Superconductivity in 2D Transition Metal Dichalcogenide

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1. Abstract

With broken space symmetry and strong quantum fluctuation, 2 dimensional materials in different systems always show distinguished properties from the bulk. Here I investigate the 2D superconductors which involves both strongly correlated and low dimensional system following recent years' development in this field. Unlike superconductivity, more accurate and precise technologies are needed in low dimension transport experiments and magnetic measurements, whose rapid development in recent years gives the way to explore the nano-world. It is the time to combine both. I try to clear up the researching logic used in this topics: the essential basic technology, criterion of the 2D superconducting system, anisotropic magnetic field response and Josephson effect are covered. And I also put forward some possible experiments could be done to explore this field in the end of section 6 and the discussion.

2. Introduction

How to define 2-dimensional materials which are distinguished from bulk systems? Different kind of materials has different criterion, such as semiconductor which should have a character length less than its Bohr radius in one of the dimensions. For metal, we need to compare its thickness with the Fermi wavelength, while in superconductor the criterion is the coherent length of the Cooper pairs. Of course, according to Anderson localization theory 2 dimensional metal corresponding to extended state does not exist in low-dimension system [1], which will be discussed later in the paper. Normally, scientific research follows the route that reducing dimension in the same system makes things different.

Since H.K.Onnes discovered Superconductivity in Mercury in 1911[2][3], people keep finding all kinds of superconductor in different systems of materials. From conventional metal to alloy, complex metal oxide or even covalent materials like carbon nanotubes [4], etc. Naturally, people wanted to see what happened in lower dimension material with starting doing research on the metal film in 1938. [5][6] In recent few years, people turn to the rising 2 dimensional transition metal dichalcogenide(TMD) exfoliated from the bulk materials to realize superconducting, which shows some novel properties and applicable prospect, e.g. the quantum phase transition [7], high critical magnetic field [8]. Besides, the 2-D TMD promises us a transition between 2 dimensional metal/insulator to 2 dimensional superconductors without dimension crossing.

3. Doping to induce superconductivity

Normally, the 2-dimensional materials give us semiconductors not metals because Anderson localization forbid the extended state existing in 1 or 2 dimensional materials. The standard procedure is doping the material to get a high density of carrier to induce superconducting in low temperature just like what people do in high Tc cuprate oxide system. Thus in the picture of quantum phase transition, there will be a criterion carrier density to induce superconducting. According to the experiments and calculations, several papers support that the criterion should be around 10^-14(8*10^-13) cm^-2. [9][10][11][12]

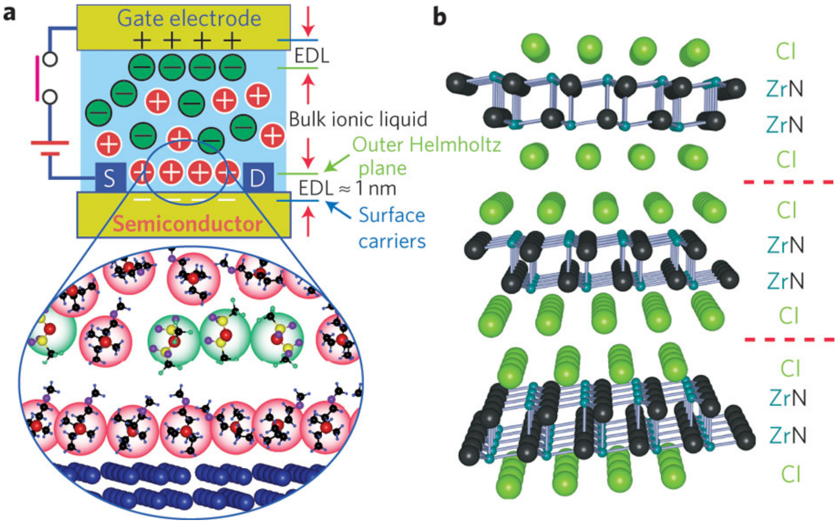

Figure 1 the principle of the Electric double-layer transistors[9]

Electric double-layer transistors

In bulk system, chemical doping is the most widely used to achieve the criterion carrier density. But here are two shortcomings of this method: it will introduce new ingredients into the original system which may cause additional defects, structure deforming or unnecessary complexity of the physical properties[13], etc. The field effect transistor(FET) is also a handy method to increase the carrier density without introduce unintentional disorder. [14] However, the conventional FET doping is not sufficient to achieve the criterion carrier density[ueno2008] thus far from measuring the unabridged region of the superconducting phase diagram. To fix this problem, a new type of FET: electric double-layer transistors (EDLT) was invented [15] and is commonly used in the electrical doping induced superconducting. Now we can use it to achieve the density as high as 10^-15cm^-

2. In traditional way, people use high-κ material(HfO_2) as the back gate to improve the carrier density. In EDLT, the ion liquid can accumulate carriers as 2-order high as conventional FET. [16] Its configuration shows as figure 1. The FET is covered with a droplet of ionic liquid called DEME-TFSI (N,N-diethyl-N- methyl-N-(2-methoxyethyl) ammonium bis (trifluoromethyl-sulfonyl) imide [16] which is normally used in ion gating. Then the Helmholtz electric double layer is formed at the interface, which can be regarded as a capacitor with one sheet of ions in liquid and another sheet of accumulated charges in solid. [12] Several remarkable demonstrations made by electrical doping are to control the critical temperature of superconducting which was shown in the high Tc system [17] and quantum phase transition from superconducting to insulating state in $LaAlO_3/SrTiO_3$ interface. [18] In low temperature during which superconducting works, the ion gate is no longer liquid, thus its quality will reduce slightly in some specific working voltage. [16] Meanwhile, the high-κ material keeps working in the frozen state which guarantee the performance of the EDLT.

