A general model for superconductivity

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Abstract

The discovery of superconductivity is more than a hundred years with a variety of superconductors reported so far. The conventional BCS theory works for the superconductive metals, but it fails to oxide superconductors. The absence of general theory greatly restricts the deep understanding and future study for superconductivity. Here, we successfully propose a universal theory for superconductivity through modeling the microscopically atomic interactions. The general formula for critical temperature is developed to display a strong relationship to a few parameters of materials. Specifically, the critical temperature displays a power law correlation for carrier concentrations and is inversely proportional to thermal expansion coefficient. Then, this model is well confirmed and supported by the reported experiment data of superconductive metals and oxides, showing excellent universality. This work fills the century gap of superconductivity and thus provides a deep insight and powerful guidance for future study.

Full Text

Preamble

A general model for superconductivity Zi-an Liu1,2,† , Zhiqiang Gao1,† , Xun Shi1,2,∗

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The discovery of superconductivity is more than a hundred years with a variety of superconductors reported so far. The conventional BCS theory works for the superconductive metals, but it fails to oxide superconductors. The absence of general theory greatly restricts the deep understanding and future study for

superconductivity. Here, we successfully propose a universal theory for superconductivity through modeling the microscopically atomic interactions. The general formula for critical temperature is developed to display a strong relationship to a few parameters of materials.

Specifically, the critical temperature displays a power law correlation for carrier concentrations and is inversely proportional to thermal expansion coefficient. Then, this model is well confirmed and supported by the reported experiment data of superconductive metals and oxides, showing excellent universality. This work fills the century gap of superconductivity and thus provides a deep insight and powerful guidance for future study.

A material with electrical superconductivity is called a superconductor, in which the electrical resistance vanishes and magnetic flux fields are expelled from the material when it is cooled to its critical temperature TC 1,2 .

Now, it became the central topic in condensed matter physics and materials science for fundamental research. Furthermore, it has extremely important practical applications in industry and daily life such as highfield magnets3 , medical imaging4 , power transmission5 , quantum devices6 . A higher TC is especially required because it can reduce cooling requirements, lower operating costs, and make superconducting technologies easier to use in large-scale applications7,8 . The phenomenon of superconductivity was discovered in solid mercury in 19119 . Later, some metals10 and their alloys11 are found to display superconductivity at very low temperatures, i.e., usually around several Kelvin. Until 1986, the ceramic materials are found to have a high TC above 90 K, leading to the discovery of high-temperature superconductors12 . Today, a variety of materials like metals10,13 , cuprates12,14 , and iron-based8,11 superconductors have been discovered to greatly expand the family of superconducting materials with the TC ranging from extremely low temperature to about 150 K under normal pressure15 .

The physical origin for superconductivity is extremely complex and it is still not clarified yet. The BardeenCooper-Schrieffer16 (BCS) theory, proposed in 1957, explains the key aspects of microscopic origin of superconductivity in low TC metals, in which the superfluid of Cooper electron pairs that interact through the exchange of phonons, contribute to superconductivity. However, the BCS theory cannot explain the high-temperature superconductors, in which the high TC is theoretically im-

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possible for a conventional superconductor17 . It remains a key scientific question and challenge until today. The absences of universal theory for superconductivity not only restricts the physical understating of superconductivity, but also greatly holds back the design and discovery of new superconductors with high TC 17 .

In this work, we propose a general theory for superconductivity through modeling microscopically atomic interactions. It well covers the low TC metals and high TC superconductors, which not only explains many physical phenomenon and requirements for superconductivity, but also provides a powerful guidance for future study.

In real space, a superconductor should possess the long-range connected superconducting channel throughout the material. Otherwise, it is not superconductive.

For an electrical conductor, when the carrier concentration (n) is small, the movement and transfer of carriers (electrons or holes) can be scattered by various particles and defects, like acoustic and optical phonons, ionized impurities, and a large variety of defects. In this case, the superconducting channel is easily destroyed. Therefore, in order to obtain superconducting channel, the n should be large enough to screen these carrier scattering, i.e., a threshold value (nt) for carrier concentration is required. Only when n $\geq$ nt , the material is superconductive. A degenerated system is thus required to provide an enough large n. This is the reason why the superconductivity is found in metallic state.

