Moiré Physics of One-Dimensional Related Systems and Their Measurement

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Abstract. Currently, the magic-angle graphene has given a tremendous boost to the study of unconventional superconductors. On the other hand, there were still limited experimental studies on superconductivity in one-dimensional (1D) carbon nanotube systems. The study of experimental systems in demonstrating superconductivity was therefore scientifically important. In this review, we have shown strategies toward demonstrating the superconductivity for the single double-wall carbon nanotube (DWCNT). In general, there have been two directions to analyse superconducting properties of one-dimensional materials: (i) strong correlated states (ii) anomalous electron transport operations. We introduced the transmission electron microscope (TEM) and Rayleigh scattering spectroscopy to describe the strong correlation. The theoretical foundations of moiré physics have also been described. Given all the methods, we concluded that the most intuitive way to demonstrate the superconductivity of single double-walled carbon nanotubes is the critical temperature. The sharp drop of the resistance could be directly observed, and the $T_c$ could be obtained from the electrical transport data. In the last section, we also summarized the challenges that need to be addressed in future superconductivity studies of 1D carbon nanotubes.

Keywords. Carbon nanotubes, Magic-angle graphene, Moiré physics, Superconductivity.

1. Introduction
Moiré physics is of central importance in condensed matter physics. However, the absence of a research system poses a significant challenge to understand the moiré physics. Recent research has provided an ideal platform for experimental probing of moiré physics, the magic-angle graphene [1]. The behavior of magic-angle graphene superconductivity is similar with the classical superconducting system of copper oxide, but the former system is simpler to reveal the mechanism of unconventional superconductivity [1]. It is a quite fascinating phenomenon resulting from the control of the mutual angle between two graphene monolayers [2]. Two layers of graphene rotated at an angle of 1.1° form large number of the moiré superlattice structures. The large number of atoms aggregated allows moiré superlattices to function uniformly. By applying an electric field, electrons act collectively to make the conductor-insulator-superconductor transition. The researchers were able to measure precisely how tightly the pairs of electrons were held together. In the two-dimensional moiré superlattice structure, magic-angle graphene can switch different phase states by simply twisting an angle [3-6]. This model provides a new route to study the superconducting system and the Hofstadter butterflies effect [7-8]. It also holds for studying the correlation effect of multi-body physics. Here, we will review the
theoretical foundation and experimental strategies on the superconductivity for the single double-wall carbon nanotube (DWCNT).

2. Theoretical Foundation

2.1. Magic-angle graphene in two-dimensional materials

The metamaterial is a type of man-made material with special properties, like van der Waals heterostructure [1]. The peculiar properties of metamaterials result from their precise geometry and size. Similarly, with the van der Waals forces, the two-dimensional construction blocks stack vertically forms the van der Waals heterostructure. The torsional angle between the layers takes a significant part in the electronic properties. Therefore, the twisted magic-angle in bilayer graphene heterostructures can also result in some abnormal quantum phenomenon. The fractional quantum Hall effect and superconductivity often derive from the high dense states. And one hallmark of achieving the high dense states is having flat bands.

Recent theoretical and experimental studies predict that the existence of flat bands in magic-angle graphene [9-11]. The two-dimensional materials provided another way of achieving flat bands. The moiré pattern was generated by the mismatch between two similar lattices [1]. And it also modulates the interlayer hybridization in twisted layers. Two Dirac cones with the twist angle θ give rise to the mini-Brillouin zone [1] (figure 1(a)). This situation means that each of the two single-layer graphene has its own unique band-gap structure. The resulting moiré pattern forms a new cell and new property. The corresponding positions of these moiré patterns have a relatively high density of states. The Fermi velocity at the Dirac points goes to zero. Also, the flattening of the energy bands near the magic angle can be observed [1] (figure 1(b)). The bandwidth of a flat-band system represents the kinetic energy of the electrons which corresponds to Coulomb interaction. And the flat band exists when the interaction of Coulomb forces is much larger than this kinetic energy [12]. The superconducting state and insulating state can be achieved with the flat band caused by the moiré superlattice structure at certain “magic” angles [13-16]. These two states can be switched simultaneously with some simple ways, like band-gap engineering. The correlations between electrons may help to explain that flat bands exhibit insulating states at half-filling [1]. The half-filling state will be suppressed under a certain magnetic field and then the insulating state becomes conductive. The study of superconductor systems in magic-angle graphene is therefore scientifically important.

