The higher superconducting transition temperature $T_c$ and the functional derivative of $T_c$ with $\alpha^2 F(\omega)$ for electron–phonon superconductors

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Abstract

This work presents an analysis of the functional derivative of the superconducting transition temperature $T_c$ with respect to the electron-phonon coupling function $\alpha^2 F(\omega)$ $[\delta T_c / \delta \alpha^2 F(\omega)]$ and $\alpha^2 F(\omega)$ spectrum of H$_3$S (Im$\overline{3}$m), in the pressure range where the high-$T_c$ was measured (155–225 GPa). The calculations are done in the framework of the Migdal–Eliashberg theory. We find for this electron–phonon superconductor, a correlation between the maximums of $\delta T_c / \delta \alpha^2 F(\omega)$ and $\alpha^2 F(\omega)$ with its higher $T_c$. We corroborate this behavior in other electron–phonon superconductors by analyzing data available in the literature, which suggests its validity in this type of superconductors. The correlation observed could be considered as a theoretical tool that in an electron–phonon superconductor, allows describing qualitatively the proximity to its highest $T_c$, and determining the optimal physical conditions (pressure, alloying or doping concentration) that lead to the superconductor reaching its highest $T_c$ possible.

Keywords: Migdal–Eliashberg theory, functional derivative, high-$T_c$ superconductivity, electron–phonon superconductor

(Some figures may appear in colour only in the online journal)

1. Introduction

From the linearized version of the Migdal–Eliashberg gap equations, it is possible to obtain the functional derivative of the superconducting transition temperature $T_c$ with respect to the electron–phonon coupling function $\alpha^2 F(\omega)$ $[\delta T_c / \delta \alpha^2 F(\omega)]$, from which the effectiveness of the phonons at different frequencies in building up the superconducting state can be evaluated [1, 2]. The first numerical calculations of the $\delta T_c / \delta \alpha^2 F(\omega)$ were performed by Bergmann and Rainer [3, 4] for isotropic superconductors. Their results showed that this function has a universal shape: it increases from zero at $\omega = 0$ to a maximum at $\sim 7k_B T_c$, and then decreases slowly to 0 as $\omega \to \infty$ [5]. This maximum is related to the importance of relaxation effects in the electron–phonon interaction as it enters superconductivity [1]. The functional derivative describes the change in $\Delta T_c$ to a small change in the function $\Delta \alpha^2 F(\omega)$ due to pressure or alloying [5, 6]

$$\Delta T_c = \int_0^{+\infty} d\omega \frac{\delta T_c}{\delta \alpha^2 F(\omega)} \Delta \alpha^2 F(\omega)$$  \hspace{1cm} (1)

Some researches [7–11] have studied the derivation and characteristics of the functional derivative for isotropic and anisotropic superconductors, and other few works [1, 5, 6, 12] have explored its application an important number of con-
oretical studies on new high-
T ordering were important to point out that the application of Migdal–Eliashberg theory, in this case, was possible since the superconductivity mechanism in HfS is the electron–phonon interaction [15]. Overall, the theoretical studies on new high-Tc superconductors are aimed at fully understanding the superconducting mechanism for achieving the prediction of Tc in new similar compounds. In this paper, we report a study of the functional derivative of Tc with α2F(ω) and α2F(ω) spectrum for HfS under pressure. For this purpose, we calculate the δTc/δα2F(ω) from the linearized version of the Migdal–Eliashberg gap equations and work with α2F(ω) spectra provided by a previous paper [16]. Our results are compared with data available in the literature for other conventional superconductors [Nb3Ge and Li (bcc)].

2. Method of calculation

The functional derivative of Tc with α2F(ω), δTc/δα2F(ω), is determined by using, in a first step, the solution of the linearized Migdal–Eliashberg equations on the imaginary axis [3, 4, 17](n, m are integers):

\[
\rho T \delta T_c = \pi T \sum_m \left( \lambda_{mn} - \mu^* - \delta_{nm} \frac{\omega_n}{\pi T} \right) \bar{\Delta}_n, \tag{2}
\]

with

\[
\bar{\omega}_n = i \omega_n + \pi T \sum_m \lambda_{nm} \text{sgn}(\omega_m), \tag{3}
\]

where at temperature T, \( \omega_n = \pi T(2n - 1) \) is the n-th Matsubara frequency; and

\[
\Delta_n = \frac{\Delta_n}{\rho + |\omega_n|}, \tag{4}
\]

in terms of \( \Delta_n \), the superconductor gaps, \( \rho \) is a pair break parameter without physical meaning. The solution \( T = T_c \) is getting at the limit when \( \rho \rightarrow 0 \). The kernel \( K_{nm} \) is given by,

\[
K_{nm} = \pi T \left[ \lambda_{nm} - \mu^* - \delta_{nm} \frac{|\omega_n|}{\pi T} \right]. \tag{5}
\]

