ to λ_D in the limit of $\Omega_0 \gg D \sim E_F$.

Besides the questions related to possible limits of T_c in hydrides, where possibly some small pockets of the Fermi surface with small Fermi energies exist [5], the interest to the problem of superconductivity in strongly antiadiabatic limit is stimulated by the discovery of a number of other superconductors, where adiabatic approximation can not be considered valid and characteristic phonon frequencies is of the order or even exceed the Fermi energy of electrons. Typical in this respect are intercalated systems with monolayers of FeSe, and monolayers of FeSe on substrates like $Sr(Ba)TiO_3$ (FeSe/STO) [20]. With respect to FeSe/STO this was first noted by Gor'kov [21, 22], while discussing the idea of the possible mechanism of increasing superconducting transition temperature T_c in FeSe/STO due to interactions with high- energy optical phonons of SrTiO₃ [20]. Similar situation appears also in an old problem of superconductivity in doped $SrTiO_3$ [23].

2. LIMITS FOR SUPERCONDUCTING TRANSITION TEMPERATURE IN CASE OF VERY STRONG ELECTRON – PHONON COUPLING

The general equations of Eliashberg – McMillan theory determining superconducting gap $\Delta(\omega_n)$ in Matsubara representation ($\omega_n = (2n+1)\pi T$) can be written as [5, 12, 13]:

$$\Delta(\omega_n)Z(\omega_n) = T \sum_{n'} \int_{-D}^{D} d\xi \int_0^{\infty} d\omega \alpha^2(\omega)F(\omega) \times D(\omega_n - \omega_{n'};\omega) \frac{\Delta(\omega'_n)}{\omega''_n + \xi^2 + \Delta^2(\omega_{n'})}$$
(2)

$$Z(\omega_n) = 1 + \frac{\pi T}{\omega_n} \sum_{n'} \int_{-D}^{D} d\xi \int_0^{\infty} d\omega \alpha^2(\omega) F(\omega) \times D(\omega_n - \omega_{n'}; \omega) \frac{\omega'_n}{\omega_{n'}^2 + \xi^2 + \Delta^2(\omega_{n'})}$$
(3)

where we have introduced

$$D(\omega_n - \omega_{n'}; \omega) = \frac{2\omega}{(\omega_n - \omega_{n'})^2 + \omega^2}$$
(4)

Here $\alpha^2(\omega)F(\omega)$ is McMillan's function, $F(\omega)$ is the phonon density of states, and for simplicity we assume

here the model of half-filled band of electrons with finite width $2D (D \sim E_F)$ with constant density of states (two – dimensional case).

We also neglect here the effects of Coulomb repulsion leading to the appearance of Coulomb pseudopotential μ^* , which is usually small and more or less irrelevant in the region of very strong electron – phonon attraction [5, 12, 13].

Then, taking into account:

$$\int_{-D}^{D} d\xi \frac{1}{\omega_{n'}^{2} + \xi^{2} + \Delta^{2}(\omega_{n'})} =$$

$$= \frac{2}{\sqrt{\omega_{n'}^{2} + \Delta^{2}(\omega_{n'})}} \operatorname{arctg} \frac{D}{\sqrt{\omega_{n'}^{2} + \Delta^{2}(\omega_{n'})}} \rightarrow$$

$$\to \frac{2}{|\omega_{n'}|} \operatorname{arctg} \frac{D}{|\omega_{n'}|} \operatorname{при} \Delta(\omega_{n'}) \rightarrow 0 \quad (5)$$

the linearized Eliashberg equations take the following general form:

$$\Delta(\omega_n)Z(\omega_n) = T \sum_{n'} \int_0^\infty d\omega \alpha^2(\omega)F(\omega) \times \\ \times D(\omega_n - \omega_{n'};\omega) \frac{2\Delta(\omega_{n'})}{|\omega_{n'}|} \operatorname{arctg} \frac{D}{|\omega_{n'}|}$$
(6)

$$Z(\omega_n) = 1 + \frac{T}{\omega_n} \sum_{n'} \int_0^\infty d\omega \alpha^2(\omega) F(\omega) \times \\ \times D(\omega_n - \omega_{n'}; \omega) \frac{\omega_{n'}}{|\omega_{n'}|} 2 \operatorname{arctg} \frac{D}{|\omega_{n'}|}$$
(7)

Consider the equation for n = 0 determining $\Delta(0) \equiv \Delta(\pi T) = \Delta(-\pi T)$, which follows directly from Eqs. (6), (7):

$$\Delta(0) = T \sum_{n' \neq 0} \int_0^\infty d\omega \alpha^2(\omega) F(\omega) \frac{2\omega}{(\pi T - \omega_{n'})^2 + \omega^2} \times \frac{2\Delta(\omega_{n'})}{|\omega_{n'}|} \operatorname{arctg} \frac{D}{|\omega_{n'}|}(8)$$

Leaving in the r.h.s. only the contribution from n' = -1, we immediately obtain the *inequality*:

$$1 > \frac{2}{\pi} \int_0^\infty d\omega \alpha^2(\omega) F(\omega) \frac{2\omega}{(2\pi T)^2 + \omega^2} \operatorname{arctg} \frac{D}{\pi T} \quad (9)$$

which generalizes the similar inequality first obtained in Allen – Dynes paper [14] and determining the *lower* boundary for T_c . For Einstein model of phonon spectrum we have $F(\omega) = \delta(\omega - \Omega_0)$, so that Eq. (9) is reduced to:

$$1 > \frac{2}{\pi} \lambda arctg \frac{D}{\pi T} \frac{\Omega_0^2}{(2\pi T)^2 + \Omega_0^2}$$
(10)

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where $\lambda = 2\alpha^2(\Omega_0)/\Omega_0$ is dimensionless pairing coupling constant. For $D \gg \pi T$ we immediately obtain the Allen – Dynes result [14]:

$$T_c > \frac{1}{2\pi} \sqrt{\lambda - 1} \Omega_0 \to 0.16 \sqrt{\lambda} \Omega_0$$
 при $\lambda \gg 1$ (11)

which in fact determines the asymptotic behavior of T_c in the region of very strong coupling $\lambda \gg 1$. The exact numerical solution of Eliashberg equation [14] produces for T_c the result like (11) with replacement of numerical coefficient 0.16 by 0.18. This asymptotic behavior rather satisfactory describes the values of T_c already for $\lambda > 2$.

