Unveiling The Unconventional Superconductivity Via Cuprates and Twisted Bilayer Graphene

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Abstract. Since the twentieth century, the exploration of superconducting material, a type of material that exhibits zero electrical resistance, has remained a paramount topic of inquiry within the scientific community seeing its excellent potential in bringing dramatical improvement to the material science and electric power and high-field magnet technology. However, earliest materials with superconducting aptitude requires extreme condition for critical temperature: It needs a temperature close to 0K, which is hard for application. This severe technical issue then led to an exploration to materials with higher temperature to reveal superconductivity. With decades of efforts, despite there are still arguments in working principle, new material like cooper oxide and iron-based material were found to have critical temperature higher than the limit predicted by Bardeen–Cooper–Schrieffer theory. However, recent experiment and accompanying working principle for the superconductivity of twisted layer graphene may suggest that it has greater potential in high temperature superconducting, due to its tunable property and relatively simpler working mechanism.

Keywords: High-temperature superconductivity; Cooper pair; Bardeen–Cooper–Schrieffer theory, Twisted bilayer graphene.

1. Introduction

For many materials in industry like metals or compounds, an eccentric property that these materials suddenly have their resistivity drop to zero can be seen when the temperatures of the materials are in a value that is low enough. This phenomenon is also known as superconductivity and the material that possess superconductivity is the superconductor. In history, the discovery of Superconductivity was contributed by the Dutch physicist Heike Kamerlingh Onnes, who succeeded in liquify the helium at a temperature 4.2K. In 1911, Kamerlingh found that when mercury was also cooled to a temperature of 4.2K, it shows superconductivity [1]. After this discovery, in 1933, W. Meissner and R. Ochsenfeld, a group of German physicists, found that for a superconductor in a temperature below T_c, the critical temperature for the superconductor to have superconductivity, there will be no magnetic field anywhere inside the superconductor [1]. This phenomenon is called as the Meissner effect. Since both Meissner effect and zero resistance are so unique and profound, these two things are often denoted as condition for a material to be superconductor. Ever since that, researchers have found more and more materials to have superconductivity with different temperature.

Due to the possible improvement that can be bring to the electric power generation and transmission by superconductor, the research in superconductor is always popular. As a result, the working principle for these conventional superconductors at extreme low temperature, the Bardeen–Cooper–Schrieffer (BCS) theory, were also developed in 1956. The theory states that the appearance of superconductivity in material is linked to the interaction between phonons and electrons in lattice vibration in the material. This type of interaction eventually forced the electrons to combine into pairs known as Cooper pairs and have a net zero spin for each pair. These pairs are then unlikely to be affected external by magnetic field and the thermal motion of lattice in low temperature. Since it can be used to explain the property of superconductor, it was widely agreed and still being considered as the base of the superconductivity. However, the temperature required by superconductor to show its magic were always too difficult to be used in application since the cooling have a considerable cost. Therefore, researchers continued to search for clues for superconductors with higher critical temperature. In 1986, although the BCS theory predicts that the limit for critical temperature for

superconductor were 40 Kelvin, Swedish scientist K. A Müller and his colleagues successfully discovered the superconductivity of Ba−La−Cu−O system for a temperature higher than the limit anticipated by the BCS theory [2]. Nevertheless, the underlying theory developed for this new high temperature superconductor is not complete and still being argued. Similar things can also be found in iron-based superconductor discovered by Japanese physicist Fujiwara and his team [3]. Despite superconductivity were observed on this new material with high critical temperature, the principle for this unconventional superconductivity is still in debate. In recent years, a group led by Pablo Jarillo-Herrero found that the overlapping of two sheets of graphene that are twisted relative to each other by a specific angle around 1 degree somehow showed unconventional superconductivity with a tunable range for critical temperature [4]. This type of material not only possess a relative complete and indisputable theory, it is also highly reproducible and adjustable since graphene has mature manufacturing process and is easy to edit on structure.