Then a \( \delta K_{nm} \) variation drives to change \( \delta \rho \),

\[
\delta \rho = \frac{\sum_n \Delta_n \delta K_{nm} \Delta_m}{\sum_n \Delta_n^2}, \tag{6}
\]

where \( \Delta_n \) is the corresponding eigenvector to \( \rho = 0 \) and \( \delta K_{nm} \) is evaluated at \( T = T_c \). Finally, functional derivative is calculated by

\[
\frac{\delta T_c}{\delta \alpha^2 F(\omega)} = \frac{\delta \rho}{\alpha^2 F(\omega) \rho}, \tag{7}
\]

where to \( \left( \frac{\delta K}{\delta \alpha^2 F(\omega)} \right) \) were used

\[
\left( \frac{\delta K_{nm}}{\delta \alpha^2 F(\omega)} \right)_{T_c} = \pi T_c \left[ \left( \frac{\delta \lambda_{nm}}{\delta \alpha^2 F(\omega)} \right)_{T_c} - \delta_{nm} \sum_m \left( \frac{\delta \lambda_{nm}}{\delta \alpha^2 F(\omega)} \right)_{T_c} \text{sgn}(\omega_m^q \omega_n^q) \right]. \tag{8}
\]

The Eliashberg spectral function \( \alpha^2 F(\omega) \) calculated at different pressures were taken from a previous work [16], which were obtained in the pressure range where the high Tc was measured (155–225 GPa) [14]. The dynamic stability of HfS \( Tm \) in this pressure range was previously confirmed [16].

3. Results and discussion

The Eliashberg spectral functions \( \alpha^2 F(\omega) \) and functional derivative \( \delta T_c/\delta \alpha^2 F(\omega) \) for HfS calculated at different pressures are shown in figure 1. By and large, our \( \alpha^2 F(\omega) \) spectra are in good agreement with previous theoretical reports (at 155 and 200 GPa) [18–22].

It is observed in figure 1 that pressure induces an almost rigid displacement of the \( \alpha^2 F(\omega) \) spectrum and its maximum peak (\( \omega_m \)) towards higher energies. A considerable decrease in the area under the curve of this spectrum, mainly in the low-frequency region (10–90 meV), is also observed. For more details see reference [16].

For functional derivative, the increase in pressure induces a slight displacement of its maximum peak (\( \omega_m \)) towards lower energies. It has a sharper maximum for the sample with the lowest Tc (138 26 K to 215 GPa). This maximum gets broader as Tc goes up. In general, it is observed that the maximum is sharp for weak coupling and it gets broader as the coupling strengthens [11]. It is also possible to observe that Tc is located at \( \omega \sim 7k_B T_c \) (\( k_B \) is the Boltzmann constant), although a slight difference grows with increasing pressure.

The Tc, \( \delta \) M and \( \alpha M \) values obtained at different pressures are shown in table 1. Tc values were calculated in a previous work [16], which are in good agreement with the experimental values [14].

An important feature in figure 1 and table 1 is observed. When comparing the values of \( \delta \) M and \( \alpha M \) energies, their proximity reveals to be correlated with the value of the Tc (\( \omega \)). A small difference in \( \delta \) M – \( \alpha M \) implies the system being closer to the optimal frequency (\( \omega_{opt} \sim 7k_B T_c \)), this is to a higher critical temperature, and vice versa.
Figure 1. Pressure effects on the Eliashberg function $\alpha^2 F(\omega)$ and functional derivative $\delta T_c/\delta \alpha^2 F(\omega)$ for H$_3$S. The functional derivatives were scaled up to permitting comparison with the respective $\alpha^2 F(\omega)$ spectrum. Each figure is labeled with its corresponding pressure and $T_c$. The arrows indicate the evolution of maximum peaks for $\alpha^2 F(\omega)$ and $\delta T_c/\delta \alpha^2 F(\omega)$, $\alpha_M$ and $\delta_M$ respectively. The frequencies $\omega_{ln}$ and $\omega = 7k_BT_c$ are also calculated and indicated (dashed lines).

