

# Mechanism of high temperature superconductivity - the BCS theory and a new electron pairing medium

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## Abstract

High temperature superconductivity, BCS theory and the mechanism of high temperature superconductivity are introduced. The author proposed, for the first time, the change of the electron cloud ions can be used as a new medium for electron pairing. Calculation results are given. The results show that the frequency of the change of the electron cloud is close to (and higher than at normal pressures) the frequency of the lattice vibration. It is possible to explain the phenomenon of high temperature superconductivity by using the change of electron cloud instead of lattice vibration.

**Key words:** high temperature superconductivity; BCS theory; electron cloud;

## 0. Introduction

High temperature superconductors generally refer to superconductors with critical temperature ( $T_c$ ) above 77K (liquid nitrogen). Historically, high temperature superconductors (HTS) refer to copper-oxide ceramic superconductors, such as YBaCuO and HgBaCaCuO. High temperature superconductivity is of great theoretical significance and practical application value.

Up to now, the mechanism of high-temperature superconductivity (How are electrons paired?) is still unresolved. In addition, it is expected to find superconductors with higher  $T_c$ s, which can expand the application of superconductors. It is believed that if someone can explain the phenomenon of high temperature superconductors or find room temperature superconductors, he is sure to win the Nobel Prize.

## 1. the BCS theory <sup>[1][2]</sup>

In 1911, Onnes (Fig. 1) discovered superconductivity in Hg at 4.2K. The DC resistivity is zero. Onnes won the Nobel Prize in Physics in 1913 (for liquefying helium in 1908). In addition to zero resistivity, superconductors have another important property called "Perfect diamagnetism", that is, the magnetic field cannot enter the superconductor. So, superconductors are not completely equivalent to conductors with zero resistivity.

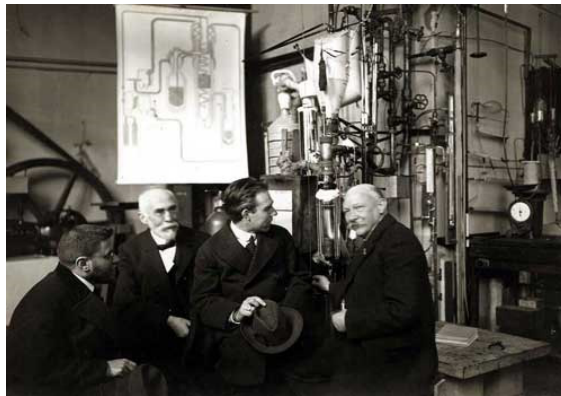


Fig.1 Onnes (the first one on the right)

Why does superconductivity occur? In 1957, 44 years after the discovery of superconductivity, Bardeen, Cooper and Schrieffer (Fig. 2) proposed the famous BCS theory. The theory can explain the superconductivity of conventional superconductors, such as Hg ( $T_c=4.2\text{K}$ ) and Pb ( $T_c=7.2\text{K}$ ). They won the Nobel Prize in Physics in 1972 (15 years after the BCS theory proposed).

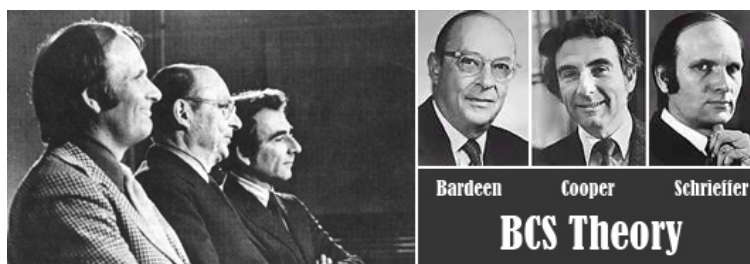


Fig. 2 Bardeen, Cooper and Schrieffer

There are several points about the BCS theory to introduce.

(1) When the theory was first proposed, many physicists did not understand it. The BCS theory was put forward in 1957, while the Nobel Prize was won 15 years later.

(2) In the BCS theory, two electrons form a pair (Cooper pair). There must be an

attraction between two electrons to form a pair. Electrons are negatively charged, so they repel each other. How can they attract each other? There must be a medium (or "glue") that creates attractive interactions between electrons. The medium is the lattice in conventional superconductors. The simple and intuitive model (Fig. 3) is as follows. When a free electron comes to a new position, the electric field of the electron causes the distortion of the lattice. When the free electron leaves, the lattice will not relax immediately (there will be a lag between the movement of the lattice and that of the electron) In this way, a positive charge region will appear, which will attract another free electron, thus creating the attraction between two free electrons.

