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High-Temperature Superconductor $PbH_4(H_2)_2$ under the Pressure of 200 GPa

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In the paper, we analyze the thermodynamic properties of high-temperature superconducting state induced in the $PbH_4(H_2)_2$ compound under the pressure of 200 GPa. We have shown that the critical temperature T_C varies from 81 K to 121 K, for the Coulomb pseudopotential $\mu^\star \in \langle 0.1, 0.3\rangle$. Due to the significant retardation and strongly coupling effects, the values of other thermodynamic parameters differ significantly from the predictions of mean-field BCS theory. The results have been obtained in the self-consistent manner under the Eliashberg formalism.

topics: Eliashberg, superconductivity, strong coupling constant, lead, BCS theory, thermodynamic properties, pressure effects

1. Introduction

In December 2014, after many years of research, there was the breakthrough in the superconducting physics. The high-temperature superconducting state has been found to induce in the H_2S and H_3S compounds under high pressure ($[T_C]_{H_2S} = 150$ K for $p \approx 200$ GPa, and $[T_C]_{H_3S} = 203$ K for $p = 155$ GPa) [1]. It should be noted that the relevant experiments were carried out in connection with the emergence of convincing theoretical works, which clearly pointed to the H_2S and H_3S compounds [2, 3]. Further intense theoretical research has strengthened the belief that the high temperature superconducting state in hydrogen-rich compounds is induced by the electron–phonon interaction [4–9].

In 2018, in the compound LaH_{10} the existence of the high-temperature superconducting state with the critical temperature comparable to the room temperature was experimentally detected: $T_C = 260$ K for $p \in \langle 180{-}200\rangle$ GPa [10]; $T_C = 215$ K for $p = 150$ GPa [11, 12]. Also in this case, the superconducting phase is induced by the electron–phonon interaction [13].

In the paper, we will examine the properties of the $PbH_4(H_2)_2$ compound under the pressure of 200 GPa. We will show that it can induce the superconducting state with the high critical temperature. Additionally, we will determine the basic thermodynamic parameters of this state. The results are based on the Eliashberg formalism [14], and the DFT calculations [15].

2. Eliashberg formalism

The thermodynamic parameters of the superconducting state were determined using the Eliashberg equations defined on the imaginary axis [16]. In the case of $PbH_4(H_2)_2$ compound, the infinitely wide electronic band can be assumed, as the consequence of which, we do not have to consider equations for the energy shift function and the chemical potential. The other two thermodynamic functions, i.e. the order parameter function ($\phi_n \equiv \phi(\mathrm{i}\omega_n)$), and the wave function renormalization factor ($Z_n \equiv \phi(\mathrm{i}\omega_n)$), we determined using the equations

$$\phi_n = \frac{\pi}{\beta}\sum_{m=-M}^{M}\frac{\lambda(\mathrm{i}\omega_n - \mathrm{i}\omega_m) - \mu^\star\theta(\omega_c - |\omega_m|)}{\sqrt{\omega_m^2 Z_m^2 + \phi_m^2}}\phi_m, \tag{1}$$

$$Z_n = 1 + \frac{1}{\omega_n}\frac{\pi}{\beta}\sum_{m=-M}^{M}\frac{\lambda(i\omega_n - i\omega_m)}{\sqrt{\omega_m^2 Z_m^2 + \phi_m^2}}\omega_m Z_m, \tag{2}$$

where $M = 1100$.

The Matsubara frequency is given by $\omega_n = (\pi/\beta)(2n-1)$, and $\beta = (k_\mathrm{B}T)^{-1}$; k_B is the Boltzmann constant. The pairing kernel of electron–phonon interaction is given by $\lambda(z) = 2\int_0^{\Omega_\mathrm{max}} \mathrm{d}\Omega \frac{\Omega}{\Omega^2 - z^2}\alpha^2F(\omega)$. The Eliashberg function ($\alpha^2F(\Omega)$) determining the form of electron–phonon interaction has been calculated using the density functional theory (DFT) method by Cheng et al. in the paper [15]. $\Omega_\mathrm{max} = 421.2$ meV means the value of maximum phonon frequency. Note that knowledge of the Eliashberg function allows

estimating the electron–phonon coupling constant $\lambda = 2\int_0^{\Omega_{\max}} \mathrm{d}\Omega \frac{\alpha^2 F(\Omega)}{\Omega} = 1.3$. The size θ in Eq. (1) means the Heaviside function; ω_c is the cut-off frequency with $\omega_c = 3\Omega_{\max}$. We took into account the values of the Coulomb pseudopotential ($\mu^\star$) in the range of 0.1 to 0.3 [14].

We solved Eliashberg equations numerically using methods that were developed in the paper [17]. Physically correct solutions can be obtained in the temperature range from $T_0 = 20$ K to T_C. Please note that there is the restriction on solutions from the low temperatures side. It results from the fact that for the temperature near absolute zero, we would have to take into account the infinite number of the Matsubara frequencies ($M = +\infty$).

