Thermodynamic adsorption potential of superconductors

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Abstract

Based on the general thermodynamic analysis of Polanyi adsorption potential, the adsorption potential condition for superconductors is obtained exactly by using the quantum state equation we presented. Because this adsorption potential results in changes of electron concentration, temperature and pressure in a certain volume (adsorption space) adjacent to the surface of the lattice, the composition and structure of superconductors are of course decisive for the adsorption potential. Then we calculate the molar adsorption potentials for those typical superconductors, and find that it is positively correlated to the superconductivity temperature T_c, which reveals that those high-T_c superconductors are mainly determined by the higher molar adsorption potentials. In addition, the adsorption potential at 〖T=T〗_c still works despite the disappearance of the energy gap of the BCS theory. This shows that beyond the electron-phonon interaction mechanism, the Cooper-paired electrons are mainly formed by this physical adsorption potential for high-T_c superconductors. This adsorption potential theory could explain almost all common facts about high-temperature superconductors, including many anomalies of the normal and superconducting states.

Full Text

Preamble

Thermodynamic Adsorption Potential of Superconductors

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Abstract

Based on the general thermodynamic analysis of Polanyi adsorption potential, we obtain the exact adsorption potential condition for superconductors using the quantum state equation we previously presented. Because this adsorption potential induces changes in electron concentration, temperature, and pressure within a certain volume (adsorption space) adjacent to the lattice surface, the composition and structure of superconductors are naturally decisive for the adsorption potential. We calculate the molar adsorption potentials for typical superconductors and find that they are positively correlated with the superconducting transition temperature T_c, revealing that high-T_c superconductors are primarily determined by higher molar adsorption potentials. Additionally, the adsorption potential remains active at $T = T_c$ despite the disappearance of the energy gap predicted by BCS theory. This demonstrates that beyond the electron-phonon interaction mechanism, Cooper-paired electrons in high-T_c superconductors are mainly formed by this physical adsorption potential. This adsorption potential theory can explain nearly all common features of high-temperature superconductors, including many anomalies in both normal and superconducting states.

1. Introduction

Since Kamerlingh Onnes discovered superconductivity in 1911, the study of superconductivity has remained a prominent yet challenging topic in physics for over a century. Early research focused on metal and alloy systems with low superconducting transition temperatures, whose mechanism could be explained by the BCS theory established in 1957 and related subsequent developments. According to BCS theory, conventional superconductors cannot achieve T_c higher than 40 K at atmospheric pressure, so superconductors with $T_c > 40$ K are often referred to as high-temperature superconductors. In 1986, Müller and Bednorz discovered the $(\mathrm{La,Ba})_2\mathrm{CuO}_4$ superconductor, and in 1987, the discovery of $\mathrm{YBa_2Cu_3O_{7-\delta}}$ raised the superconducting temperature to the liquid nitrogen range for the first time, ushering in a new era of high-temperature superconductivity research. The highest T_c at atmospheric pressure currently stands at 133 K, achieved in 1993. Since the 1990s, high-precision epitaxial growth technology has been introduced into superconductivity research, enabling the formation of heterostructures through artificial stacking of materials with different structures or chemical compositions. Novel superconducting states may also emerge based on the coupling of these atomically smooth heterogeneous interfaces. Through artificial design of heterostructures or precise control of single-crystal film thickness, researchers can explore superconductivity under dimensional changes in highly crystalline samples. More recently, Yanagisawa investigated correlated-electron systems, emphasizing strong electron correlation as a driver of superconductivity in high-temperature cuprates.

To date, numerous experimental results reveal several common features of high-temperature superconductors: (1) They are strongly correlated electronic systems; (2) Some form of electron-electron mediation must be considered; (3) Due to severe anisotropy, interlayer coupling must be taken into account; (4) There should be an appropriate carrier concentration—too much or too little is detrimental to superconductivity; and (5) It is necessary to establish a theory of superconductivity based on the anomalous electronic states of the normal phase, because these anomalies contain the special interaction mechanisms of the electronic system. At present, the mechanism of high-temperature superconductivity remains unsolved, and the journal *Science* has repeatedly included it among the 125 most important scientific questions.