2.2. Molar patterns in one-dimensional systems

Although rapid progress has been made in studying the physics of molar patterns in two-dimensional van der Waals heterostructures [17-18], molar patterns experimental studies in 1D systems remain limited. The way to regulate the phase state of one-dimensional material system is still a focal point of research. In analogy with the bilayer graphene heterostructures mentioned earlier, double-wall carbon nanotubes (DWCNTs) provide an ideal platform to describe the one-dimensional moiré superlattice [19]. The Luttinger parameter can help to analyze one-dimensional Mott states in nanotubes from the perspective of electron-electron interactions [19]. The traditional doping method gives rise to the superconducting state of carbon nanotube [20], but it also brings the instability of electron fluctuations. Therefore, the chiral angle can be changed by forming the magic angle of carbon nanotubes to eliminate the instability caused by doping. The magic angle of the carbon nanotubes is a transverse deformation based on the magic angle graphene [21]. The transverse deformation caused by the curved surface combines with the original chiral carbon nanotube produces a new moiré pattern [22-25]. The strong coupling between nanotubes and the changes in electronic band structure is due to the moiré superlattice potential [13]. The relative orientation of the chiral vectors C and C₀ of the internal and external nanotubes basically determines the van der Waals coupling between the internal and external nanotubes in the DWCNT moiré superlattice [6]. There are two main types of superlattice correlation states (figure 1(c)) [2]. The state of strong coupling state happens when the difference between the two chiral vectors C~C₀ (C and C₀ are almost parallel) is parallel to the armchair direction.
And the subband edges’ energy shifts drastically. In figure 1(d), interpenetrating bands are presenting in the band structure. The flat band state because of moiré interference potential occurs when C – C₀ (C and C₀ are almost parallel) is parallel to the zigzag direction [13]. It was expected that the individual zigzag TWCNT would be a band-gap semiconducting. In figure 1(e (a)), it is metallic when the interactions between the tubes are switched on [2]. When the Fermi energy is 0 eV, the electronic band will create a new Dirac cone. Rotating along the axis of the central tube (for avoiding crossing points) will form the large pseudo-gaps (figure 1(e (b))). The coupling between bands is the origin of the pseudo-gap effect [2]. Dirac points in the conduction band and valence band are also opened because of interpenetration. The Dirac cone caused by the strong interaction can produce the transition between metallic and semiconductor states by the twist Angle. However, some edge structures have mixed the armchair and zigzag [13]. And the coupling effect structure will be weak because of the lattice dislocation.

The transition energy shifts because of the electronic coupling between tubes can be predicted based on the knowledge of the matrix elements. The matrix elements can be stated as follows (equation (1)):

\[
M_{\alpha\beta} = \frac{1}{2(\pi N')^2} \sum_{R_j} e^{i(\mu'-\mu)\theta_j + (k'-k)z_j} \sum_{R'_i} e^{i(\mu'\Delta\theta + k'\Delta z)} t_{\sigma\sigma'}(\Delta\theta, \Delta z)
\]  

(1)

Here \(\mu\) is the quantized angular wavevector; the \(N(N')\) is the total number of individual graphene cells on the internal (external) nanotube; the \(k\) is the continuous linear wavevector. The way to estimate the transition energy shifts can be expressed as (equation (2)):

\[
\delta E_{i\beta}^{el} = \sum_{\beta=1}^{3} \frac{|A_{i\beta}|^2}{E_i - E_\beta} = \sum_{\beta=1}^{3} \frac{|A_{i\beta}|^2}{\Delta E_{i\beta}}
\]

(2)