3. Results

In Fig. 1, we plotted maximum value the order parameter as a function of temperature. It can be seen that the critical temperature is in the range of 81 K to 121 K. This means that the high temperature superconducting state can be induced in the $PbH_4(H_2)_2$ compound. Therefore, conducting appropriate experiments seems justified.

The full dependence of order parameter on the temperature can be reproduced using the formula $\Delta_{n=1}(T) = \Delta_{n=1}(T_0)\sqrt{1-\left(\frac{T}{T_C}\right)^{\Gamma}}$, where $\Gamma = 3.6$. Note that in the BCS theory, the exponent Γ is equal to 3 [18, 19].

By determining the values of order parameter and the wave function renormalizing factor using the Eliashberg equations, the remaining thermodynamic parameters of superconducting state can be calculated.

In the first step, let us estimate the half-width of energy gap $\Delta(T) = \mathrm{Re}[\Delta(\omega = \Delta(T))]$. On this basis, the value of dimensionless ratio can be determined: $R_\Delta = 2\Delta(0)/k_B T_C$, where $\Delta(0) = \Delta(T_0)$.

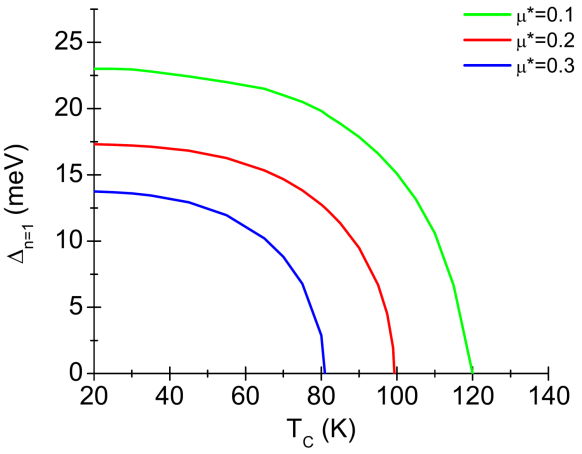

Fig. 1. The dependence of order parameter on the temperature for selected values of Coulomb pseudopotential.

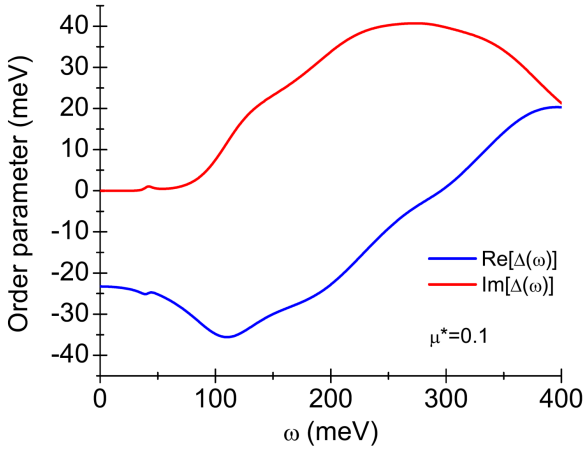

Fig. 2. The real part and the imaginary part of order parameter at the real axis. The case of $\mu^\star = 0.1$.

In addition, one can remember that the order parameter function specified on the imaginary axis should be analytically continued at the real axis. The procedure of analytical continuation has been explained in detail in the paper [20]. For example in Fig. 2, we have plotted the order parameter on the real axis for the case $\mu^\star = 0.1$ and $T = T_0$. It can be seen that the imaginary part of order parameter for the low frequencies is zero. Physically, this means no dissipative effects in the superconducting state. As the result of analysis, we obtained the following values of R_Δ: 4.22, 4.04, and 3.96, respectively for $\mu^\star = \{0.1, 0.2, 0.3\}$. Note that the BCS theory predicts $R_\Delta = 3.53$ [18, 19]. The observed discrepancies result from the existence of significant strong-coupling and retarding effects, as evidenced by the high value of electron–phonon coupling constant λ.

Next, we have calculated the difference in free energy between the superconducting and normal state

$$\Delta F/\rho(0) = -\frac{2\pi}{\beta}\sum_{n=1}^{M}\left(\sqrt{\omega_n^2+\Delta_n^2}-|\omega_n|\right) \times\left(Z_n^S - Z_n^N\frac{|\omega_n|}{\sqrt{\omega_n^2+\Delta_n^2}}\right), \quad (3)$$

wherein $\rho(0)$ is the value of electronic density of states at the Fermi surface. Z_n^S and Z_n^N are the wave function renormalization factors for the superconducting state (S) and the normal state (N), respectively.

The thermodynamic critical field is given by the formula $H_C/\sqrt{\rho(0)} = \sqrt{-8\pi[\Delta F/\rho(0)]}$.