Due to strong electron interactions in high-temperature superconductors, describing their electronic behavior inevitably involves extremely complex many-body quantum physics. Consequently, the mechanism of high-temperature superconductivity is still not fully understood, particularly the source of the attractive interaction responsible for carrier pairing, which remains one of the core challenges in current physics.

Grounded in the work of Henri Poincaré, R. Thom, and others, catastrophe theory can explain phenomena involving gradual quantitative change leading to sudden qualitative change. This highly generalized mathematical theory summarizes the rules of non-equilibrium phase transitions through several catastrophe models. According to Thom' s classification theorem, as long as the number of control variables causing mutations does not exceed four, various natural mutation processes can be described using seven basic potential function models. Due to its inherent structural stability during the mutation process of any non-equilibrium system, any phase transition can be analyzed quantitatively using one of these catastrophe models, even without knowing the system' s differential equations. In our previous work, we used catastrophe theory to quantitatively investigate the general non-equilibrium phase transition process from laminar to turbulent flow, as well as to derive a revised relativistic Schrödinger equation from a phase transition perspective.

More recently, we quantitatively investigated the thermodynamic quantum phase transition process using structural-stability-based catastrophe theory. For a canonical ensemble composed of N identical particles, we adopted the cusp catastrophe model to express the average free energy and further obtained a general quantum state equation for pressure to describe the phase transition process from quantum to macro scales using dimensional analysis. Subsequently, we derived the ensemble free energy while considering the interaction potential energy among particles, along with the canonical partition function and the specific heat capacity of the system.

The transition from a normal state to a superconducting state is a phase transition problem that clearly falls within the thermodynamic framework. Superconductivity represents a second-order phase transition, meaning the superconducting phase transition occurs without latent heat changes, while the free energy of the superconducting state must be lower than that of the normal state. By analogy with Polanyi adsorption potential theory in chemistry, we propose a physical adsorption potential between free electrons and the superconductor lattice through van der Waals forces. We assume these electrons can be adsorbed and condensed due to changes in electron concentration, temperature, and pressure in a certain volume (adsorption space) adjacent to the lattice surface, in addition to the electron-phonon interaction mechanism.

In this paper, based on this general quantum state equation derived from catastrophe theory, we further develop the physical adsorption potential between electrons and the superconductor lattice to attempt an explanation for high-temperature superconductivity.

2. General Thermodynamic Analysis of Polanyi Adsorption Potential

The adsorption of gas or liquid on a solid surface results from adsorption displacement caused by adsorption forces between the adsorbate in the adsorption force field and the solid surface. The field strength of an equipotential surface in the adsorption force field depends not only on the properties of the adsorbate and solid and its surface structure but also on the distance between the equipotential surface and the solid surface. The field strength decreases with increasing distance and becomes zero at infinity, which is defined as the zero-potential surface.

In the adsorption force field, the adsorbate moves along the field strength direction under the adsorption force to perform adsorption work. Here, the effect of the adsorption force exceeds that of molecular thermal motion, resulting in reduced distances among adsorbate molecules and even producing coagulation or chemical adsorption. The adsorption layer between the normal phase of the adsorbate and the solid surface is called the adsorption phase, where the adsorbate density is greater than that of the normal phase, and the concentration of the adsorption phase shows a gradient and continuous change, as shown in [Figure 1: see original paper].

[Figure 1: see original paper] Diagram of the physical adsorption potential adjacent to the surface of the lattice

From the above, we see that the adsorption process involves adsorbate molecules entering the adsorption phase from the normal phase under certain conditions, undergoing displacement or chemical changes—in other words, changes in the amount of substance. Below, we use chemical potential and adsorption work to reflect the rules of the adsorption process.