Here \(\delta E\) is the transition energy shift of the nanotube. As clearly seen, the \(1/\Delta E_{i\beta}\) plays a decisive role in the determination of the final energy shift, and its value changes with the inner-wall (outer-wall) diameter, the optical transitions, and chiral angle. Combining the information on \(M_{i\beta}\) and \(\Delta E_{i\beta}\), the transition energy shifts caused by the inter-tube electronic can be estimated.
Figure 1. (a). The difference between the two wavevectors in two layers constitutes the mini-Brillouin zone. (b). Result of interlayer hybridization. (c). One-dimensional (two-dimensional) moiré pattern (DWCNT with similar (different) tube chirality). Copyright 2016 Scientific Reports. (d). Simulations of electronic properties. Copyright 2016 Scientific Reports. (e). Superlattice Dirac point and pseudogaps. (EF = 0 eV) Copyright 2016 Scientific Reports.
2.3. Hamiltonian
Two separate semiconductor carbon nanotubes can form insulating or semiconductor carbon nanotubes, or even double-walled metallic carbon nanotubes (SWCNTs), depending on the resulting moiré superlattice [26]. The torsion and compression of a one-dimensional molar lattice equate to building a Hamiltonian to describe [6]. Any position \( r \) in the middle and lower layers above is offset by mapping \( \delta (r) \), the periodic vector of the molar pattern \( \mathbf{L}_i^M \), and the corresponding reciprocal vector is obtained [6]: \( \mathbf{G}_i^M = (I - M^{-1} R) \mathbf{b}_i \) (\( i = 1, 2 \)). The effective Hamiltonian near each corner of the intra-layer Brillouin region can be used to describe low-energy electrons. The total Hamiltonian is obtained by coupling between layers [6], \( H_\xi = \begin{pmatrix} H_1(k) & U \\ U & H_2(k) \end{pmatrix} \). Assume that the lowest energy bands of decoupled internal and external SWNTs for each band center are expressed [6] as \( E = \pm [m_i^2 + k^2]^{1/2} \) and \( E = \pm [m_0^2 + k^2]^{1/2} \), in the presence of coupling between \( u_0 \) tube [6]. If \( \Delta K_\xi \cong \xi G_4^M \), the following equation further explains the energy-momentum relationship derived from the Hamiltonian model,

\[
E(k) = -u_0 \pm \left\{ [m - m_D(u_0)]^2 + [k + k_D(u_0)]^2 \right\}^{1/2}
\]

and

\[
E(k) = +u_0 \pm \left\{ [m + m_D(u_0)]^2 + [k - k_D(u_0)]^2 \right\}^{1/2}
\]

When \( u_0 > |m| \), the closure gap, the interaction between pipe can be under the condition of strong coupling will bare single-walled carbon nanotubes semiconductor bandgap in the metal band [27]. When the energy gap is larger than \( 2u_0 \), they maintain their semiconducting properties [6]. Based on the model of effective Hamiltonian, we found the experimental data is still limited.

3. Experimental strategies
Here we show some experimental methods that can demonstrate the strongly correlated states and anomalous electron transport operations of one-dimensional materials.

3.1. Strong correlated states
The transmission electron microscope (TEM) and Rayleigh scattering spectroscopy are the common means that are often used to characterize the microscopic structure of nanomaterials, so they can be used to precisely identify the character structure of the single double-wall carbon nanotube (DWCNT). For the transmission electron microscope, it can determine the chiral structure of the double-wall carbon nanotube by nanobeam electron diffraction [13, 28-29]. On the central part of the DWCNT, the broadband light was focused. To probe optical transitions, the laser light needed to be polarized (figure 2(a)). The light which was scattered by the DWCNT needed to be collected then directed to the spectrometer and the CCD camera [21]. Because of the hexagonal lattice of graphitic layers, the diffraction pattern of the double-wall carbon nanotube usually shows some mutually twisted hexagonal patterns [13]. The oscillation profile along the equatorial line in the diffraction pattern is the decisive characteristic to determine DWCNT chirality [30]. A strong correlation occurs when the chirality is almost parallel to the direction of the armchair. For example, two constituent nanotubes in figure 2(b) are almost armchair nanotubes. And the changes along the equatorial line by strong correlation are the superposition of interlayer enhancement.

Rayleigh scattering spectroscopy can determine the chiral structure of the double-wall carbon nanotube by probing electronic transitions. By normalizing the scattered light intensity with incident laser intensity, Rayleigh scattering spectra can be obtained [13, 31-33]. In the spectra, each optical resonance arises from the interband transitions with inner or outer carbon nanotubes. And the nonperturbative intertube coupling raises different electronic transitions. Theoretically, the essential characteristic of van der Waals coupling is the relative orientation of the chiralities. The total number
of spectra will not change in the weak coupling state, but the weak coupling will bring the shift of peak position in Rayleigh scattering spectra. Contrary to the simple sum of two nanotube spectra, the strong correlation state of the double-wall carbon nanotube produces some additional peaks in Rayleigh scattering spectra. For example, the observed transitions in figure 2(c) indicate the extra number of peaks that expressing the drastic change of band structure [13,34].