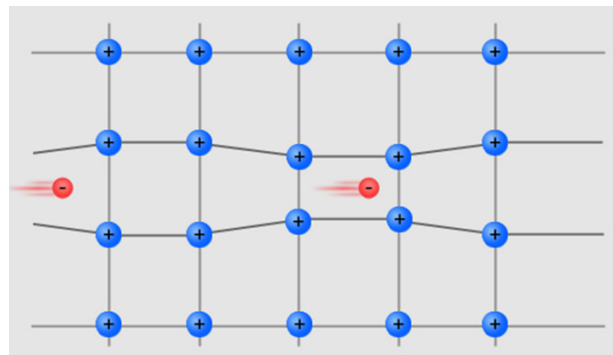


Fig. 3 Real space schematic diagram of the attraction between two free electrons caused by the lattice

(3) Isotope effect <sup>[3][4]</sup>. Before the BCS theory, scientists have discovered the isotope effect. For the same metal (for example, Hg), the atomic weight of different isotopes is different. Their superconducting critical temperatures are also different. The smaller the atomic weight, the higher the superconducting critical temperature. This is because the smaller the atomic weight, the higher the lattice vibration frequency. Higher frequencies cause higher critical temperatures. The BCS theory can satisfactorily explain the isotopic effect.

But the frequency cannot be too high and must be within a certain range. This is easy to understand. If the vibration frequency of the lattice is too low, it is difficult to respond to free electrons. If the lattice vibration frequency is too high, there will be no delay in the movement of the lattice relative to the electrons. In both cases, effective attractions cannot be achieved. As shown in Fig. 4, there is a typical curve where an

optimal frequency exists. The author believes that, for the lattice, the frequency is relatively low. For the superconducting critical temperature, the higher the frequency, the better. The optimal frequency will not be exceeded.

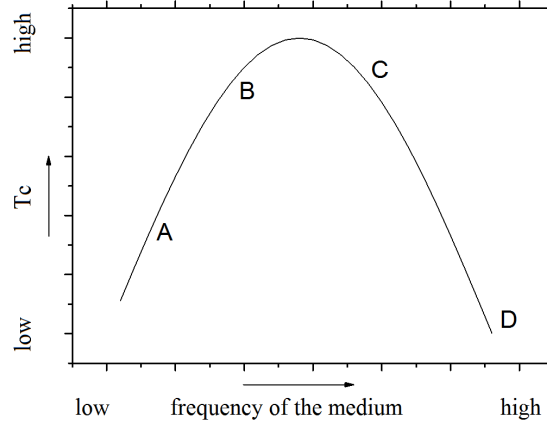


Fig. 4 Approximate dependence of the superconducting critical temperature on the frequency of the pairing medium

(4) McMillan proposed that, for conventional superconductors (Hg and Pb, et al, with the lattice as the pairing medium), superconducting critical temperature will not exceed 40 K (McMillan limit) at normal pressures. This is the famous Macmillan limit. The main reason is that the vibration frequency of the lattice under normal pressures is very low (at the point A of the curve in Fig. 4).

Can the superconducting critical temperature be improved by increasing the lattice vibration frequency? The answer is yes. The frequency of the lattice vibration will increase under high pressures. In 2015, Nature reported <sup>[5]</sup> a superconducting critical temperature of 203K in H<sub>3</sub>S system under 90 GPa. In 2019, Nature reported <sup>[6]</sup> a superconducting critical temperature of 250K (-23 °C) in a LaH<sub>10</sub> system under 170 GPa. The critical temperature is very close to room temperature, but it is achieved at ultra-high pressures. This severely limits its application. The frequency of the lattice vibration may be approximately at the position of point B in the curve. This kind of superconductivity can be explained by the BCS theory just like Hg and Pb, which is called conventional superconductivity.

## 2. Challenges encountered by the BCS theory

Not all superconductors can be explained by the BCS theory. Superconductors that cannot be explained by BCS theory are called unconventional superconductors.