Table 1. The $T_c$, $\delta_M$ and $\alpha_M$ values obtained at different pressures. $\alpha_M$ and $\delta_M$ are the maximum peaks of $\alpha^2 F(\omega)$ and $\delta T_c/\delta \alpha^2 F(\omega)$ respectively. $T_c$-values taken from reference [16].

| P (GPa) | $T_c$ (K) | $\delta_M$ (meV) | $\alpha_M$ (meV) | $\delta_M - \alpha_M$ (meV) |
|---------|-----------|------------------|------------------|-----------------------------|
| 155     | 203 000   | 121.7            | 126.8            | 5.1                         |
| 160     | 197 400   | 113.9            | 128.7            | 14.8                        |
| 165     | 191 388   | 115.5            | 132.1            | 16.6                        |
| 170     | 186 263   | 112.8            | 132.0            | 19.2                        |
| 175     | 181 438   | 110.2            | 134.3            | 24.1                        |
| 180     | 169 300   | 103.6            | 139.5            | 35.9                        |
| 185     | 158 670   | 98.00            | 147.1            | 49.1                        |
| 195     | 147 305   | 91.80            | 152.7            | 60.9                        |
| 205     | 138 258   | 86.90            | 166.1            | 79.2                        |

Similar behavior can be observed in Nb$_3$Ge and Li (fcc) as reported by Baquero et al [1] and Yao et al [2], respectively. Figure 2(a) describes the evolution of the Nb–Ge system towards stochiometry and figure 2(b) shows the effects of pressure superconductor properties on Li. In both cases, it is observed that the closest proximity of $\delta_M$ and $\alpha_M$ occurs for the configuration in which the system has the highest $T_c$. However, for Li, the highest $T_c$ does not imply the largest broad of the maximum of $\delta T_c/\delta \alpha^2 F(\omega)$, as it occurs in H$_3$S and Nb$_3$Ge. It is also possible to observe that in both systems $\delta_M$ and $\alpha_M$ are close at $\omega = 7k_BT_c$ when the system has the highest $T_c$. According to our best knowledge, there are no other data available to extend the verification of this correlation.

These results are consistent with the idea of moving all the spectral weight in the $\alpha^2 F(\omega)$ to a delta function placed at $\omega \sim 7k_BT_c$, which would then allow obtaining the highest $T_c$ of that system [1, 13]. In this regard, Nicol and Carbotte [13] proposed that the Allen–Dynes characteristic phonon frequency $\omega_{ln}$ ($\omega_{ln} = \exp \{2/\lambda \} \int_0^\infty \ln(\omega)\alpha^2 F(\omega)/\omega d\omega$) [26] could be used as a close measure to this delta function, for H$_3$S. However, our results show $\omega_{ln}$ varies significantly with pressure ($\omega_{ln}$ increases with increasing pressure, see figure 1), which implies that possibly $\omega_{ln}$ is not a suitable reference parameter for such comparison. For H$_3$S, the variation of $\omega_{ln}$ versus pressure have been also reported by Akashi et al [27].

Based on our results, we conclude from the behavior seen in these systems that electron–phonon superconductors
Figure 2. Eliashberg spectral function $\alpha^2 F(\omega)$ and functional derivative $\delta T_c / \delta \alpha^2 F(\omega)$ for NbGe and Li (fcc) given in references [1, 2]. (a) NbGe system to different Ge concentration (in %). (b) Pressure effects on Li (fcc). Each picture is labeled with its corresponding experimental $T_c$ [23–25]. The arrows indicate the evolution of the difference $\delta M - \alpha M$. The frequency $\omega_{opt} = 7k_B T_c$ are also indicated (dashed lines).

will reach their highest possible $T_c$ ($\omega_{opt} = 7k_B T_c$) when the maximum peaks of the $\alpha^2 F(\omega)$ spectrum and the functional derivative of $T_c$ with respect to $\alpha^2 F(\omega)$ manage to get as close as possible.

This behavior could be considered as a theoretical tool that, in an electron–phonon superconductor, allows testing the degree of proximity to its highest $T_c$ possible for physical conditions defined. Furthermore, it could establish [through variations of spectral function $\alpha^2 F(\omega)$] the optimal physical conditions (pressure, alloying or doping concentration) that lead to the superconductor have the highest spectral density of the electron–phonon interaction $\alpha M$ at the optimal vibrational frequency $\omega_{opt}$, that is to say, its highest $T_c$.