If the material has superconductivity, the electrons can freely pass through the superconducting channels without any barriers. In materials, the shortest width of superconducting channels is determined by the distance between the nearest adjacent atoms. Taking metals as the first example, in the classic diagram, the atoms lose their outer electrons to form ionized cations with positive charges (see Fig. 1a [Figure 1: see original paper]). The donated electrons from the atoms have common movement within the lattice, which is called free electron gas. Usually, metals have

expansion coefficient (α), which is defined as the change of the volume in response to a change in temperature.

Therefore, the relationship between W and TC can be given by

α $\cdot$ TC =

Substituting Eq. 2-4 into Eq. 1, we can get

2$ $0 −1/6 d0 − 2r+

Eq. 5 can be rewritten as TC = TU − Bn−1/6

interactions and the general model for superconductivity in (a) metals and (b) oxides. a very high n in the range of 1028 − 1029 m−3 , a typical degenerated system that can easily screen those neutral impurities and defects. At 0 K, the phonons are frozen; thus, the scattering from phonons vanishes. For a degenerate electron gas, there exists a screening length (λ) for ionized impurities. When the distance is larger than λ, the ionized impurities have no effect on carrier transfer.

Therefore, for metals, the shortest width (W) of superconducting channel is described by W = d0 − 2r+ − 2λ

where

λ = $ $0 · n−1/6

where $ $0 is the screening coefficient constant $ $0 2 π 4/3 m0 e2 31/3

$ $0 is the vacuum permittivity, m0 is the rest mass of electron, and e is the elementary charge. Clearly, a (−1/6) power-law relationship is observed between λ and n.

At temperature T , all atoms are thermally vibrated, which can strongly scatter transferred carriers. The harmonic vibration is believed to has no effect on the superconducting channel because all the atoms vibrate synchronously. However, the channel is significantly affected by the anharmonic vibration, i.e., the adjacent atoms may simultaneously move towards each other to close the channel. In this case, the material will lose its superconductivity. The anharmonic vibration of all atoms in materials can be described by the parameter of thermal

d0 − 2r+

TU can be regarded as the theoretical upper limit of the superconducting critical temperature when n is infinity.

Then we take high-temperature oxide superconductor as the second example. As shown in Fig. 1b, different from the metals, there are cations and anions bonded in oxides. Therefore, the TU is

where d0 is the distance between the nearest adjacent atomic centers and r+ is the radius of cations.

The λ in three-dimensional free electron gas can be obtained by the Thomas-Fermi screening theory (see Ref. 18 and the Supplementary Note 1)

d0 − r+ − r−

where r− is the radius of anion. Another significant difference between oxides and metals is the occupied volume for electron gas. In metals, the electron gas can fill all the free three-dimensional space, i.e., metals have a large occupied volume. However, for oxides, the distribution of electron gas is greatly restricted in a very small and narrow space. Taking famous yttrium barium copper oxide (YBCO) as the example, it is typical layered material and only the CuO2 planes are superconductive (see Fig. 2a [Figure 2: see original paper]). The effective volume for electron gas is thus greatly limited by the orientational Cu-O bonds. First, the superconductivity in YBCO is strictly confined within the two-dimensional CuO2 planes (Fig. 2a), while the other planes are not superconductive. This suggests that the electron gas is also confined within these two-dimensional planes. Second, even within the CuO2 plane, the electron gas is also constrained into long and thin "pipes" along the Cu-O bonds (see Fig.

2b). Third, for each Cu and O atom, the distribution of electron gas is restricted to a narrow solid angular range too (see Fig. 2c), instead of the fully symmetric sphere (4π) in metals. In total, all these factors work together, leading to an extremely small effective volume for electron gas in YBCO as compared with metals. A volume factor δ is thus used to describe the

and Y-doped BSCCO.The dashed lines in d are the fitted curves by using Eq. 6.

ratio of effective volume to total free space, which can be roughly estimated based on material' s crystal structure and bonding features. In YBCO, the δ is in the range of $10{-}3 - 10{-}4$. Such small δ in YBCO indicates that only extremely small volume in the lattice is needed to be screened for superconductivity. Therefore, the screening effect in YBCO is extremely strong, which is proportional to $1/\delta$. This leads to a greatly reduced screening length $\lambda =$ \$ \$0 δ • n−1/6 . Then the B is

For other materials, parameter TU and B may vary according to material' s structures and bonding details.