(1) Nb. The  $T_c$  of Niobium 9.2K. For alloys with Nb, the superconducting critical temperature can be higher, for example,  $Nb_3Ge$  ( $T_c=23K$ ). However, Nb does not conform to the isotope effect. Other superconducting metals also deviate significantly from the isotope effect (while Hg and Pb are strictly obeying the isotope effect). This became the first challenge of the BCS theory. However, electron-lattice coupling is only part of the BCS theory, not all. If there is a new medium replacing the lattice to achieve electron pairing, then the BCS theory is still valid.

(2) Cuprate high temperature superconductors. This is the second serious challenge to the BCS theory. In 1986, Bednorz and Müller <sup>[7]</sup> discovered that superconductivity may exist in La-Ba-Cu-O with a critical temperature of 31K. In 1987, Wu Maokun et al <sup>[8]</sup> and Zhao Zhongxian et al, almost simultaneously produced Y-Ba-Cu-O superconductors. The superconducting critical temperature reached 93K under normal pressure. But the BCS theory predicts that the superconducting critical temperature caused by the electron-lattice coupling under normal pressure cannot exceed 40K. Fig. 5 shows the crystal structure of the Y-Ba-Cu-O superconductor. The most obvious feature is the layered structure, and there is a copper oxide plane. Scientists believe that the copper oxide plane is the main factor for superconductivity.

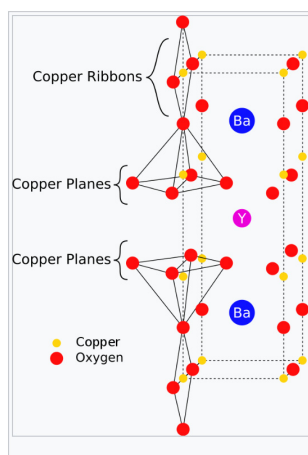


Fig. 5 Crystal structure of Y-Ba-Cu-O

(3) Iron-based superconductors. This is the third serious challenge to the BCS theory. In 2006, scientists discovered superconductivity for the first time in LaOFeP with a critical temperature of only 4K <sup>[9]</sup>. However, iron-based superconductors with  $T_c$ s more than 40K were found. The  $T_c$  of PrFeAsO<sub>0.89</sub>F<sub>0.11</sub> reaches 52K <sup>[10]</sup> and SmFeAsO<sub>0.85</sub> 55K <sup>[11]</sup>. In 2012, Q. K. Xue et al. successfully grow FeSe thin films with one unit-cell thickness on the surface of SrTiO<sub>3</sub> substrates by the MBE method <sup>[12]</sup>. The monolayer FeSe showed a  $T_c$  above the liquid nitrogen temperature (77K). In 2014, Q. K. Xue et al. reported the FeSe monolayer with a  $T_c$  over 100K <sup>[13]</sup>. Fig. 6 shows the crystal structure of the 122-type iron-based superconductor, which also has a distinct layered structure.

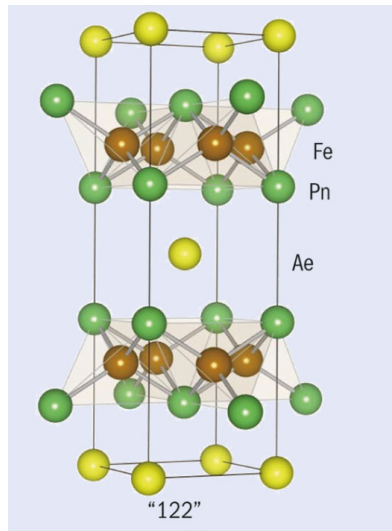


Fig. 6 Crystal structure of 122-type iron base superconductor

From whether they can be explained by the electron-lattice coupling, superconductors can be divided into two types: conventional superconductivity (which can be explained by the electron-lattice coupling) and unconventional superconductivity (which cannot be explained by the electron-lattice coupling). In fact, the problem of "mechanism of high temperature superconductivity" is the problem of electron pairing in unconventional superconductivity. There is no consensus on this problem.