As a result, the article will give introduction to the BCS theory first. Then give a comparison between iron and cooper compound with the twisted bilayer graphene to illustrate why this is a new promising material for high temperature superconductor then proceed to delineate a comparative analysis among some of the most representative high temperature superconductor, i.e., copper-based compounds and twisted bilayer graphene. This is essential to underscore the burgeoning potential of the latter as a propitious candidate for high-temperature superconductivity.

2. BCS theory

Since the superconductor require sharp critical temperature for its unique property, the idea that superconductivity is a cooperative phenomenon where many conduction electrons participate had been formulated. However, there were two difficulties to confirm that idea as the actual mechanism. Firstly, the fermion nature of electrons requires then to obey Fermi-Dirac distribution and therefore forbidden two electrons to occupy the same quantum state. This leads to the possibility of electrons having cooperative phenomenon to be zero. Secondly, the Coulomb repulsion between electrons will prevent them from approaching each other regardless how small the repulsion could be. In 1956, Cooper proposed that electrons can have attraction through a lattice distortion that can overcome the coulomb repulsion [5]. By considering electron-phonon interaction, which relates to the absorption or emission of the phonon, paves a way to a new, systematic explanation to the superconductivity. Those isolated electrons, in this case, could attract other electrons in the long range and then combined to form a bound pair that behaves like a boson seeing that the screening by phonon can effectively reduce the repulsion between electrons. Since all bosons are capable of being condensed coherently into just a single quantum state, the coherent state of these bosonic Cooper pairs is separated from other excitations by an energy gap at condensation. This means that any external perturbation, like thermal fluctuations or impurities, needs energy at least equivalent to this gap to break the Cooper pairs. As a result, these pairs can flow without scattering, resulting in zero electrical resistance. This theory is later known as the BCS theory.

2.1. Cooper Pair

2.1.1 Formation

The formation of cooper pair can be understood by flowing argument. When electron passing through the lattice, distortion will be made due to the attracting force between positive ions and electrons. This distortion will then affect the motion of electron. Usually, it will drag the electron for a distance.

$$d \approx v_f \frac{2\pi}{\omega_D} \approx 100 - 1000nm, \tag{1}$$

where ω_D is the Debye frequency, a cut off frequency in lattice vibration that above this frequency no oscillations would take place, and v_f is the fermi velocity, the velocity of electrons at

the Fermi level in a metal at absolute zero temperature [1]. As shown in Fig.1, this deformation then led to the accumulation of positive charge in a small area. As a result, another electron will also be attracted to pass through this region. Then attraction will then continuously increase and reach a maximum when the second electron goes along the same path of the first electron and is now at a distance d away. The attraction brought by positive ion is a polarization force therefore it can last for a longer period compared to the instantaneous coulomb repulsion (Here, relativity is not considered in this background, classical field theory is applied to this model). In other words, despite the lattice distortion is weak, it can cause long term effects that eventually mediated the repulsion force between electrons and force them to combine into a pair. For a more detailed physics picture, Cooper studied what would happen to two electrons with momentum $\overrightarrow{p1} = -\overrightarrow{p2}$, where the magnitudes of the momentum are slightly greater than the known Fermi momentum pf, the average momentum of electrons in the material, are added to a filled Fermi plane. It can be deduced that no energy state below the Fermi level can be occupied since they are already fully occupied by other electrons. Hence the electrons, in order to follow the Pauli exclusion principle, would prefer to approach to each other and form a pair. This can be achieved by exchange of a virtual phonon to lose energy. In this case, the pair can be considered as a Boson and no longer follow the Fermi-Dirac statistics. The interaction is also allowed by uncertainty principle.