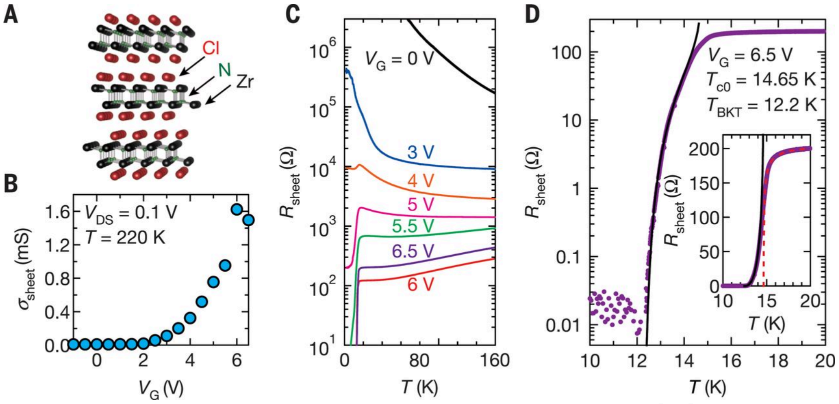

Figure 2 transport properties of ZrNCl with ion gate[11]

4. Phase transition and phase diagram

With the assistant of the ion gating, the carrier density criterion of induced superconducting is easily exceeded. From figure 3 we can clear see that in a specific range of voltage, the transition to superconducting is observed. While in the other range, the insulating state remains whose resistance increases with the decreasing of the temperature corresponding to the semiconducting property. When $V_{LG} \geq$ criterion, the R-T relation shows metallic transporting character which has positive slope before the superconducting transition.

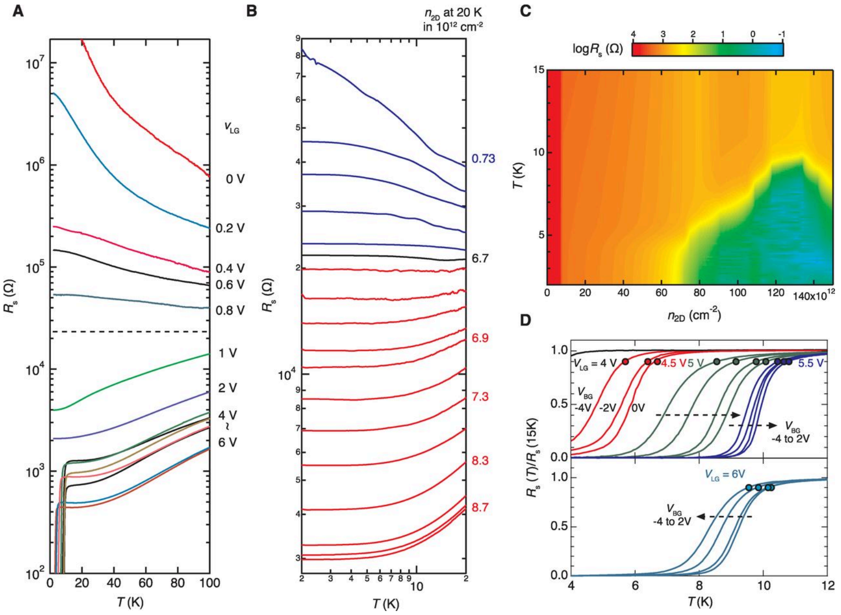

Figure 3 transport properties of thin flake MoS2 and dome phase diagram[10]

Normally, studying in 2D materials using the technique of reducing the film thickness, which inevitably introduces the defects causing disorder. In this kind of system, the direct Superconductor-Insulator transition(SIT) is supposed to be present, which is a kind of quantum phase transition(QPT). [19] Caused by quantum fluctuation, QPT does not require the thermal fluctuation as the thermal phase transition does, which means even in zero temperature, the ground state the QPT can still happen.