But Eq. 6 should be the general and universal formula for superconductivity. Next, we check the validity of our model by using the reported experiment data. In the crystalline materials, the radius of cations and anions are determined by their valence state and coordination numbers (CN). When the valence state is not an integer, we chose the closest integer as the CN. We carefully searched these data and listed them in Table I and II. In principle, the data for α between 0 K and TC , and the data for other parameters at TC should be used. However, many of data are absent in experiment. Thus, the roomtemperature values for d0 , r+ , r− and n are used. This is acceptable since they usually change very little from 0 to 300 K for metallic state. In addition, the (-1/6) power law for n in Eq. 2 also indicates it is insensitive when n is not changed too much.

YBCO is taken as the first example because of the rich experiment data in literature. It has orthorhombic structure, which is anisotropic and the α varies along different directions. For simplifying, an average value of $0.92 \times 10{-}6$ K−1 for α at about 50 K is used. In YBCO, a large range of hole concentration (still marked as n) is realized through adjusting the partial pressure of oxygen.

In this case, the d0 , r+ , and r− change little when just changing the content oxygen in a certain range. Therefore, the TC is dominated by the variation of n as shown

in Fig. 2d. Clearly, the nt in YBCO is around 1027 m−3 , a typical degenerated range. We use Eq. 6 to fit the experiment data and the fitted curve agree with the experiment as shown in Fig. 2d, in which the experiment data follow well along the (-1/6) power law. The fitted B is 5.651×106 K m−1/2 and the fitted TU is 212.46 K, indicating the upper limit of TC in this material. Furthermore, we also checked the model by another superconductor, Bismuth Strontium Calcium Copper Oxide (BSCCO)25 . The data from the literature are also listed in Table I. Similarly, the fitted curve by Eq. 6 is consistent with the experiment data (Fig. 2d) with the fitted parameters listed in Table I too. All these data strongly suggest that our model works for high-temperature superconductors.

Then the metals are used as the second exzample to check our model. Different from YBCO, there are many different types of metals to show superconductivity with different n values for each metal. Furthermore, the d0 , r+ , and α are also different each other. Therefore, it is hard to obtain the similar diagram as shown in Fig. 2d.

We carefully searched the literature and then list the data for above parameters in Table II. In order to compare the theoretical calculation with experiment data, we plot (TU − TC) vs (n−1/6 /(αd0)) in Fig. 3a [Figure 3: see original paper]. The experiment data are almost fully located in the dashed line with the slope of 2$ $0 , which is obtained by Eq. 5. This strongly suggests that the model also works for superconductive metals.

Finally, we try to combine superconductive metals and high-temperature superconductors into one diagram. the power law of (-1/6). The YBCO and BSCCO materials are located in the low n range, and the metals are located in the high n range. Despite the vast differences in their crystalline structures and electronic properties, all these three types of materials follow well the (-1/6) power law, suggesting the excellent universality of our model.

Table I and II show that the shortest width of superconducting channels is in the range of 10−14 m for ox-

metals is calculated by Eq. 7. TABLE I: Physical parameters for orthorhombic YBCO and tetragonal Y-doped BSCCO.The valence states of Cu and O are +2 and -2, respectively. The in-plane CN is 4 for Cu and 2 for O. Sample No. of YBCO is taken from ref. 19. The data for d0 of YBCO at 300 K are taken from ref. 20, and the data for d0 of BSCCO are taken from ref. 21. The data for r+ , and r− at 300 K are taken from ref. 22. The data for n and TC are taken from ref. 20. A constant α value in the ab plane at 50 K is used. For YBCO, the data for α are taken from ref. 23. For BSCCO, the data for α are taken from ref. 24. TU , B and λ are obtained by fitting the experiment data in Fig. 2d by Eq. 6, 9-10. W is calculated by Eq. 4.