$$\Delta E \Delta t \geq \frac{\hbar}{2} \tag{2}$$

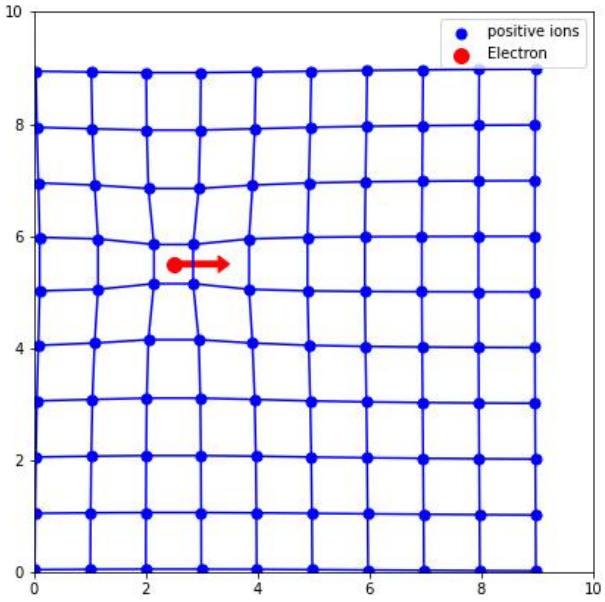


Fig. 1. The deformation of lattice brought by the motion of an electron.

These so-called Cooper pairs possess a binding energy around 10^{-3} eV to 10^{-4} eV. Since the binding energy is small, a low temperature is required to prevent thermal excitation from breaking the combinations. In addition, when the two electrons have momentum opposite in direction but equal in magnitude, the binding is the strongest since the binding pair in this case would have a 0 momentum and stay in the spin-singlet state by having a symmetric spatial wavefunction. Under this circumstance, the radius of the Cooper pair can be determined by using Hamilton operator to be.

$$r = \frac{\hbar v_F}{E_B} \tag{3}$$

Where E_B is the binding energy and v_F is the Fermi velocity, v_F is usually around 10^8 cm/s in metals. Hence, r is usually a value also around 100-1000 nm, similar to the value of d [1].

2.1.2 Condensation

With derivations above, it can be seen that Cooper pairs are actually large in size when being compared with the distance between different electrons, which is around 0.01 nm. Cooper pairs can therefore be considered as extended entities, leading to a significant overlap between them. As a result, one can find for the internal space occupied by one pair, there actually exists thousands of other pairs. At the instant that two electrons form a Cooper pair, the majority of nearby electrons would also realign into bound pairs. At temperature below critical temperature, these Cooper pairs can then be condensed back to the state with the lowest energy, the ground state. They then creating an energy gap with a sum value of 2Δ in the energy spectrum, which is the energy required for even one single pair to be separated into electrons. The isolated electrons will then occupy the new, empty levels above the energy gap. In general, when the material is at a temperature below T_c, but greater than absolute zero, all the pairs are in ground state, all the solo electrons stay in the levels above the Fermi level, as illustrated in Fig. 2.