For a system consisting of adsorbate components, the adsorption force is a generalized force, and the Gibbs free energy of the adsorption system is expressed as $G = G(T, P, L, N_1, N_2, \cdots, N_k)$, whose full differential is

$$dG = \left(\frac{\partial G}{\partial T}\right)_{P,L,N_j} dT+\left(\frac{\partial G}{\partial P}\right)_{T,L,N_j} dP+\left(\frac{\partial G}{\partial L}\right)_{P,T,N_j} dL+\sum_i \left(\frac{\partial G}{\partial N_i}\right)_{P,T,L,N_{j\neq i}} dN_i$$

where P is pressure, T is temperature, L is adsorption displacement, N_j is the number of particles of the j-th component in the adsorbate, and the subscript $N_{j\neq i}$ indicates that the amount of other components except the i-th component remains constant.

Because the adsorption work done by the system in a reversible process equals the decrease in free energy, we have

$$\left(\frac{\partial G}{\partial L}\right)_{P,T,N_j} dL = f dL = -\delta W$$

where $f = \left(\frac{\partial G}{\partial L}\right)_{P,T,N_j}$ is the generalized adsorption force, and $\left(\frac{\partial G}{\partial L}\right)_{P,T,N_j} dL$ represents the contribution of the adsorption system at a certain equipotential surface to the free energy change with respect to adsorption displacement L.

Additionally, the adsorption process mostly occurs at constant temperature and pressure, i.e., $dT = dP = 0$, so Eq. (2) simplifies to

$$dG = -\delta W + \sum_i \left(\frac{\partial G}{\partial N_i}\right)_{P,T,L,N_{j\neq i}} dN_i$$

Since this adsorption process is spontaneous, according to the principle of free enthalpy reduction, i.e., $dG \leq 0$, we have

$$\delta W = \sum_i \mu_i dN_i$$

where the chemical potential $\mu_i = \left(\frac{\partial G}{\partial N_i}\right)_{P,T,L,N_{j\neq i}}$. Eq. (5) represents the contribution of a multi-component system to the adsorption work when the amount of substance changes under constant temperature and pressure.

When the amount dN_i^b of the i-th component at the zero point of the bulk phase in the system is adsorbed to an equipotential surface of the adsorption phase, the amount added to the i-th component at the equipotential surface is dN_i^a, and $-dN_i^b = dN_i^a = dN_i$. The adsorption potential of an adsorbate molecule in the adsorption force field on the solid surface is defined as the work required to move the molecule from its equipotential position in the adsorption phase to the zero position. Thus, Eq. (5) becomes

$$\varepsilon_i^a dN_i \leq \delta W = \mu_i^b dN_i - \mu_i^a dN_i$$

When $dN_i = 1$ mol, the work done by the system to adsorb 1 mol of substance equals the Polanyi molar adsorption potential ε, giving

$$\varepsilon = A(\mu_i^b - \mu_i^a)$$

where A is Avogadro' s number, indicating that ε represents the change in chemical potential when 1 mol of substance is reversibly adsorbed from the bulk phase b to the adsorption phase a.

3. Adsorption Condition at the Solid-Gas Interface of Superconductors

For superconductors, this adsorption potential is also caused by enhanced condensation, which should depend on the properties of the superconductor's lattice structure and the position of electrons near the lattice surface. Since the electron concentration in normal states is uniform, the adsorption potential is zero at the interface between normal states and the adsorption phase. The adsorption potential of free electrons in equilibrium with the solid surface can take any equilibrium value from zero to saturation.

3.1 Thermodynamic Adsorption Condition for Superconductors

According to Ref. [21], we have obtained the correlation among electron concentration, temperature, and pressure of Fermi electrons through catastrophe theory:

$$Pv^{5/3} = \left(\frac{4\alpha}{3}\right)^{3/2} \frac{\hbar^2 n^{2/3}}{m} \left(\sqrt{B + \sqrt{B^2 - \frac{9}{\beta^2}}} + \sqrt{B - \sqrt{B^2 - \frac{9}{\beta^2}}} \right)$$

where $\hbar$ is Planck' s constant, m is the rest mass of electron, k_B is Boltzmann constant, $n = N/V$ is the number of electrons per unit volume, α is the phase transition index, $\beta = \frac{\hbar^2 n^{2/3}}{mk_{BT}}$ is a dimensionless temperature, and $B = \left[(3\pi^2)^{2/3} + \frac{3}{\beta}\right]/2$.