![Figure 2](image)

**Figure 2.** (a). Schematic diagram of theoretical simulation of equipment. (b). Experimental and simulated electron diffraction pattern. Copyright 2020 Physical review letters. (c). Rayleigh spectrum of DWCNT showing a strong-coupling effect. Copyright 2020 Physical review letters.

### 3.2. Anomalous electron transport operations

#### 3.2.1. Temperature related standards

Here we provide two ways to describe temperature-related standards: the critical temperature (T_c) and differential resistance signature. The critical temperature (T_c) in this article means the temperature at which the resistance drops sharply. The resistance drops can be revealed by the London effect. The DWCNT may become a superconductor when the temperature drops below the T_c. Once it becomes the superconductor, it will repel the magnetic field, known as the Meissner effect. However, if the magnetic field is applied strong enough, it will break the shield and the Meissner effect will fail. In fact, there is a certain penetration width of the magnetic field, which is called the penetration length, λ. When the magnetic field is particularly strong because of the non-local behaviour, it will penetrate the inside. It will stop the dropping in temperature and destroy the superconductivity to some degree. The two-probe device can measure the electrical transport data [35]. And the device needs to be annealed in an ultra-high vacuum. In the image of the temperature dependence of resistance, the resistance drop will largely be suppressed when the magnetic field is larger than the critical point. Because the magnetic field is strong enough to penetrate the shield, the Meissner effect breaks down. It shows that superconductivity is related to temperature and magnetic field. For example, in figure 3(a), the sharp drop of the resistance at the T_c was observed [35]. By analogy, the magnetoresistances (MR) at different temperatures can also be observed. The MR that remained positive usually represents the superconductivity. For example, in figure 3(b), the resistance remains constant at first [35]. The MR remains positive after an obvious turning point. The reluctance will decrease as the temperature increases. The critical magnetic field can be obtained by extrapolating the turning point field to 0 K at each temperature. And the perpendicular critical field is given by \( H_c = 4\sqrt{3}H_c\lambda/d \). \( H_c \) is the magnetic field strength of the vertical field, \( \lambda \) is the penetration depth of the magnetic field, and d is the tube bundle's diameter. Then the critical temperature (T_c) is given by \( \frac{T_c}{T_{c0}} = \left[ 1 - 2.49 \left( \frac{n^2 \lambda^2}{l^2} \right) \right]^{-2} \). Above the T_c, the situation that temperature-dependent MR properties almost disappeared shows the existence of superconductivity.

The differential resistance drops (dV/dI) shows the appearance of a super-current. The type of superconductor for the DWCNT is given by the Ginzburg-Landau parameter. When \( k < 1/2^{1/2} \), it is the type I superconductor. The DWCNT is a superconductor when the magnetic field is weak. When \( k > 1/2^{1/2} \), it is the type II superconductor that has three regions with a transitional mixing region.
The superconductivity occurs as the magnetic field is relatively weak. When the magnetic field exceeds a certain limit, it will be a mixture state. As the magnetic field increases, the superconductivity disappears. The relationship between differential resistance at zero magnetic fields and the bias current during different temperatures can be measured. Like the temperature-dependent of the resistance, the resistance drop centered at zero bias current will appear and gradually vanish with the increase of temperature. For example, in figure 3(c), the vanishment of the resistance drop was observed [35]. Therefore, the superconductivity of single double-walled carbon nanotubes can be demonstrated by the sharp drop of the resistance and the positive MR at Tc.

![Figure 3](image_url)

**Figure 3.** (a). At different magnetic fields, the resistance was the function of temperature. Copyright 2012 Scientific Reports. (b). Magnetoresistances at different temperatures. Copyright 2012 Scientific Reports. (c). At different temperatures, the differential resistance at zero magnetic fields was the function of current. Copyright 2012 Scientific Reports.