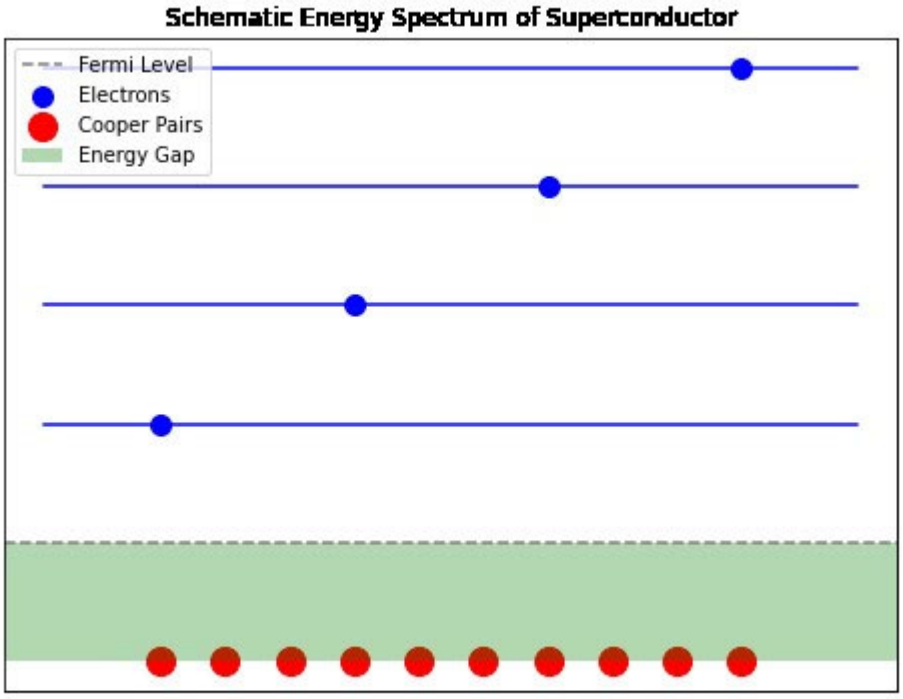


Fig. 2 Schematic Energy Spectrum for electrons in superconductor

2.2. Formulation of the theory in microscopic view

With the introduction of Cooper pairs, BCS theory can explain several effects of superconductors including the Meissner effect. The interaction between electrons via virtual phonons is calculated to be attractive when the difference in energy between different energy states of electrons is lower than the energy for a phonon, $\hbar\omega$. The superconducting phase will be constructed when the attractive force is greater than the screened Coulomb repulsion. Then as mentioned above, electrons combined to form Cooper pairs and condensed at the ground state. Two fascinating properties of these pairs in the ground state are the facts that they are not only indistinguishable from each other but also in phase. Hence the pairs can then exchange their companions within a thin energy band around Fermi energy and be represented by a single wavefunction as one ensemble. At this state, the pairs are highly ordered and well-structured that this order spreads over the whole lattice and is the main reason for most peculiar properties. For instance, when an external magnetic field is applied to the

superconductor and intend to pass through it, it would induce motion in these Cooper pairs (since they still carry charge). This motion doesn't encounter any resistance because the superconductor has zero resistivity. This would in principle disturb the order of Cooper pairs. However, the binding energy would prevent this, so it requires magnetic field to provide energy larger than the gap to break the Cooper pairs. As a result, the weak magnetic field are all repelled by the superconductors in this case.

In addition to that, BCS theory can calculate the specific value for the transition temperature and the energy gap. In the weak coupling limit that $kT_c \ll \hbar\omega$, the expression for T_c is obtained by solving ground state wavefunctions with expression.

$$kT_c = 1.14\hbar\omega \exp\left[-\frac{1}{N(0)V}\right], \tag{4}$$

where $N(0)$ is the density of states for single spin per unit energy at Fermi level and V is the electron-phonon interaction parameter [1]. It can be seen that kT_c is proportional to the phonon energy, which agrees with the experiment of the isotope effect in superconductors. In principle, the highest critical temperature can be reached is around 30 K, which is calculated by considering highest Debye frequency for materials, with the introduction of typical values of N and V for known superconductors. This limit is later being broken by a new type of superconductor, the Ba−La−Cu−O oxide. The relationship between energy gap and critical temperature is given by equation.

$$2\Delta = 3.5kT_c, \tag{5}$$

which can also be calculated from the wavefunction. The detailed analysis of all equations in this section is beyond the aim of the article, and the detailed derivation can be found in Ref. [5].

3. Conventional Cooper Oxide Superconductor

Despite the BCS theory has well explained many properties of the conventional superconductors like metals, it does not solve the core issue that it requires an incredibly low temperature for the superconductor to reveal its magic. Hence new materials have been made to seek whether there is a chance to increase the critical temperature. Luckily, in 1986, superconductivity in Ba−La−Cu−O oxide proved that there do exists material that can have a temperature higher than the limit predicted by Eq. (1.4). The final critical temperature reached by this metallic, oxygen deficient material is 35 K, which is slightly higher than the 30 K anticipated by BCS theory [2].