In the case of general phonon spectrum it is sufficient to replace here $\Omega_0 \to \langle \Omega^2 \rangle^{1/2}$, where

$$\langle \Omega^2 \rangle = \frac{2}{\lambda} \int_0^\infty d\omega \alpha^2(\omega) F(\omega) \omega$$
 (12)

is the average (over the spectrum) square frequency of phonons, and the general expression for the coupling constant is [5, 12, 13]:

$$\lambda = 2 \int_0^\infty \frac{d\omega}{\omega} \alpha^2(\omega) F(\omega) \tag{13}$$

For $D \ll \pi T$ from Eq. (10) we obtain

$$T > \frac{1}{2\pi} \sqrt{\lambda^*(T) - 1} \Omega_0 \tag{14}$$

where

$$\lambda^*(T) = \frac{2D}{\pi^2 T} \lambda \tag{15}$$

so that in strongly antiadiabatic limit we get:

$$T_c > (2\pi^4)^{-1/3} (\lambda D\Omega_0^2)^{1/3} \approx 0.17 (\lambda D\Omega_0^2)^{1/3}$$
 (16)

From the obvious requirement of $\lambda^*(T) > 0$ we obtain the condition:

$$\Gamma_c < \frac{2}{\pi^2} \lambda D \tag{17}$$

which limits T_c from the above.

Thus we require the validity of the following inequality:

$$(2\pi^4)^{-1/3} (\lambda D\Omega_0^2)^{1/3} < T_c < \frac{2}{\pi^2} \lambda D$$
 (18)

which reduces to:

$$\Omega_0 < \frac{4}{\pi} \lambda D \approx 1.27 \lambda D$$
 или $\frac{D}{\Omega_0} > \frac{0.78}{\lambda}$ (19)

so that for our analysis to be self – consistent it is required to have:

$$\lambda \gg \frac{\Omega_0}{D} \gg 1 \tag{20}$$

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Fig. 1. Temperature of superconducting transition in Einstein model of phonon spectrum in units of $2\pi T_c/\Omega_0$, as function of pairing constant λ for different values of the inverse adiabaticity parameter $\frac{D}{\Omega_0}$. Dotted lines show the dependencies for $2\pi T_c/\Omega_0$ in the region of weak and intermediate couplings (1) [16]. Black dashed line — Allen – Dynes estimate valid in adiabatic limit [14]



Fig. 2. Temperature of superconducting transition in Einstein model of phonon spectrum in units of $2\pi T_c/D$, as function of pairing constant λ fro different values of adiabaticity parameter $\frac{\Omega_0}{D}$. Dotted lines show the dependencies for $2\pi T_c/D$ in the region of weak and intermediate couplings (1) [16]. Black line — the limiting behavior given by $\frac{2}{\pi^2}\lambda D$

where the last inequality corresponds to strong antiadiabatic limit. Correspondingly, all the previous estimates are not valid for $\lambda \sim 1$ and can only describe the limit of very strong coupling.

In Fig.1 and Fig. 2 we show the results of numerical comparison of the boundaries for T_c , following from Eq. (10) with the values of transition temperature in the re-

gion of weak and intermediate coupling following from Eq. (1), for different values of adiabaticity parameter Ω_0/D . It is clear that in the vicinity of intersections of dotted and continuous lines on these graphs we actually have the smooth crossover from T_c behavior in the region of weak and intermediate coupling to its asymptotic behavior in the region of very strong coupling $\lambda \gg 1$. It is also seen that the increase of phonon frequencies and crossover to antiadiabatic limit does not lead, in general, to the increase of T_c as compared with adiabatic case.

3. CONCLUSIONS

In this paper we have considered the case of very strong electron - phonon coupling in Eliashberg -McMillan theory, including the antiadiabatic situation with phonons of very high frequency (exceeding the Fermi energy E_F). The value of mass renormalization is in general determined by the coupling constant λ [16], which is small in antiadiabatic limit. At the same time, the pairing interaction is always determined by the standard coupling constant λ of Eliashberg – McMillan theory, appropriately generalized by taking into account the finite values of phonon frequencies [16]. However, the simplest estimates [16, 18] show, that in antiadiabatic situation this constant in general rather rapidly drops with the growth of phonon frequency Ω_0 for $\Omega_0 \gg E_F$. In this sense, the asymptotics of T_c for very strong coupling, discussed above, can be possibly achieved only in some exceptional cases. Even in this case, as it is clear from our results, the transition into antiadiabatic region can not increase T_c as compared with the standard adiabatic situation.

While the usual expression for T_c in terms of pairing constant λ and characteristic phonon frequency $\Omega_0 \sim \langle \Omega^2 \rangle^{1/2}$ are quite convenient and clear, it is to be taken into account that these parameters are in fact not independent. As it is seen from expressions like (12) and (13), these parameters are determined by the same Eliashberg – McMillan function $\alpha^2(\omega)F(\omega)$. Correspondingly, there are limitations for free changes of these parameters in estimates of optimal (maximal) values of T_c .

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