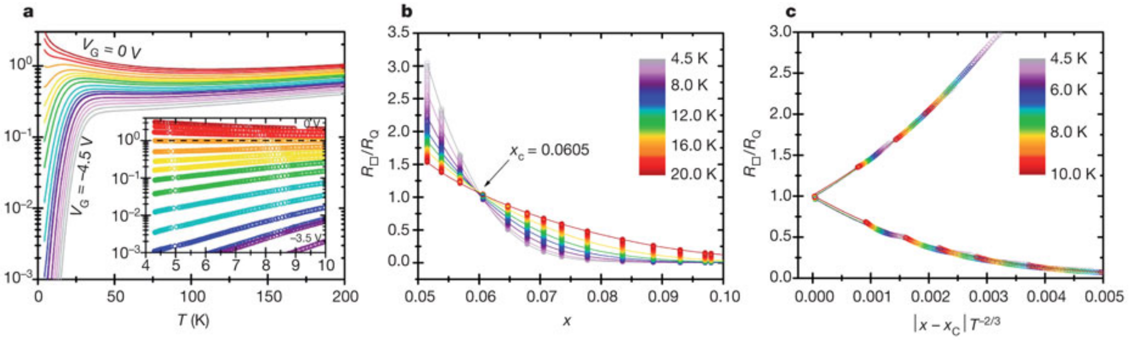

Figure 4 SIT driven by electric field[16]

The mechanism behind the SIT is controversial. There are two different models which are put forward to illustrate the QPT. [7] One is called fermion model, which is the transition is driven by the breaking and condensing of the Cooper pair. In insulating phase, the Cooper pairs are broken thus vanish. The other is bosonic model, saying that the vortex-antivortex pairs are the order parameter of the transition. In both sides insulating or superconducting, the Cooper pairs always exist. But the difference is the vortex-antivortex pairs are strongly pinned in superconducting phase, while in insulating phase, they are broken by quantum fluctuation and weakly pinned thus free to move. So the key feature is to examine whether Cooper pairs still exist before and after SIT. In $La_{2-x}Sr_xCuO_4$ ultrathin film synthesized by EMB, the quantum transition has been detailed discussed. [7] By measuring the quantum resistance of this system, $R_Q=h/(2e)^2$ is obtained in both sides, which convincingly prove that the carriers in both sides consist by two electrons. Thus it more coincides with the bosonic model.

Metallic ground state

Specifically, an intervening metallic phase was observed between the superconducting and insulating phase in a less-ordered 2D material with magnetic field applied. [11] The metallic ground state in 2D materials is an untrivial problem. Since Anderson put forward his localization theory proving that the extended state which corresponding to the metallic diffusion does not exist in 1 or 2-dimensional system. [1] Using thermal activated vortex flow model, the Arrhenius plot in ZnNCl is compared with the experimental data. [11] Below the T_c, the resistance of the sheet is described by

$$R = R'\exp(-\mathrm{U(H)}/k_BT_c)$$

Here the bias voltage is 6.5V and the out-of-plane magnetic field varies from 0.05T to 9T.

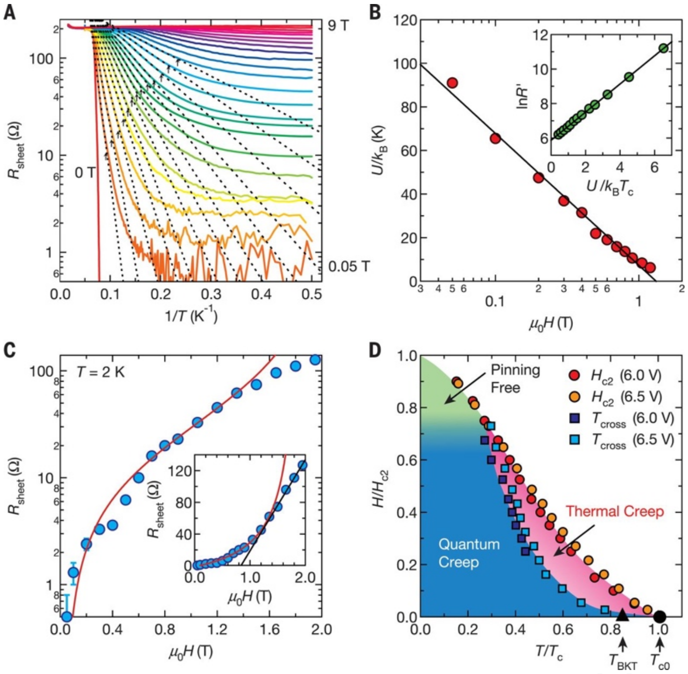

Figure 5 vortex dynamics and the metallic ground state[16]

The experiments align with the activation model well at high temperature region. But at low temperature, the R-T curve deviates from the theoretical dashed line, and tend to a finite resistance at zero temperature, which shows a metallic ground state.

Domed-shape phase diagram

In the phase diagram of the electrical induced superconducting, a domed shape phase is observed in many system [20][21], which is quite a coincident with the phase diagram plotted in the high T_c cuprate superconductor [22][23].

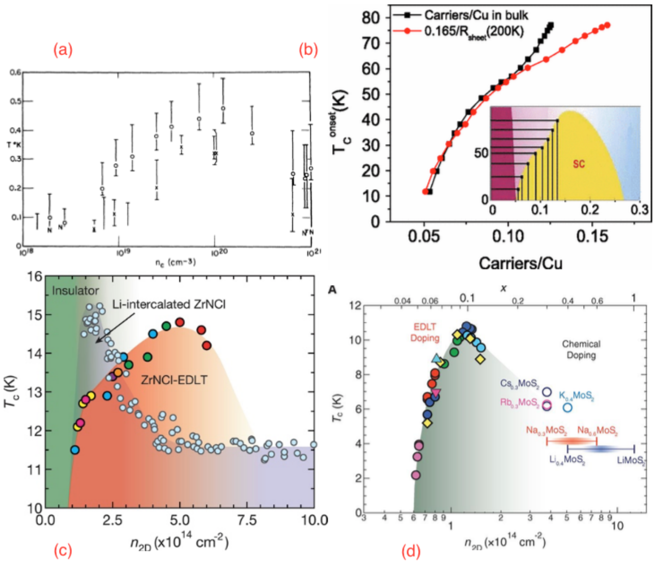

Figure 6 dome-shape phase diagram in different systems. (a) $SrTiO_3$ [21]. (b) Ultrathin YBa2Cu3O7 x Films.[17] (c)ZrNCl thin film.[16] (d) MoS2 thin flakes.[16]