3.1. Discovery

The discovery of cooper oxide superconductor is based on the consistent searching and attempts among the elements in the periodical table. Especially for materials like alloys and compounds. Before the Ba−La−Cu−O system, superconductivity was found in different families. However, none of them have succeed in breaking or even approaching the theoretical limit of T_c predicted by the BCS theory, which implies a possibility of zero for application. Some of the materials are listed below in Table 1 [6]. All attempts to approaching the limit were failed until in 1986 suddenly, two physicists in Zurich, Bednorz, and Muller, realized the superconductivity for a T_c at 35K in the oxide compound of the type $La_{5-x}Ba_xCu_5O_{5(3-y)}$, where x and y can be adjusted manually for optimization of electronic property. Varying the content of Ba and La affects the charge carrier concentration and the crystal structure, both of which are critical for the onset and optimization of superconductivity.

Table 1. Three materials comparing

No.	Type of superconductor	Example	Max. Tc (K)
1	Pure elements	Fe	9.2
2	Carbides	NbC	11
3	Semimetal superconductor	La_3Se_4	10

3.2. Sample Prepartion and Measurement

The material was prepared by the method called co-precipitation, a method used to remove ions, particular metal ions, from solution. By adding different substance to the solution, one substance can initiate precipitation, and as it forms solid particles in the solution, it can eventually adsorb other solutes in the solution that would not precipitation by themselves in the similar situations. By co-precipitating the nitrates of Ba, Cu, and La in appropriate ratios, a solid-state reaction was initiated at 900 degrees in a reduced atmosphere. This process yielded samples shaped as pellets, which were sintered at the same temperature. The sample's resistivity, which fluctuates based on temperature, has been identified to transition through three distinct phases. As illustrated in Fig. 3, for samples where x = 0.75 and x = 1 with y > 0, the resistivity initially falls linearly with a decrease in temperature, then ascends logarithmically, and finally plunges dramatically by three orders of magnitude. This eventually results in the manifestation of superconductivity and the Meissner effect [2]. Additionally, variations in the resistivity curve were observed when changing the sample current, suggesting the superconductivity is granular in nature. Ultimately, this kind of superconductor was identified as having an O^{2-} deficient phase with mixed valences of the Cu components. This means both Cu^{2+} and Cu^{3+} ions are present, leading to high metallic conductivity and a pronounced λ, which is indicative of strong electron-phonon interactions that contribute to superconductivity.

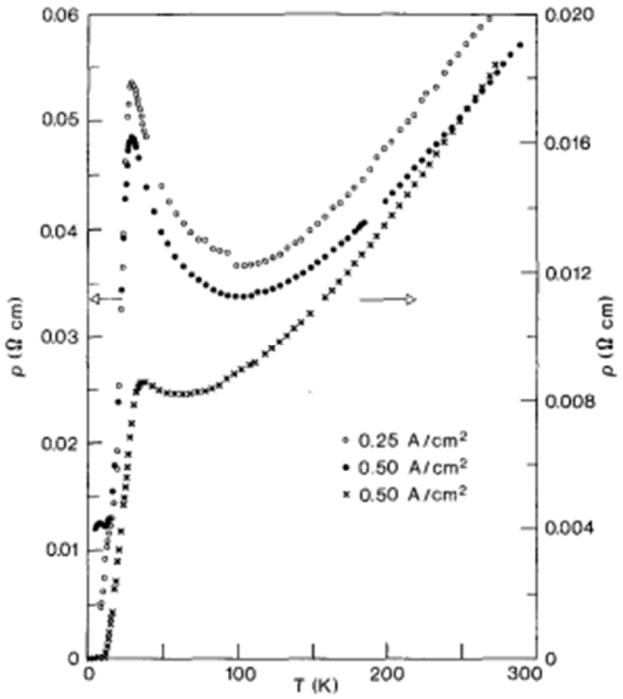


Fig. 3 Resistivity against temperature curve for $La_{5-x}Ba_xCu_5O_{5(3-y)}$ with $x = 1$(upper curves left scale) and $x = 0.75$ (lower curve right scale) [2]