There is an important difference between these two types of superconductors. Both Hg and Pb are conventional superconductors. The valence electron configuration of Hg is  $5d^{10}6s^2$ . The valence electron configuration of Pb is  $5d^{10}6s^26p^2$ . The d orbitals of both

elements are fully filled. While, for unconventional superconductor Nb, its valence electron configuration is  $4p^6 4d^4 5s^1$ . In the cuprate superconductors, the valence electron configuration of  $Cu^{2+}$  is  $3d^9$ . In iron-based superconductors, the valence electron configuration of  $Fe^{2+}$  is  $3d^6$ . There are atoms or ions in the three unconventional superconductors with not-fully-filled orbitals. The author believes that this is the most important difference between the two types of superconductors. It is possible to explain the electron pairing mechanism in unconventional superconductors from this perspective.

### **3 A possible new electron-pairing medium**

The electrons in the unconventional superconductors are also paired. This has been proved experimentally. However, it cannot be explained by the electron-lattice coupling. This has created a problem that has not been solved until now. There may be hundreds of theories that have been proposed <sup>[14][15][16]</sup>, but there is still no consensus. There are two possibilities. One is that there is a lattice-like medium in the unconventional superconductors that pairs the electrons. One is that there is no medium similar to the lattice. The electrons are naturally paired due to the principles of quantum mechanics. P. W. Anderson <sup>[17]</sup> raised an important question in 2007: Is There Glue in Cuprate Superconductors?

If there is a medium in an unconventional superconductor similar to the lattice that pairs the electrons, how should the medium be found or discovered? There is no doubt that this medium should be able to change under influences, and the frequency of the change should be appropriate (see Fig. 4). The frequency cannot be too high or too low. What should the value of the frequency of the new medium be? If it is close to the frequency of the lattice vibration, then it is possible to pair the electrons. Table 1 shows the highest lattice vibration frequencies of three typical conventional superconductors. It can be reasonably guessed that the frequency of the electron pairing medium in the unconventional superconductor should also be approximately within this range, such as 100meV - 300meV.

Table 1 Maximum phonon frequencies and  $T_c$  of three typical conventional superconductors

	Pb	MgB <sub>2</sub>	H <sub>3</sub> S
$\omega / \text{meV}$	9	90	250
$T_c / \text{K}$	7	39	164

To verify the above ideas, the author has studied the Nb, iron-based superconductors and cuprate superconductors by first-principles calculations. First-principles calculations use software and high-performance computers to study the properties of materials. One of its important advantages is the ability to calculate phenomena and properties that are experimentally unobservable.

Firstly, the author explores the change of the superconductor under certain influence. It is found that the charge density changes (electron clouds) obviously. Then, the real-time evolutionary method is used to study the frequency of this change.

Fig. 7 shows the charge density change of iron-based superconductor LaFeAsO under influence. Please refer to the literature [18] for the author's calculation methods. It can be seen that whether a Li<sup>+</sup> ion is inserted or a uniform electric field is applied, the electron cloud of Fe<sup>2+</sup> ions changes significantly. And the change is not strictly along the direction of the electric field, but is similar to a rotation. Some areas increase, while some areas decrease. No similar changes occurred in other ions (La<sup>3+</sup>, O<sup>2-</sup> and As<sup>3-</sup>). Why? This is because the 3d orbital of Fe<sup>2+</sup> ions is not fully filled (3d<sup>6</sup>). One explanation is that Fe<sup>2+</sup> ions do not have spherical symmetry, so rotation occurs under the influence of an electric field. Another explanation is that the 3d orbital of Fe<sup>2+</sup> ions recombines and electrons refill, so the electron cloud changes.



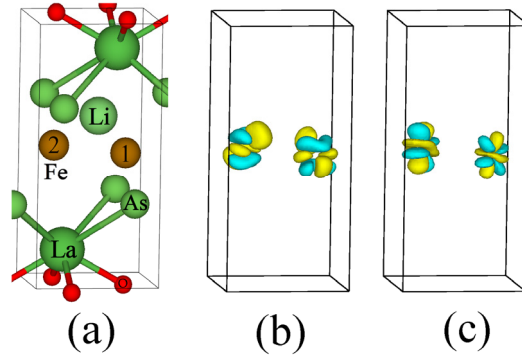


Fig. 7 (a) Crystal structure of LaFeAsO; (b) charge density change caused by insertion of a Li<sup>+</sup> ion; (c) charge density change caused by a uniform electric field (yellow indicates increased charge density, blue decreases)

Fig. 8 shows the charge density change of cuprate superconductor HgBa<sub>2</sub>Ca<sub>2</sub>Cu<sub>3</sub>O<sub>8</sub> under influence. It can be seen that the electron cloud of Cu<sup>2+</sup> ions undergoes a distinct rotation-like change, which is the same as the change of the Fe<sup>2+</sup> ion iron-based superconductors.