From Eq. (7), when $\beta \gg 1$, meaning the mean distance among particles $v^{1/3}$ is much larger than the thermal wavelength $\lambda = \sqrt{2\pi\hbar^2/(mk_{BT})}$, the microscopic quantum effect can be ignored. In the infinite limit of $\beta \to \infty$ and $\alpha = 1/2$, Eq. (7) degenerates to the state equation of ideal Boltzmann gas with $P = nk_{BT}$ [21]. When $\alpha = 0$, $\beta \to 0$, i.e., $\beta\alpha = 1$, the pressure of the Fermi gas is not zero due to the Pauli exclusion principle, giving $P = \frac{\hbar^2 n^{5/3}}{m}(3\pi^2)^{2/3}$ [21].

For high-temperature superconductors such as perovskite copper oxides ($Sr_8CaRe_3Cu_4O_{24}$), there is high superconducting temperature (up to $T_c = 135$ K) with low carrier concentration ($n = 3.0 \times 10^{27}$). For low-temperature superconductors such as Ag, there is low superconducting temperature (0.9 K) with high carrier concentration ($n = 1.044 \times 10^{31}$). In any case, $\beta = \frac{\hbar^2 n^{2/3}}{mk_{BT}} \ll 1$ for all superconductors. Therefore, Eq. (7) simplifies to

$$P = 1.74nk_{BT}\left(\frac{4\alpha}{3} - 1\right)$$

i.e.,

$$1.74\left(\frac{4\alpha}{3} - 1\right) = \frac{P}{nk_{BT}}$$

Eq. (8) is a thermodynamic quantum equation of state for Fermi gases, indicating that under the same pressure, when electron concentration n increases, temperature T decreases.

Using Eq. (8a), the Gibbs free energy of N identical electrons is [21]

$$G(P,V,T) = F(P,V,T) + PV = Nk_{BT}\left[2.638\left(\frac{4\alpha}{3} - 1\right) + 12.88\left(\frac{4\alpha}{3} - 1\right)^2\right]$$

where $F(P,V,T)$ is the free energy of the system.

From Eq. (9), the chemical potential at the normal phase is

$$\mu_n = 4.397k_{BT}\left[(1 - \frac{4\alpha}{3}) + 4.88(1 - \frac{4\alpha}{3})^2\right]$$

which shows that the chemical potential depends on temperature T, electron concentration n, and phase transition index α.

At the adsorption phase, due to work done by the adsorption force, electron concentration increases, resulting in a temperature decrease according to Eq. (8a). As T decreases to the superconducting temperature T_c, some additional form of electron order begins to form and increases as the temperature drops further. At this point, the phase transition index becomes α', and the superconducting electron concentration becomes $n_s = \omega n$ ($\omega \leq 1$), where ω is the relative proportion.

Since the special properties of the superconducting state are determined by the condensed matter of Cooper pairs, and the formation of Cooper pairs allows electrons to flow through the material without resistance, this new electron pairing state plays a dominant role in the energy and particle number changes of the system. Therefore, in the superconducting state, the chemical potential is primarily considered for superconducting electrons (Cooper pairs).

The chemical potential μ_s of the superconducting phase is

$$\mu_s = 4.397 k_{BT} \left[(1-\alpha') \left(\frac{4\alpha'}{3} - 1 \right) + 4.88(1-\alpha') \left(\frac{4\alpha'}{3} - 1 \right)^2 \right]$$

where N_S is the number of superconducting electrons.