Domed phase diagram in conventional superconductor has a long history. In 1965, domed phase was spotted in $SrTiO_3$ [21], a bulk semiconductor. Chemical doping was used to induce superconductivity. Similarly, the domed shape was also observed in the ultrathin film of $YBa_2Cu_3O_{7-x}$, though it is typical high Tc superconductor. [24] In recent years, domed shape phase diagram also shows in the 2D TMD materials [10][11], which just drives people to think about the mechanism behind it.

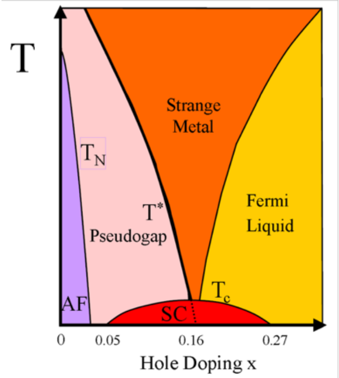

Figure 7 phase diagram of high Tc Superconductivity.[17]

Furthermore, we can ask whether the 2D superconductors follows high Tc superconducting mechanism or just the normal BCS. Basically, the coherent length obtained in this system [11] strongly supports it is a BCS superconductor since it has quite long Pippard coherent length. Meanwhile, there were some papers [11][7] which study on the Superconducting-Insulating transition(SIT) showing that the transition is driven by the vortex-antivortex pair obeying the BKT transition. Therefor it is still not quite clear to judge whether it is BCS superconductor.

5. Magnetic properties

Meissner Effect

Another regular measurement is Meissner effect. However, it is quite difficult to measure the magnetization of such small materials. Thus there is only one measurement of Meissner effect done by two-coil mutual inductance technique yet.[7] By this method, there is no need to do the lithography which may introduce other contaminations and interfere the functioning of the ion gate. However, this paper studied on the thin film grown by molecular beam epitaxy (MBE), which is nearly 1mm in length. For other materials, like exfoliated monolayer TMD, the size of the material is too small to adapt the two-coil mutual inductance technique. The principle of two-coil mutual inductance technique is shown in Fig. We measure the current of coil 2 to calculate the magnetic flux inside. If the sample is only with the size of 10um, then the size of the coil really matters. For in a simple case, provided that the coil 1 is quite large, and the magnetic flux doesn't leak, then we can get the mutual inductance coefficient

$$M_{12} = N_2 \mu \frac{N_1}{L} S$$

S is the area of the section of coils; L is the length of coil 1, and it is large. Thus we find the coefficient is proportional to the density of turns. To measure the exfoliated 2D materials, we need the radius of the cable is extremely small to get a relatively large mutual inductance.

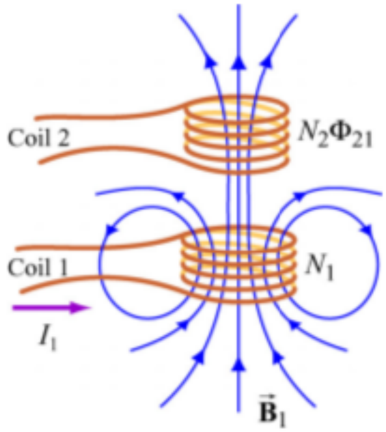

Figure 8 principle of 2 coils mutual inductance.[17]

Despite the difficulty of the measurement on Meissner effect, a lot of measurements on critical magnetic field have been done. Because of the symmetry breaking in the 3 dimensions of real space, its response to the applied magnetic field also shows anisotropic property.