3.3. Achievements and Defects

At the time, even though the prevailing belief was that electron-phonon coupling drove superconductivity, scientists were on the verge of abandoning hope for achieving high Tc, which the BCS theory deemed unattainable. A glance at the materials in the table reveals that none approached this theoretical limit. However, shortly after introducing this system, oxide superconductors boasting progressively higher critical temperatures began to emerge, reminiscent of mushrooms sprouting after a rain. The unveiling of oxide superconductivity marked the dawn of a new era dominated by high-temperature superconductors, specifically those with CuO layers and Cu components. Notably, materials with CuO layers, such as $YBa_2Cu_3O_7$, now referred to as cuprates, continually display soaring Tc values that surpass 77K, the boiling point of nitrogen [7]. The oxide superconductors are still having a debating environment for its theoretical basis of its superconductivity. Some mentioned

that it is the strong electron-electron interaction that led to this high critical temperature, while others proposed that the true reason could be due to the multi-layer structure of material or spin fluctuations in the cooper atoms. In general, a comprehensive theory is not yet been proposed. Furthermore, the preparation of oxygen deficient material usually involves high temperature environment and therefore complicate for massive production for application. In addition, the Cu oxide was usually fragile and unable to extend compared to traditional metal superconductors, which makes it hard for mechanic application in practical use. Finally, the three phases contained in this type of superconductor is complicate for analysis that makes it difficult to optimize the structure of material.

4. Twisted Bilayer Graphene

Soon after the discovery of the Cu oxide superconductor, other types of compounds were also found to have superconductivity with a relatively high critical temperature. Despite most of the new compounds have similar issues that could be found in the cooper oxide, still, new material with less defects were discovered. Among all the materials, the twisted bilayer graphene (TBG), now seems to be another promising one that can be used in real life application. This amazing material, when being first studied and recognized as a superconductor in 2018, is not only having an easy process of production but also has a tunable electronic property. In addition, study of the phase diagram of this material also possesses an unprecedented resolution. In addition, this material is thought to have a less complicate theoretical background to explain its superconductivity since most experiments agree with the theories very well.

4.1. Surprising Discovery

In 2018, Pablo Jarillo-Herrero and his group started their experiment on investigating the TBG [4]. Despite the single layer of graphene was successfully isolated in 2004 by Andre Geim and Konstantin Novoselov, physicists were then amazed by the moiré pattern that can be seen in graphite that contains multiple layers of graphene [8]. The term moiré is to refer the pattern formed when two sheets were squeezed together without being perfectly aligned. As for the case of graphene, when the two periodic lattices of carbon are overelaid with a slight angle, the hexagonal lattices will be unable to align perfectly everywhere. The mismatch and rotation would in the end create a larger periodic pattern and form a superlattice structure. The new pattern would possess a larger periodicity compared with the two separate sheets. Physicist Eva Andrei also found that by using scanning tunneling spectroscopy to observe the superlattice, one would find that the electronic band structure of the graphene has changed depending on the twisting angle of the two sheets. Especially when the angles is around 1.1 degree, a flat band, a band structure that electrons are effectively away from the other, enabling other interactions like magnetism to happen, were observed. This angle is later being dubbed as the ‘magic angle’ by Allan MacDonald and Rafi Bistritzer in 2011 [9]. At that time, no one would expect superconductivity to appear on this material.