### 3.2.2. Magnetic field related standards

The evaluation of magnetic field-related standards is divided into two methods: Zero-field–cooled (ZFC) and field-cooled (FC). Zero-field–cooled was used to measure the change of magnetic moment to temperature during the sample heating process. The carbon nanotubes were cooled to the critical temperature (Tc) and then apply the magnetic field. The field cooling means the relationship between the polarizability and T after adding a magnetic field. The measurements were measured by cooling with the applied field. All the magnetizations were measured with a stable temperature. Temperature-dependent magnetic susceptibilities can be measured with the superconducting quantum interference device (SQUID) magnetometer. And the measurement results of ZFC and FC were related to superconductivity. For example, in figure 4(a), the magnetic susceptibility decreases monotonically as the electric field increases. This phenomenon is quantitatively consistent with the Meissner effect of one-dimensional fluctuation superconductivity [36]. The magnetization in figure 4(b) is linearly
proportional to the field in the low field range. This behavior is expected from a superconductor [37]. Figure 4(c) shows the remnant magnetization (Mr) as a function of temperature [37]. As Mr remains up to 400 K, the bulk superconductivity might persist up to 400 K. Therefore, the superconductivity of single double-walled carbon nanotubes can be proved by the temperature-dependent of the ZFC and FC susceptibility.

![Graph](image)

**Figure 4.** (a) For five magnetic field values, the normalized susceptibility of SWNTs is plotted as a function of temperature. Copyright 2001 Science. (b-c). The sample tubes are assembled into a bundle, and then these bundles are assembled into ropes. (b) The temperature dependence of FC susceptibility. Copyright 2001 Physics. (c). The remnant magnetization as a function of temperature. Copyright 2001 Physics.

3.2.3. **Shubnikov–de Haas oscillations**

The Fermi velocity decreases at a high temperature can demonstrate the existence of the flat band that was related to the superconductivity. The effective mass is inversely proportional to the Fermi velocity, \( m^* = \frac{\hbar^2}{2E_F^2} \). The effective mass of the electron \( (m^*) \) can be extracted by measuring the temperature dependence of the Shubnikov–de Haas oscillations. The Shubnikov–de Haas oscillation frequency can support the existence of gaps at half-filling. And a plausible explanation for the gap is the Coulomb interactions between electrons. The charge conduction will occur when Zeeman energy exceeds the charge gap. Therefore, the transformation of metal and conductor states
can be achieved by adjusting the magnetic field. For example, in figure 5(a), it exhibits an abnormal reduction in capacitance and a strong increase in dissipation [1]. When the device is warmed up, the enhancement in dissipation vanishes. In figure 5(b), it shows that the charge gap was exceeded by Zeeman energy and the charge conduction occurs. As a result, the superconductivity of single double-walled carbon nanotubes can be explained by the Shubnikov–de Haas oscillations demonstrating the existence of the flat band.

Figure 5. (a). Capacitance measurements. Copyright 2018 Nature. (b). When the Zeeman field (B ≠ 0) was applied, the charge gap could be closed when the Zeeman energy was equal to the gap. Copyright 2018 Nature.

4. Summary and perspectives
In summary, combining with the latest magic-angle graphene theory, this paper has extended to the one-dimensional correlation system. It was proved that one-dimensional double-walled carbon nanotubes are also a strongly correlated system. We then presented a detailed overview of the theoretical foundations of molar patterns in one-dimensional systems and the energy-momentum relationship derived from the Hamiltonian model. By theoretical analysis, some experimental methods that could demonstrate the superconductivity for the single double-wall carbon nanotube (DWCNT) have been summarized. The chiral structure could be characterized based on the transmission electron microscope (TEM) and Rayleigh scattering spectroscopy to describe the strong correlation in the carbon nanotube. In addition, in terms of the anomalous electron transport operations, the temperature and magnetic field-related standards helped to demonstrate the superconductivity. Theoretical studies have shown that the phenomenon of a sharp drop in the resistance and the positive magneto-resistances (MR) at Tc could become viable evidence of superconductivity. The temperature-dependent of the ZFC and FC susceptibility could also be a good choice. And the existence of the flat band demonstrated by Shubnikov–de Haas oscillations could help to explain the transformation of metal and conductor states. The discovery and proof of the one-dimensional correlation system were helpful to scientifically and systematically find a better and cleaner platform to explore the electron strong interaction in the one-dimensional system. On the other hand, it also provided an excellent material foundation for exploring the superconductivity of one-dimensional materials or the molar physics of condensed matter.

However, the methods of demonstrating superconductivity listed in this paper were still limited. The intrinsic mechanism of ZFC and FC on superconductivity involved extremely complex processes and the mechanism was not yet well understood. In addition, Shubnikov–de Haas oscillations seem like a good choice, but it requires a high-intensity magnetic field which is difficult to achieve. For more rigorous proof, researchers need to resort to representations at lower temperatures or under more extreme conditions. We believe that by the cooperation between theoreticians and experimentalists, a more efficient way will be available soon.

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