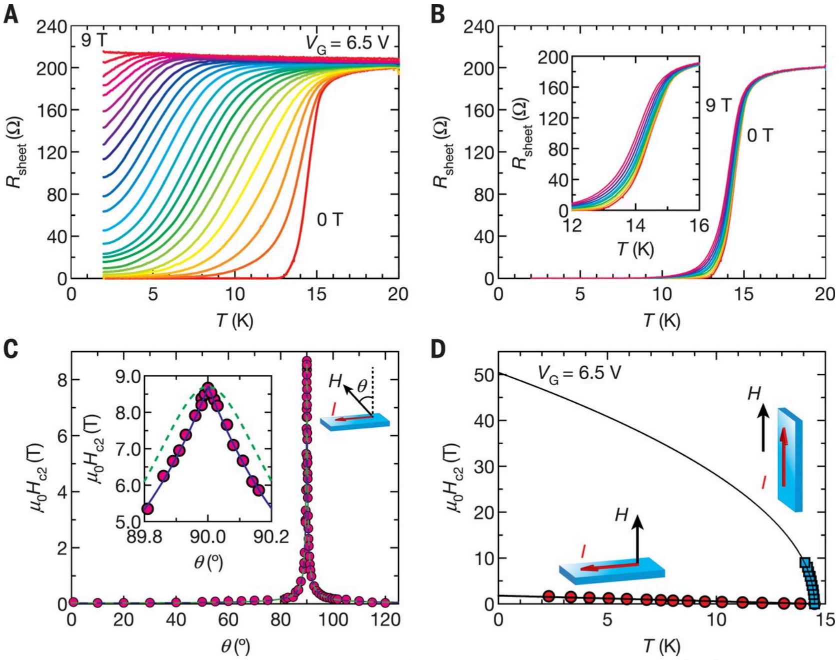

Figure 9 magnetic response of 2D ZrNCl superconductor.[16]

As a type-2 superconductor, people investigate the relation between H_{c2} with the angle between magnetic filed and material plane and temperature. According to 3D Ginzburg-Landau theory, we can calculate the relation between H_{c2} with the angle in the anisotropic material model. However, the measurements' outcomes are in line with the 2D Tinkham formula perfectly, which is a strong proof supporting idea of 2D superconductor.

The mechanism of superconductivity destroying caused by magnetic field is very clear now. Both Pauli paramagnetic and orbital effect work on this. [8] To form the total diamagnetic inside the material, the carriers i.e. Cooper pairs in superconductor flow circularly establishing loop currents to minimize the the magnetic field inside, which strongly relies on the coherent length of the Cooper pairs i.e. orbital effect. And Pauli paramagnetic mechanism arises because magnetic field can destroy the singlet sate Cooper pair by Zeeman effect. Interestingly, there are some papers studying on the triplet state Cooper pair in uranium compound[25][26][27][28], which is beyond my discussion. In 2D materials the orbital mechanism is specifically anisotropic because in in-plane direction the strong magnetic field produce small flux which can be minimized by finite coherent length. This contributes to the high critical magnetic field in the in-plane direction. Thus critical magnetic field mainly rely on the interaction between the magnetic field and the singlet Cooper pairs. [8] However, it still cannot explain why the critical field can exceed the Pauli limit [29][30] so much.

Enhanced critical magnetic field

Now we are heading questions why the critical magnetic field can be enhanced and how large the critical magnetic field can be, which mainly determined by the interaction between the magnetic field and the singlet Cooper pairs. [8] According to a recent paper, 52T criterion magnetic field has been achieved in MoS_2 monolayer [31], which is a quite large field. Several mechanisms to explain how to protect superconducting from high B field has been put forwarded. Spin orbital scattering (SOS) was thought as the reason of the high critical field. [32] The spin-orbital scattering can protect the singlet Cooper pair from the applied magnetic field by spin flipping with the supposing of small scattering time, which means high scattering rate. But in recent papers regard the SOS as a minor factor in 2D superconductors considering high scattering rate does not exist in the bulk material of MoS_2. [8][31] Instead, the spin-valley locking could be the main reason. Applied B field prefer the aligning of the two electrons while the Cooper pair consists of 2 electrons with opposite spins. The SOS make spins randomized to weak the energy splitting of opposite spins caused by applied B field. However, SOS mechanism was overestimated because scattering ratio should be quite high which is not observed in the bulk materials. [8]

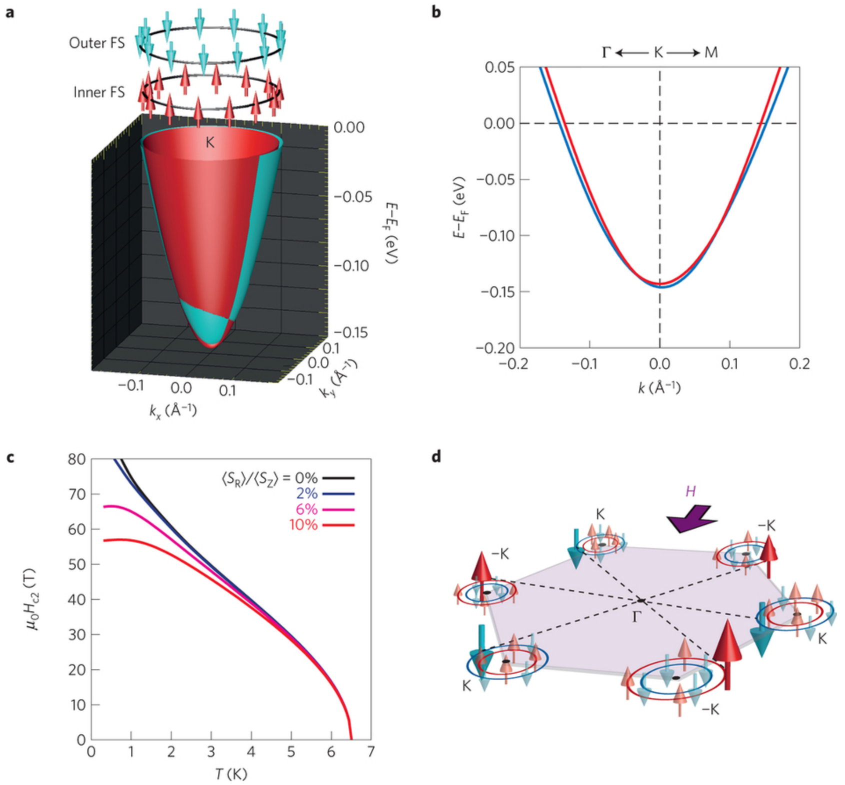

Figure 10 the principle of spin-valley locking in MoS2.[17]