Materials

BSCCO

Sample

m) (10

n) (10

o) (10 m

) (K) (10

(10 K m

) (10

m) (10

ides and 10−15 − 10−19 m for metals, which are extremely small. This is the reason why the TC is low in superconductors. Detailed analysis shows that the large width of superconducting channels in oxides is mainly from the ex-

tremely strong spatial confinement screening effect. Theoretically, if we have enough accurate values for all the parameters, we can directly calculate TC by Eq. 6-10.

However, current measurement accuracy of the distance

TABLE II: Physical parameters for superconductive metals.Valence state Z is calculated by Z = ρN , where M is the Molar mass, ρ represents density, and NA is the Avogadro constant. Considering the anisotropy of HCP structures, the in-plane CN of 6 is taken. The data for r+ at 300 K are taken from ref. 22. For r+ , the value corresponding to the Z is used or is extrapolated/ interpolated according to the values in ref. 22. The data marked with for d0 at 300 K are taken from ref. 26, and the others are from ref. 27. The data marked with ∗ for n are from ref. 28, and the others are from ref. 29. The data of TC are taken from ref. 30. The data for α of Al, In, Nb are taken from ref. 31, ref. 32 and ref. 33, respectively, and the others are taken from ref. 34. The temperature for α is listed in the bracket. TU is calculated by Eq. 7 and B is calculated by Eq. 8. λ is calculated by Eq. 2 and W is calculated by Eq. 4.

Materials

Crystal structure

+3.004 (Z = 3) +2.244 (Z = 2) +2.018 (Z = 2) +2.999 (Z = 3) +1.249 (Z = 1) +1.430 (Z = 1)

(10−10 m) (10−10 m) (1027 m−3) (K) (10−6 K) (106 K) (1010 K m−1/2) (10−11 m) (10−18 m)

3.01∗

82.0∗

is usually in the order of 10−12 m, which is far larger than the width of superconducting channel. Therefore, it is strongly recommended to improve the measurement technique to obtain high accurate data for future study.

Adjusting pressure (P) is a feasible approach for high TC materials. The pressure definitely can change all the parameters shown above. For simplifying, the effect of pressure on d0 , r+ , and r− can be ignored because they offset each other. The n can be increased when increasing P . The state equation for ideal gas gives α (P/P0)−1/3 for isotropic materials, where P0 is the atmospheric pressure.

In summary, a general and universal theory for superconductivity is proposed, which well explains both the low TC metals and high TC superconductors. According to the model, in order to obtain high TC materials, a high TU can increase the upper limit value for TC ; a large n and a small $1/\delta$ are required to provide strong carrier screening effect for small screening length. In addition, a small α is also required to protect the superconducting channel to a high temperature. This model, does provide a useful insight and powerful guidance for the future study of superconductivity.

(2 K) (10 K) (10 K) (3 K) (6 K) (20 K)

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AUTHOR CONTRIBUTIONS

X.S. proposed the idea, model, and theory of this work.

Z. L. and X.S. performed the primary theoretical derivations and established the mathematical model. Z.G collected the experiment data. Z.L., Z.G. and X.S, analyzed the experimental data. X.S. wrote the original draft of manuscript. All authors contributed to the subsequent revision, discussion, and polishing of the final text. X.S. supervised the entire project.

DATA AVAILABILITY

The data that support the findings of this study are included within the article.

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Supplementary Note 1: Derivation of the n−1/6 Dependence of the Thomas-Fermi Screening Length

The density of states at the Fermi level is given by

According to the Thomas-Fermi screening theory, we provide a derivation of the power-law relationship between the screening length λ and electron concentration n. In a degenerate electron gas, the screening wave number k is defined by the density of states at the Fermi level, g(EF)

Consequently, the relationship for the screening wave number becomes

g(EF)

g(EF) =

m0 e2 31/3 1/6 $ $0 2 π 4/3

Thus, the Thomas-Fermi screening length λ, which is the the reciprocal of k, follows the formula below

For three-dimensional free electron gases, The Fermi wave number kF is directly related to the carrier concentration n through the relation kF = (3π 2 n)1/3

m 0 kF π 2 2

$ $0 2 π 4/3 −1/6 m0 e2 31/3

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

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