Therefore, according to Eq. (6b), the adsorption potential condition for the superconducting phase transition is

$$\varepsilon_p \geq \mu_s - \mu_n$$

where ε_p is the average adsorption energy per electron.

3.2 Identification of the Phase Transition Index α for Superconductors

When a normal metal is cooled, conductive electron entropy typically decreases. At temperatures below T_c, some additional form of electronic ordering must begin to form, making an additional contribution to the specific heat capacity.

According to Eqs. (8) and (9), the conductive electron entropy S and specific heat capacity C_p can be obtained respectively as

$$S = \frac{15 k_B \alpha}{(8\alpha - 15)} \left[44.8494 \left(\frac{4\alpha}{3} - 1 \right) \left(\frac{5\alpha}{3} - 1 \right) + 154.56 \left(\frac{4\alpha}{3} - 1 \right)^2 \right] \frac{\hbar^2 n^{2/3}}{m k_{BT}}$$

$$C_p = T \left(\frac{\partial S}{\partial T} \right)_P = \frac{15 k_B \alpha}{(8\alpha - 15)^2} \left[44.8494 \left(\frac{5\alpha}{3} - 1 \right) \left(\frac{4\alpha}{3} - 1 \right) + 154.56 \left(\frac{4\alpha}{3} - 1 \right)^2 \right] \frac{\hbar^2 n^{2/3}}{m k_{BT}}$$

From an entropy perspective, the formation of Cooper pairs and their motion states affect entropy calculations. Therefore, when calculating the entropy of a superconducting state, we primarily consider the influence of superconducting electrons, i.e., the electron contribution n in Eq. (13) is replaced by the superconducting electron concentration $n_s = \omega n$.

It is well known that $C_p \sim T^3$ in the superconducting phase and $C_p \sim T$ in the normal phase. Considering Eq. (8b) where $n \sim T^{8\alpha-15}$ and $\beta = \frac{\hbar^2 n^{2/3}}{m k_{BT}} < 1$, we have $\alpha = \frac{16}{15}$ for $T > T_c$, which results in a jump in specific heat capacity C_p at T_c. Meanwhile, since the superconducting phase transition occurs without latent heat changes, the continuity of entropy S at T_c uniquely determines the change in electron concentration from the normal phase to the superconducting phase.

In fact, when a superconducting phase transition occurs in a material, electrons form Cooper pairs, their motion states become bound, and the interaction mode between electrons changes, altering the effective interaction potential energy among electrons and consequently changing the pressure of the electron gas. Thus, for the superconducting phase, according to Eq. (8b) we have

$$P_s = 1.74 n_{sk_{BT}} c \left(\frac{4\alpha'}{3} - 1\right) = 1.74 \omega n k_{BT} c \left(\frac{4\alpha'}{3} - 1\right)$$

where P_s is the pressure of the electron gas in the superconducting phase.

The following example of metal Sn illustrates its superconducting phase transformation process. For Sn, $T_c = 3.72$ K, at which $n = 8.8 \times 10^{28}$. According to the continuity of entropy S at T_c, from Eq. (13) we obtain $\omega = 0.0006223$, meaning that the superconductivity phenomenon occurs only when the superconducting electron concentration increases to 0.6223% under the action of adsorption potential.

[Figure 2: see original paper] The entropy and the specific heat capacity of Sn varying with temperature

Figure 2 shows the conductive electron entropy S and specific heat capacity C_p of 1 molar electrons of Sn varying with temperature T, calculated using Eqs. (13) and (14).

3.3 The Adsorption Potential Condition for Superconductors

Based on the above analysis, the adsorption potential condition for the superconducting phase transition can be further obtained as

$$\varepsilon_p \geq \mu_s - \mu_n = k_{BT} c \left[1.256 \left(\frac{4\alpha'}{3} - 1\right) \frac{\hbar^2 (\omega n_c)^{2/3}}{m k_{BT} c} - 9.194 \left(\frac{4\alpha'}{3} - 1\right)^2 \left(\frac{\hbar^2 (\omega n_c)^{2/3}}{m k_{BT} c}\right)^2 - 1.256 \left(\frac{4\alpha}{3} - 1\right) \frac{\hbar^2 n}{mk} \right.$$

where ω is determined by the continuity of entropy S at T_c from Eq. (13).