In the 2D materials with inversion symmetry broken the spin-valley locking has been found. [8][31] Because of spin-orbital coupling, the electrons at K point feel effective B field. Thus the energy band splits into 2 bands. The electrons with opposite momentum feel opposite effective B field, then it turns out that the electrons with opposite spin and momentum can couple to form a Cooper pair. So strong coupling with small moment produces large effective magnetic field (B_{eff}~100T) [8] in the out-of-plane direction. Large applied in-plane magnetic field can not destroy the spin-valley locking. Since the spin-valley locking is caused by the breaking of the inversion symmetry, then the 2D materials with inversion symmetry unbroken should have lower criterion of the magnetic field. [8] In 2H-type staking MoS_2, odd-layer MoS_2 is inversion symmetry broken while the even-layer MoS_2 is with inversion symmetry. Thus in bulk system, which can be regarded as the inversion symmetry remained, shows low threshold of the magnetic field. But the experiments on the layer with specific number has not been reported yet, which might be an interesting ending work on the relation between inversion symmetry with the critical magnetic field.

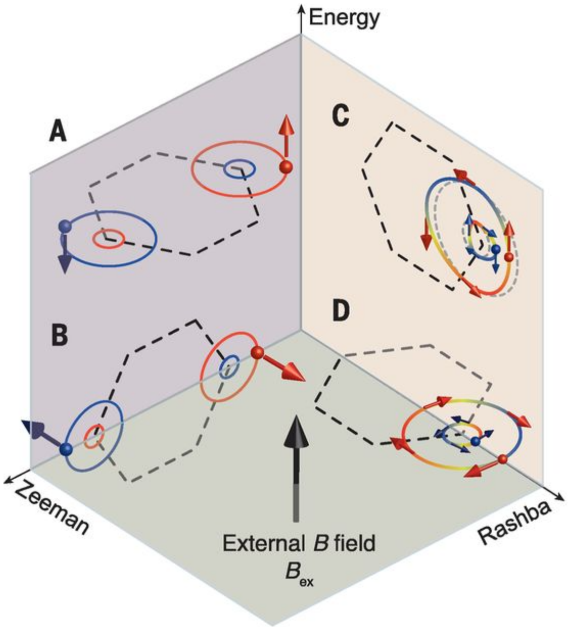

Figure 11 interaction between spins and Zeeman/Rashba type effective magnetic field. [16] It shows that the out-plane B will destroy Zeeman type splitting and protect the Rashba type, while the in-plane B protects Zeeman type splitting but destroy Rashba-type effective field.

Here we can summarize the deduction of the reason of high critical magnetic field. Experimentally, we know the in-plane criterion is quite high while in the out-of-plane direction it is vulnerable. Then we know the applied magnetic field must be perpendicular to the spin direction in Zeeman case, or perpendicular to the orbital plane in the Rashba case. Finally, it is a cooperation effect between these two mechanisms. [8]

6. Josephson Junction in 2D materials

When superconductors are separated by a thin barrier which should be thinner than the coherent length, the Cooper pairs from 2 sides will have coherence. The phase difference of the Cooper pairs from 2 sides will determine the electric response and can be tuned by applied voltage.

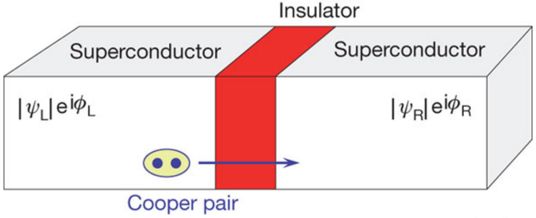

Figure 12 the structure of Josephson junction.[36]

Using Feyman's simple method [37], we can easily get the equation of Josephson Effect:

$$j = j_c \sin\varphi$$
$$\frac{\partial\varphi}{\partial t} = \frac{2eV}{h}$$

j is current density, and j_c is the critical current density. φ is phase difference, and V is the voltage bias.