It must be emphasized that, in Fig. 8, the electron cloud of O<sup>2-</sup> has also undergone significant changes. The electron cloud of Cu<sup>2+</sup> changes because its valence electron configuration is 3d<sup>9</sup>. The d orbital is not completely filled. Why does the electron cloud of the O<sup>2-</sup> ion change? The author believes that this is because the valence of the O<sup>2-</sup> ion is not strictly -2. There are holes on it, so its electron cloud does not have perfect spherical symmetry. It will rotate under the influence of the electric field.

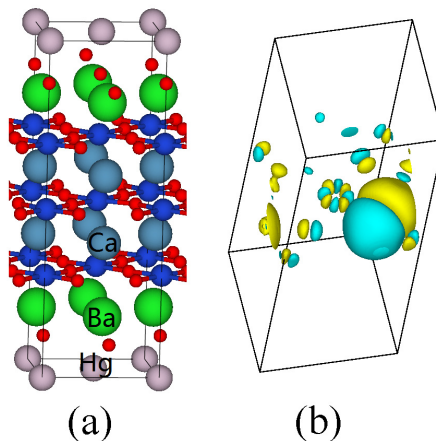


Fig. 8 Cuprate superconductor HgBa<sub>2</sub>Ca<sub>2</sub>Cu<sub>3</sub>O<sub>8</sub>, (a) crystal structure; (b) charge density change caused by slight changing the position of an oxygen ion

The author believes that the rotation of the electron cloud can act as a pairing medium for unconventional superconductors. The pairing mechanism is like this. When a free electron comes to a new location, the electron cloud of the ions in the vicinity will change. In this way, the charge density around the free electron will decrease (equivalent to the emergence of a positive charge). When the free electron leaves, the electron cloud of the *will not relax immediately*, so that there will be a region lack of charge (equivalent to a positive charge region) that will attract another free electron. Attraction between two free electrons appears. This mechanism is essentially the same as the electron-phonon interaction, except that the medium is the change of the electron cloud, not the displacement of ions.

However, according to the Bonn Oppenheimer approximation <sup>[19]</sup>, the mass of electrons is very small (about 1/1800 of the mass of the proton). The change of the electron cloud may be very fast compared with the lattice, and the frequency may be much higher than that of the lattice vibration. If it is true, the change of the electron cloud cannot be used as a medium for electron pairing.

But, can the electron cloud change as slow as the lattice? This must be verified by calculation or experiment. The author used the TDDFT method to calculate the real time evolution of charge density for Nb, iron-based superconductors and cuprate superconductors. Please refer to <sup>[20]</sup> for the detailed calculation method. It was first discovered that the frequency of the change of the electron cloud is very close to the frequency of the lattice vibration. So, the change of the electron cloud can be used as a superconducting electron pairing medium.

Fig. 9 shows the evolution of the charge density of an iron-based superconductor LaOFeP. The change was excited with a time dependent electric field. It must be emphasized that the excitation vanished after a certain period of time and then the charge density oscillates freely. Fig. 9 shows the results of the free oscillation of the charge density. 100 represents the charge density change after 100 steps. The time of one step is  $0.002 \hbar / eV$ . It can be clearly seen that there is almost no change in charge density after 100 steps. As the evolution time increases, the change of the charge density becomes more and more obvious. It reaches its maximum after about 450 steps.

Moreover, the change of the charge density is mainly the change of the electron cloud of  $\text{Fe}^{2+}$  ions. The frequency of the change of the electron cloud of the  $\text{Fe}^{2+}$  ions is approximately 280 meV. This result is very close to the lattice vibration of  $\text{H}_3\text{S}$  at an ultrahigh pressure of 250 GPa. This is unexpected, and the electron cloud has not changed very quickly. So, the electronic cloud can be used as an electronic pairing medium.