At first, Jarillo-Herrero and his team intended to investigate what effect that the flat band would bring to the electronic property of the TBG. In principle, the twisted angle would enhance the electron interactions since flat bands near Fermi level would result in slow motion of electrons. The team did observe TBG become correlated insulator at low temperature. This suggests that strong electronic correlations occured, corresponds to the prediction above. The property that varies with temperature is a precursor to superconductivity for materials like cuprates. Hence, the team then applied a perpendicular electric field to the TBG material, which allow them to add or remove electrons from the structure. They then discovered that after adding or removing a certain number of electrons, a sharp drop in resistance suddenly took place in the transport measurement performed in a dilution refrigerator at low temperature [10]. Furthermore, the superconducting phase in this case is fragile that could be affected by the magnetic field and the specific doping levels. This evidence eventually confirmed the superconductivity in the material. As shown in the Fig.4 that contains both superconducting phase and insulating phase below, at zero magnetic field, diagram a has shown a

transition from metal to superconductor. The intermediated region is being identified as the half-filling insulating phase. Once magnetic field is being applied, superconductivity breaks and transition to insulator will be seen. If the magnetic field is too strong, Zeeman effect plays the dominant role, and the system is metallic everywhere for all carrier densities.

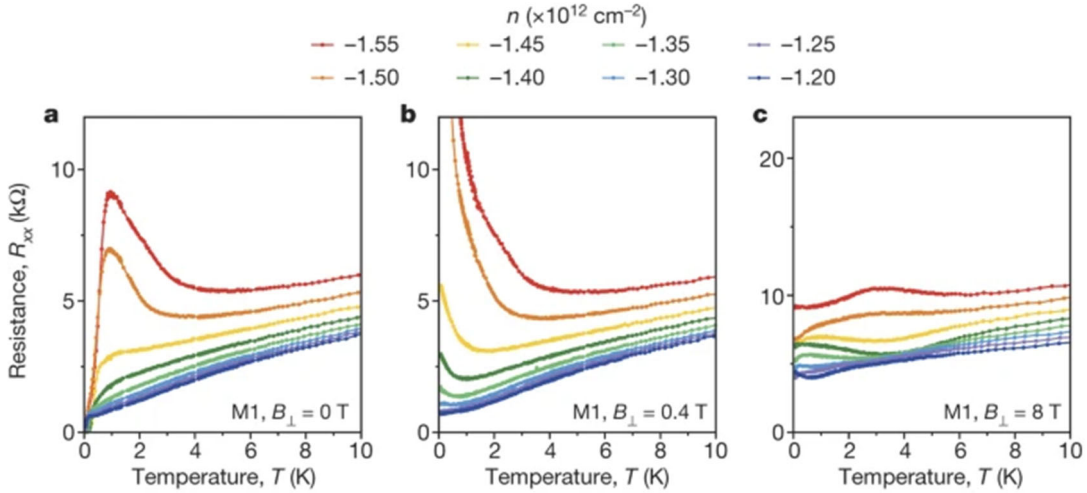


Fig. 4. Phase diagrams of TBG at different fields and carrier densities n [4].

4.2. Production of Material

The production of the TBG is actually a simple process with no annealing or similar highly cost manufacturing process involved. The samples were fabricated by using a modified dry-transfer technique. The two individual layers of graphene were obtained using mechanical exfoliation. The two layers were peeled and selected on SiO_2 chip by using optical microscopy and atomic force microscopy to determine their quality. In addition, hexagonal boron nitride with a thickness of 10-30 nm were also exfoliated to tear a graphene flake using van der Waals force. With a polymer stack to pick up and supporting the boron nitride, a graphene flake could be obtained at room temperature. The separated graphene flakes were then rotated by a twist angle about 1.05-1.16 degree and being restacked back to an ensemble. This process allows the tuning of electronic band structure to be controllable.