According to Eq. (16a), when $T \to 0$, $\omega \to 1$, and $\varepsilon_p \to 0$, which differs from BCS theory where the band gap $2\Delta(0) = 3.53 k_{BT} c$. On the other hand, when $T = T_c$, we have

$$\varepsilon_p \geq \mu_s - \mu_n = k_{BT} c \left[1.256 \left(\frac{4\alpha'}{3} - 1\right) \frac{\hbar^2 (\omega n_c)^{2/3}}{m k_{BT} c} - 9.194 \left(\frac{4\alpha'}{3} - 1\right)^2 \left(\frac{\hbar^2 (\omega n_c)^{2/3}}{m k_{BT} c}\right)^2 - 1.256 \left(\frac{4\alpha}{3} - 1\right) \frac{\hbar^2 n}{mk} \right.$$

which also differs from BCS theory where the band gap $\Delta(T_c) = 0$. Here n_c is the electron concentration at T_c, and $n_s = \omega n_c$.

Table 1 The average adsorption energy per electron ε_p for superconductors

Superconductors	T_c (K)	n_c at T_c	ω	ε_p	ε
$La_{2-x}Sr_xCuO_4$	35	3.26×10^{30}	2.898×10^{-6}	$0.21k_{BT}c$	$0.41k_{BT}c$
$YBa_2Cu_3O_{6.7}$	60	8.8×10^{28}	1.5×10^{-7}	$0.44k_{BT}c$	$0.32k_{BT}c$
$YBa_2Cu_3O_7$	90	5.0×10^{27}	1.5×10^{-7}	$0.2665k_{BT}c$	$0.27485k_{BT}c$
$Sr_8CaRe_3Cu_4O_{24}$	135	3.0×10^{27}	1.5×10^{-7}	$54.44k_{BT}c$	-

According to Eq. (16b), Table 1 calculates the average adsorption energy per electron ε_p and the corresponding molar adsorption potential ε for various superconductors. We find that the molar adsorption potential ε is positively correlated with the superconducting temperature T_c, revealing that high-T_c superconductors ($T_c \geq 40$ K) are primarily formed by higher molar adsorption potentials. At $T = T_c$, the adsorption potential remains active despite the disappearance of the energy gap in BCS theory. Regarding ω, during electron migration due to adsorption, larger ω corresponds to greater adsorption energy. This positive correlation means that ω actually reflects the composition and structure of superconductors, which are decisive for the adsorption potentials, even at the same electron concentration. Furthermore, this can explain why for copper oxides with high-temperature superconductivity, the smallest unit cell requires at least an intact layer containing the CuO_2 bilayer.

For low-T_c superconductors with high carrier concentration in the normal phase, their simple composition and structure result in adsorption potentials too low to form additional Cooper pairs beyond the electron-phonon interaction mechanism. Conversely, for high-T_c superconductors with low carrier concentration in the normal phase, when the degree of electron migration ω increases to a certain extent, the molar adsorption energy increases, resulting in the high-T_c phase transition. From this perspective, anomalies in superconducting properties can be explained for high-temperature superconductors, while anomalies in normal states can be explained for low-temperature superconductors.

4. Conclusions

In this paper, we precisely define the molar adsorption potential and propose the thermodynamic adsorption potential condition to explain high-temperature superconductivity. We reveal that beyond the electron-phonon interaction mechanism, Cooper-paired electrons in high-T_c superconductors are primarily formed by this physical adsorption potential. Thus, high-temperature superconductivity can still be explained by Cooper-paired electrons arising from the molar adsorption potential. This theory can explain many anomalies in both the normal and superconducting states of perovskite copper oxides, as well as the isotopic effects in copper oxides.

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Note: Figure translations are in progress. See original paper for figures.