4. Conclusions

In this work, we report a study of the functional derivative of $T_c$ with Eliashberg spectral function $\alpha^2 F(\omega)$ [$\delta T_c / \delta \alpha^2 F(\omega)$] and the $\alpha^2 F(\omega)$ spectrum for $H_3S$ (Im 3m) under pressure (155–215 GPa). The $\delta T_c / \delta \alpha^2 F(\omega)$ were calculated from the linearized version of the Migdal–Eliashberg gap equations. Our results were analyzed and compared with data available in the literature.

We found a correspondence between the maximum peaks of $\alpha^2 F(\omega)$ spectrum ($\alpha M$), $\delta T_c / \delta \alpha^2 F(\omega)$ ($\delta M$) and the highest $T_c$, which satisfies the idea of moving all the spectral weight in the $\alpha^2 F(\omega)$ to a delta function placed at $\omega_{opt} = 7k_B T_c$ (optimal frequency vibrational) where the system will be optimized. We observed that the convergence of $\delta M$ and $\alpha M$ (as $\alpha^2 F(\omega)$ varies) implies that the system is close to the higher $T_c$, and vice versa. This behavior was corroborated in other conventional superconductors by analyzing data available in the literature, suggesting its validity in electron–phonon superconductors.

Based on our results, we suggest that this correlation could be a theoretical tool that, in an electron–phonon superconductor, allows testing the degree of proximity to its highest $T_c$ possible for physical conditions defined, furthermore could establish [through variations of spectral function $\alpha^2 F(\omega)$] the optimal physical conditions (pressure, alloying or doping concentration) that lead to the superconductor reaching its highest $T_c$ possible.

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References

[1] Baquero R, Gutiérrez-Ibarra J, Meza L, Navarro O and Kihlstrom K E 1989 Rev. Mex. Fis. 35 461
[2] Yao Y, Tse J S, Tanaka K, Marsiglio F and Ma Y 2009 Phys. Rev. B 79 054524
[3] Bergmann G and Rainer D 1973 Z. Phys. 263 59
[4] Rainer D and Bergmann G 1974 J. Low Temp. Phys. 14 501
[5] Mitrović B 2002 Int. J. Mod. Phys. C 13 1087
[6] Mitrović B 2004 Eur. Phys. J. B 38 451
[7] Daams J M and Carbotte J P 1980 Solid State Commun. 33 585
[8] Mitrović B and Carbotte J P 1981 Solid State Commun. 37 1009
[9] Whitmore M D 1984 J. Low Temp. Phys. 56 129
[10] Marsiglio F and Carbotte J P 1985 Phys. Rev. B 31 4192
[11] Carbotte J P 1990 Rev. Mod. Phys. 62 1027
[12] Daams J M, Carbotte J P and Baquero R 1979 J. Low Temp. Phys. 35 547
[13] Nicol E J and Carbotte J P 2015 Phys. Rev. B 91 220507
[14] Drozdov A P, Erements M I, Troyan I A, Ksenofontov V and Shylin S I 2015 Nature 525 73
[15] Capitani F, Langerome B, Brubach J-B, Roy P, Drozdov A, Erements M I, Nicol E J, Carbotte J P and Timusk T 2017 Nat. Phys. 13 859
[16] Camargo-Martinez J A, Gonzalez-Pedreros G I and Baquero R 2019 Supercond. Sci. Technol. 32 125013
[17] Daams J M and Carbotte J P 1978 Can. J. Phys. 56 1248
[18] Errea I et al 2016 Nature 532 81
[19] Amsler M 2019 Phys. Rev. B 99 060102(R)
[20] Errea I et al 2015 Phys. Rev. Lett. 114 157004
[21] Flores-Livas J A, Sanna A and Gross E K U 2016 Eur. Phys. J 89 63
[22] Sanna A, JFlores-Livas J A, Davydov A, Profeta G, Dewhurst K, Sharma S and Gross E K U 2018 J. Phys. Soc. Japan 87 041012
[23] Kihlstrom K E and Geballe T H 1981 Phys. Rev. B 24 4101
[24] Kihlstrom K E, Mael D and Geballe T H 1984 Phys. Rev. B 29 150
[25] Deemyad S and Schilling J S 2003 Phys. Rev. Lett. 91 167001
[26] Allen P B and Dynes R C 1975 Phys. Rev. B 12 905
[27] Akashi R, Kawamura M, Tsuneyuki S, Nomura Y and Arita R 2015 Phys. Rev. B 91 224513