The beautiful outcome of the simple physics is the current shows tiered response under the time harmonic voltage, which is called Shapiro steps, and the current' response to the perpendicular magnetic field shows similar pattern with Fraunhofer diffraction.[38]

$$\text{Shapiro steps: } V_n = \frac{n\hbar\omega}{2e}$$

$$\text{I-B}_{\text{ext}}\text{ relation: } I(H) = I_c(0)\left|\frac{\sin\left(\frac{\pi\Phi}{\Phi_0}\right)}{\frac{\pi\Phi}{\Phi_0}}\right|$$

In 2D system, the character is in one direction the scale is much smaller than the coherent length, which is the criterion of the 2D superconductors as I discussed. But the question is that since in z direction the thickness is so small, is there still the Josephson effect in a van der Waals system without no interval? Like graphene, the layers are connected by van der Waals interaction, which shows weak coupling between the layers but the distance is much smaller than the coherent length. The answer is yes. The experiment has been done investigating the Josephson Effect in van der Waals (vdW) system, which is made by 2 few-layer $NbSe_2$.[39]

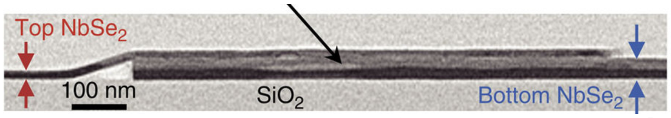

Figure 13 Josephson junction made by 2 few-layer NbSe2 stacking.[39]

There is no inserted material between the 2 few-layers, but the van der Waals gap. As the result, the Shapiro steps and Fraunhofer diffraction like I-B relation are both obtained, which is a direct proof that the $NbSe_2$, as a TMD material is a BCS superconductor.

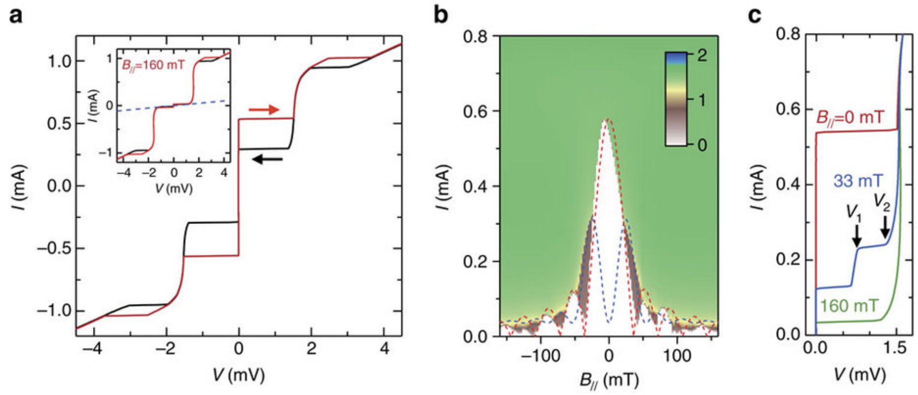

Figure 14 the I-V properties: Shapiro steps and I-B relation.[17]

Feynman's method is based on a simple condition: The wave functions in both sides are different but coherent, and voltage bias create the basis energy difference between 2 sides. So we can see the barrier inserted is essential to have Josephson Effect since the voltage bias can not be built in pure superconductors. In this system, the barrier is gap linked with weak vdW interaction. The experimental result that this vdW gap shows large transparency to the phase coherent transport [39] suggests that the weak vdW interaction can not be neglected even in this scale. Then the interesting question is how the Josephson junction behave in heterostructure. If we use 2 monolayers of different TMD materials to make up a Josephson junction in z direction, how do the character of the materials tune the I-V or I-B response?

A new experiment

Here I try to propose an experiment suggestion on 2D TMD Josephson junction. We can see that when we measure the I-B relation in Josephson junction, the magnetic field should be applied in-plane, which is exact the direction protected by spin-valley locking. Since the spin-valley locking has just been found in I think it is good time to combine both. Use 2 monolayer exfoliated from MoS_2 to make Josephson junction. What I am interested in is not the high critical magnetic value, but when the direction of the Cooper pairs' spins is locked in the direction of out-plane, what the superconducting current responses to the magnetic field? The superconducting current is generated by the magnetic field, so basically the I-B relation just shows the coherent length and the symmetry of the Cooper pairs. But MoS_2 is with strong spin-orbital coupling. Thus some nontrivial phenomenon can be expected from this experiment. Furthermore, the superconducting quantum interference device (SQUID) made by 2D materials shows interesting scenario.

7. Discussion

Superconductivity in 2D system is an induced surface state, which shows many novel properties, such as QPT, metallic ground state, spin-valley locking etc. It has been discussed in the frame of BCS, continuous phase transition theory for years. But there still remain many points unclear.

Relation between thickness and superconductivity

What is the relation between the number of layers and its superconductivity? It was reported [33] that the superconductivity of MoS_2 is weaker in the monolayer accounted by the more important role of quantum fluctuation in the low dimension system. Meanwhile in other researches, the MoS_2, ZrNCl and $Bi_2Sr_2CaCu_2O_{8+x}$ shows similar T_c with their bulk materials, and $La_{2-x}Sr_xCuO_4/La_2CuO_{4+\delta}$ and FeSe thin films even have higher T_c than bulks. As for $KTaO_3$, the bulk does not show superconducting at all. [14] Thus the relation between the thickness and superconductivity is still not quite clear. However, in BCS superconductors, the fluctuation of the lattice should be favored by electron-phonon coupling. Thus I suppose there are several problems in the conclusion made by Constanzo et al. [33] that the thin film's superconductivity is weaker than bulk. When the dimension is reduced, the material is more sensitive to the defects, which will surely affect the superconductivity of the few layer MoS_2. According to the localization theory by Anderson, the low dimension materials are more sensitive to the defects.