In Fig. 3, we presented the dependence of free energy difference and the thermodynamic critical field on the temperature. The determined critical field can be estimated on the basis of experimental data, which allows to verify the theoretical results obtained by us.

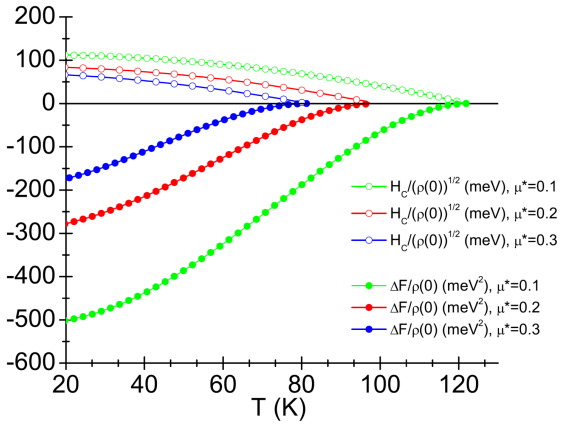

Fig. 3. The free energy difference and the thermodynamic critical field as a function of temperature.

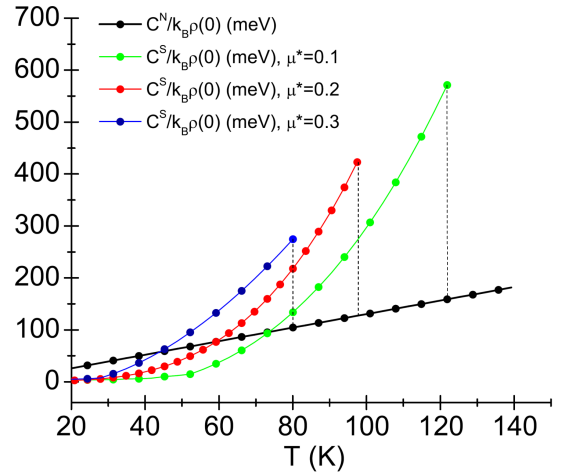

Fig. 4. The specific heat of superconducting and normal state as a function of temperature.

The difference of specific heat between superconducting and normal state ($\Delta C = C^S - C^N$) should be estimated based on the formula

$$\Delta C\left(T\right)/k_{\mathrm{B}}\rho\left(0\right) = -\frac{1}{\beta}\frac{\mathrm{d}^2\left[\Delta F/\rho\left(0\right)\right]}{\mathrm{d}\left(k_{\mathrm{B}}T\right)^2}. \qquad (4)$$

The specific heat of normal state is most convenient to estimate using $C^N\left(T\right)/k_{\mathrm{B}}\rho\left(0\right) = \frac{\gamma}{\beta}$. The Sommerfeld constant is given by the formula $\gamma = \frac{2}{3}\pi^2\left(1+\lambda\right)$.

The temperature dependence of specific heat of the superconducting and normal state was plotted in Fig. 4. Note the characteristic specific heat jump occurring at the critical temperature. This value can also be estimated on the basis of experimental data.

4. Summary and discussion of results

Obtaining the superconducting state in the room temperature is one of the main goals set by solid state physics. As we have shown in the present paper, the high-temperature superconducting state can induce in the $PbH_4(H_2)_2$ compound under the high pressure (200 GPa). Assuming reasonable values of the Coulomb pseudopotential ($\mu^\star \in \langle 0.1, 0.3\rangle$), we have shown that the critical temperature is in the range from 81 K to 121 K. Due to the high electron–phonon coupling constant ($\lambda = 1.3$), the values of other thermodynamic parameters differ significantly from the predictions of BCS theory. For example, $R_\Delta \in \langle 3.96, 4.22\rangle$.

It should be appreciated that after many years of research, the new family of chemical compounds was discovered, in which the superconducting state is induced with the critical temperature equal to or significantly exceeding the value of T_C observed for cuprates [21]. Nevertheless, the hydrogen-rich compounds are not ideal materials from the point of view of their possible technological applications. First of all, because they go into the superconducting state only in the presence of very high pressure. In our view, the importance of hydrogen-rich compounds is that the superconducting state can exist near the room temperature. In addition, we have fully decrypted the pairing mechanism leading to the induction of superconducting condensate [2, 3], which allows us to conduct advanced research at the theoretical level (and not only in the group of hydrogen-rich compounds [22]). Note that the pairing mechanism for cuprates is not fully understood to this day [23].

From the point of view of the presented results, the special attention should be paid to the huge difference between the atomic mass of lead and hydrogen. This fact is important from the point of view of the induction of the high-temperature superconducting state in $PbH_4(H_2)_2$. The lead contributes for electron–phonon coupling constant in the low frequency range, while hydrogen in the high frequency range. Therefore, these are complementary contributions. From the physical point of view, this means that it is worth investigating the superconducting properties of hydrogen-rich compounds consisting of atoms with significant atomic mass difference. For this reason, in the next step, we will consider compounds based on uranium.

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