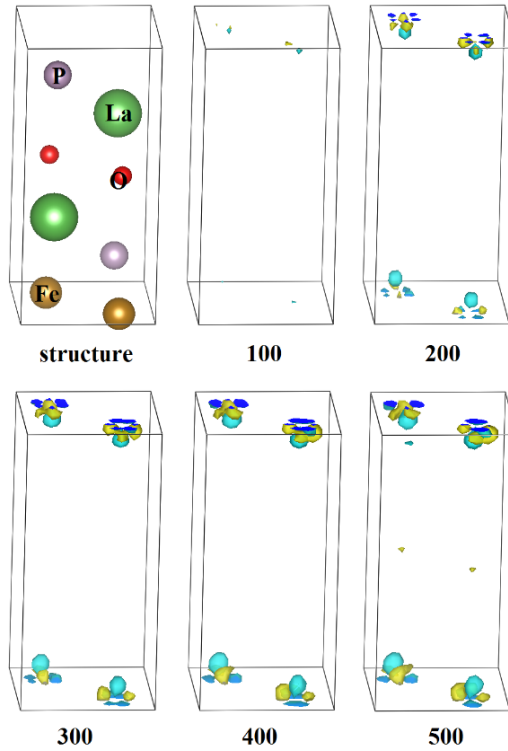


Fig. 9 Crystal structure of  $\text{LaOFeP}$  and real time evolution of the charge density (yellow indicates increased charge density, blue decreases)

Fig. 10 shows the crystal structure and the evolution of the charge density change of metal Nb. Focus on the Nb atom indicated by an arrow. It can be seen that the charge density of this Nb atom is also gradually changing. The maximum of the change density change is reached after approximately 1000 steps. The corresponding frequency is approximately 125 meV. This result is close to the frequency of lattice vibration too.

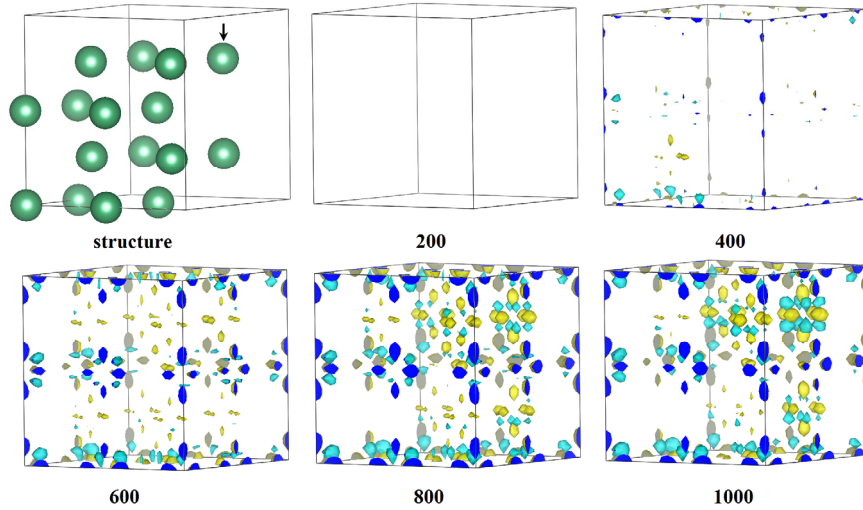


Fig. 10 Crystal structure of Nb and real time evolution of the charge density

Fig. 11 shows the crystal structure and charge density evolution of  $\text{CaCuO}_2$ .  $\text{CaCuO}_2$  is the parent structure of cuprate superconductors <sup>[21]</sup>, which contains the copper oxide plane, and has a simple structure. Focusing on the  $\text{O}^{2-}$  ion indicated by an arrow, the electron cloud of the  $\text{O}^{2-}$  ion gradually changes. The frequency is about 250 meV, which is also close to the lattice vibration frequency.