Coupling of Cooper pairs by van der Waals interaction

Another paper [8] using thin flakes with the thickness from sub-10 to 50nm to investigate the 2D superconducting. Here the material is not a 2D system, but the induced charges only accumulate in the surface. This is a cleaner system without coupling of heterostructure. Of course it is still a question whether we can use monolayer model in this case though its experimental data follows 2D superconductivity. The Cooper pairs have stronger coupling by van der Waals interaction between the layers than the monolayer with substrate. Therefore, we can suppose the superconductivity in upper layers is limited by the coupling of the lower layers. It may be argued that the van der Waals force is extremely short-ranged force, with the form:

$$\mathrm{V(r)} = \frac{C_1}{r^{12}} - \frac{C_2}{r^6}$$

But if we consider the case of monolayer to few-layer with the thickness of ~nm to ~10nm, the van der Waals still matters.

Besides, in this paper the method of using the EDLT to achieve monolayer superconducting is questioned, [33] since it is not the standard 2D system. The principle is simple, control the charges' accumulating thickness in thin flakes to model the few-layer superconducting. The skeptic thinks it's not a strong proof for the modelling that high current density accumulated on the surface. Therefore, I suppose if we want to verify the accuracy of this modelling, we can use the true mono-, bi-, tri-layer MoS_2 superconductor to compare with the gate controlled thin flakes. Then we can also see the difference between the semi-2D material on the substrate and the semi-2D system induced inside the bulk.

As we all know the van der Waals interaction arises from the election wave function’s dipole-dipole interaction. Moreover, the wave function of singlet Cooper pair is more dipolar than the single electron. Therefore, I suppose the van der Waals interaction in bi-layer MoS_2 will increase when the temperature is decreased during the superconducting phase. Thus I think using AFM [34] to measure the van der Waals energy in few layer MoS_2 should get the relation between the density of the superfluid and the van der Waals energy, which is not difficult to verify.

Besides, another interesting thing mentioned in this paper is the relation between the inversion symmetry and spin valley locking. Thus I propose that the investigation on the superconductivity of 2D materials with specific number of layers are needed. One way we can use monolayer, bilayer, tri-layer on the substrate to verify the spin valley locking in the systems without inversion symmetry. I expect the critical magnetic field shows not strict periodicity with the number of the layers.

Measurement of Meissner effect

The method of measuring Meissner effect is conventionally the 2 coils mutual inductance. But the problem of applying this technique in low dimensional system is that the coil’s size is normally larger than the scale of the materials. Thus my opinion is to find coils which has small size which is relative to the 2D samples.

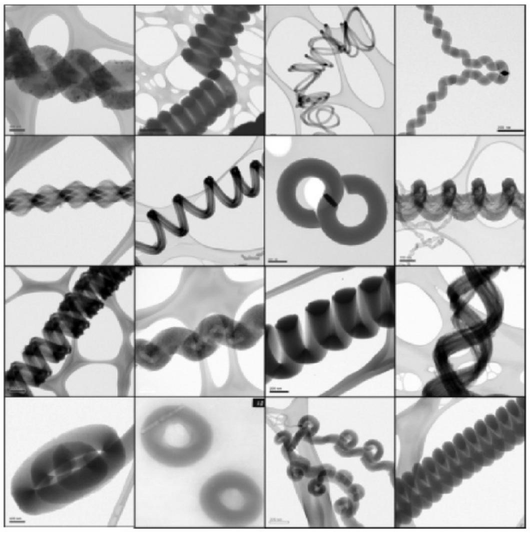

Figure 15 coils made by carbon nanotubes.[17]

The coils made by carbon nanotubes has been studied intensively. But it is still not put into used as the electric coils because of the difficulty of micro-manipulation. But recently a paper on self assembly of nanotubes focused on the nanotubes’ reacting to the applied magnetic field, which is called Teslaphoresis.

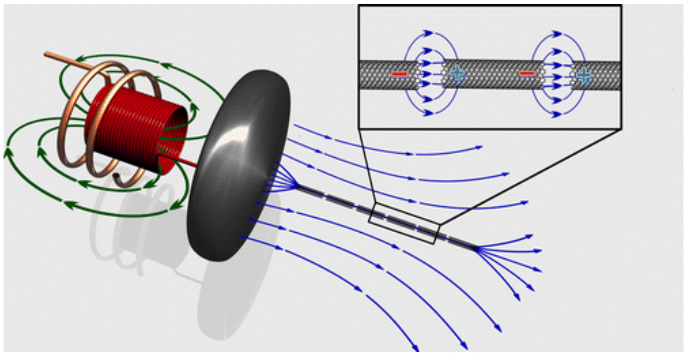

Figure 16 Teslaphoresis [17]

And it shows good electric properties, which is used to self assemble a wire driving the light. Furthermore, it is prepared in around 100μm. The problem of this technique is the uniformity. Even it can be used to carrier the current, but still to generate magnetic field or measure it requires highly uniform coil, which will not cancel the magnetic moments by itself. However, I suppose, it is still provided a possibility to make a mesoscopic coils used for measurement high magnetic field in small area.

8. Acknowledgements

I really appreciate the supervising work given by Prof. Jianting Ye, who spends a lot of time as well as much effort on discussing, answering questions and paper examining.

End

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