4.3. Properties and Possible Mechanism

To determine what is the key factor that led to the superconductivity in the TBG, experiments focused on different aspects were done to test different properties of this superconductivity. By analyzing the temperature dependent measurements of different fields in the TBG, using phenomenology designed for phonon-mediated superconductors, the fermi temperature, effective mass of charge carriers and the symmetry of the phonon-electron pairing was obtained. The fermi temperature T_F is traditionally given by.

$$T_F = \frac{m \times v_F^2}{2 \times k_B}, \tag{6}$$

where m is the effective mass, v_F is the Fermi velocity, and k_B is the Boltzman constant. With suitable assumption to the effective mass to be $0.2m_e$, the fermi temperature obtained is 3.7 K. By comparing with the transition temperature, which is around 0.136 K, the material can be regarded as unconventional superconductor. Furthermore, the analysis on field current also indicates the TBG to be a moderately strong coupled two-band superconductor with s- or p-wave symmetries [11]. The analysis in screened Coulomb interaction of TBG reveals that the pairing for superconductivity could be depended on the intravalley phonons and intervalley optical phonons. All these properties suggests that Umklapp processes is closely related to the observed superconductivity [12].

4.4. Furture Chanllenges and Prospects

Nowadays, the efficiency of the current assembly method does not match with the rapid pace of new achievements and findings. Despite chemical synthesis can also produce TBG sample for experiments, most groups are still using the conventional mechanical exfoliation for better quality since high quality sample usually allows researchers to study on other phenomenon like quantum Hall effect. To fabricate such a device, the layering of the graphene instead of merely production is still a big obstacle to the production. Even the most experienced team can only reach a probability of 50% for manufacturing normal TBG samples. For newbie groups the chance of success is usually lower than 10%. In the aspen meeting, a new equipment has been presented. This new amazing device called quantum twisting microscope combines the measuring resolution of scanning tunneling microscopes with the manipulation of atomic force microscopy which enables the adjusting of the twisted angle for stacked graphene after fabrication, maybe be a new solution to the layering.

Typically, in many scientific domains, theoretical predictions and experimental findings can sometimes be at odds or progress at different paces. However, in the case of magic-angle graphene, there's been a tight-knit collaboration between theoretical scientists and experimentalists. Furthermore, the research on magic-angle graphene has managed to stay relatively controversy-free, even though the topic has created a lot of excitement in the scientific community. Jarillo-Herrero credits this to the open nature of graphene research. He mentions his visit to Manchester in 2005 where he learned about the exfoliation technique to isolate graphene. This open sharing of techniques and knowledge might have set a collaborative tone for the community.

Noteworthily, while magic-angle graphene is crucial for those studying carbon materials and superconductivity, its significance goes beyond these groups. The research on this subject has broad implications and has garnered interest from researchers working in a diverse range of fields. While magic-angle graphene is crucial for those studying carbon materials and superconductivity, its significance goes beyond these groups. The research on this subject has broad implications and has garnered interest from researchers working in a diverse range of fields. Especially for those working on 2D materials. Many researchers have now started to find the possible effect brought to material by twisted angle.

5. Conclusion

After the discovery of superconductor, efforts have been made to explain the triggering mechanism. The BCS theory for conventional superconductor is profound and succeeded to draw a great number of researchers to explore the superconductivity and continue to attempt new materials that can break the theoretical limit predicted by the BCS theory, so that it can be one step closer to the actual application. In 1956, the discovery of high temperature superconductor, a type of copper oxide has opened a new gate to the superconductor. More and more cooper-based materials with higher transition temperature and less strict conditions have been found. However, the majority of these materials is still having technique issues in explaining working principle and having a high cost in assembly. The twisted bilayer graphene, discovered in recent years, has once again expand a new field to the research of superconductivity. This new material, despite is still having difficulties in large scale production for high quality samples, has revealed its advantages in many aspects. Compared to superconductors like cooper oxide, the TBG requires no high temperature annealing or similar process, which ensures low cost and relatively higher efficiency and reproducibility in experiments. In addition, the tunable electronic structure, significance beyond caron research and controversy-free environment also proved that this material is undoubtfully a material with great potential.

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