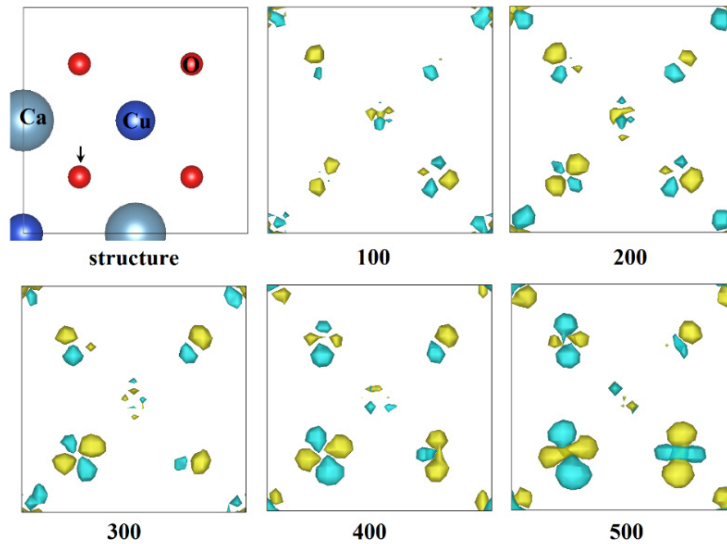


Fig. 11 crystal structure of  $\text{CaCuO}_2$  and real time evolution of the charge density

Fig. 12 shows the same crystal structure of  $\text{CaCuO}_2$  and the charge density evolution, but the mode is different from that given in Fig. 11. Focusing on the  $\text{Cu}^{2+}$  ion marked with an arrow, the change of the electron cloud reaches the maximum after about 350 steps, and the corresponding frequency is about 360 meV.

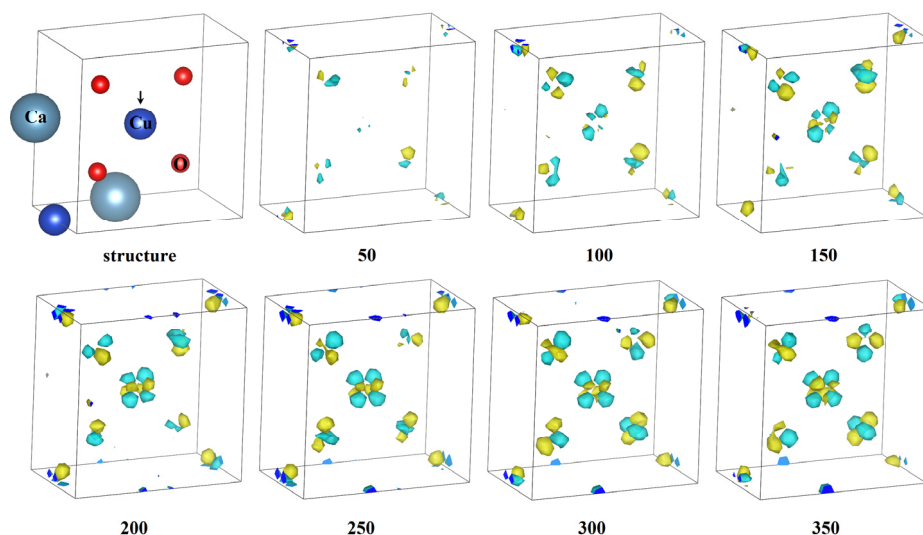


Fig. 12 Crystal structure and charge density evolution of  $\text{CaCuO}_2$  (the excitation and vibration mode are different from Fig. 11)

In addition to the above three superconductors, the author has also found similar phenomena in other superconductors [22][23][24][25][26].

Compared with the lattice vibration in conventional superconductors under normal pressure, the frequency of the change of the electron cloud can be higher. So, a higher superconducting critical temperature can be obtained.

For different unconventional superconductors, the author believes that in addition to frequency, the superconducting critical temperature also has an important relationship with the shape of the orbital. For the cuprate superconductors, the author thinks that the  $\text{O}^{2-}$  ion in the copper-oxide plane plays the most important role. The 2p orbital is not completely filled, and its shape is simple. The area of the charge density change is the largest, which is advantageous to the pairing of the electrons, so the superconducting critical temperature is the highest. For the iron-based superconductors, the 3d orbital (play the most important role in iron-based superconductors) is complicated, and the charge density change region becomes smaller, so the critical temperature is lower than the cuprate superconductors (It is not entirely true. It is In general.) For Nb, the 4d orbital active, the shape is more complicated than the 3d orbital, and it is more difficult to form a larger residual charge region, so the transition temperature is very low. There is also a type of unconventional superconductor called

heavy fermion superconductor, in which 4f or 5f orbital is active, the orbital shape is the most complicated, and the superconducting critical temperature is lower than Nb.

In short, the author believes that the BCS theory is correct. For the first time, the author proposed that the change of electron cloud can be used as an electron pairing medium. It is possible to explain the high temperature superconducting mechanism by replacing the lattice vibration with the change of electron cloud.

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