Development of high-field STM and its application to the study on magnetically-tuned criticality in Sr$_3$Ru$_2$O$_7$

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Abstract. Scanning tunneling microscopy/spectroscopy (STM/STS) is a powerful method to investigate the spatial variation of electronic states with an atomic resolution. The energy resolution of STM/STS is also very high, better than $\sim$meV, which enable us to study the details of low-lying quasi-particle excitation spectrum. In addition, STM/STS works even in extreme environments e.g. at low temperatures and under high magnetic fields. Taking these advantages into account, we have built a versatile STM/STS system working under combined extreme conditions of ultra-high vacuum, very-low temperature and high magnetic field. We apply this system to the study of strongly correlated electron systems in which subtle interplay among charge, spin and orbital degrees of freedom brings about various electronic phenomena. As one of the examples, we describe the evolution of the tunneling spectrum across the metamagnetic transition in Sr$_3$Ru$_2$O$_7$.

1. Introduction
Since the discovery of high-temperature superconductivity in cuprates, many interesting phenomena have been found in transition metal compounds, for example, colossal magnetoresistance effect in manganites [1] and giant thermopower in cobaltites [2]. These compounds are called as strongly correlated electron systems (SCES), because correlation among electrons due to the Coulomb repulsion plays an important role. A number of novel phenomena of SCES’s are found in doped Mott insulators. The Mott insulator is remarkably different from conventional band insulator. In the case of the band insulator, all the degrees of freedom associated with electrons are inactive because each single-particle energy level is empty or occupied by two electrons with opposite spin. On the contrary, spin (and in some cases orbital) degrees of freedom survive in Mott insulators and charge degrees of freedom become involved if they are doped with carriers. In general there are couplings between different degrees of freedom. Therefore, the electronic properties of SCES are determined by the delicate balance between energy scales associated with the degrees of freedom. As a result, novel electronic states and many electronic phase transitions are observed in SCES. Superstructure [3] and inhomogeneity [4, 5] in the electronic states emerge in some cases. Therefore, real-space investigation of the electronic states is very important in SCES.
Scanning tunneling microscopy/spectroscopy (STM/STS) is a powerful method to tackle this issue because spatial variation of the local density of states (LDOS) can be investigated with atomic resolution. Indeed, STM/STS studies on high-temperature superconductors, which are recognized as typical SCES, have revealed many interesting phenomena [4, 5, 6, 7, 8, 9]. However, most of previous STM/STS studies were performed at fixed conditions and the evolution of the electronic states across the phase transition has never been investigated systematically by STM/STS yet. Variable magnetic-field measurements are especially important because some functions of SCES are related with the criticality near the magnetic transition [1].

In order to examine the detailed electronic states near the magnetic phase transition in SCES, we have developed a versatile scanning tunneling microscope (STM) working under combined extreme conditions of ultra-high vacuum (UHV), very-low temperature (VLT) and high magnetic field (HF). In this article, we describe the details of the microscope design and its performance. We also report on the evolution of the electronic states across the metamagnetic transition in Sr$_3$Ru$_2$O$_7$ as one of the examples of the applications of the newly-developed STM system.

2. General remarks on STM design

Usual STM deals with the tunneling current of 10 pA ∼ 1 nA. In usual cases, this level of current is not difficult to measure if proper grounding is achieved. In many cases, the performance of STM is limited by the mechanical vibration noise. It is important to kill the vibrations coming from outside by pneumatic isolators etc. However, most effective way to reduce the vibration noise is to enhance the rigidity of the STM unit itself. The rigidity can be measured by the mechanical resonant frequency of the STM unit. To enhance the resonant frequency, the mobile part in the unit should be as light and stiff as possible. In many cases, the vibration mode giving lowest resonant frequency is the bending mode of the scanning piezo tube. The resonant frequency of the bending mode is independent of the thickness of the piezo tube in the first approximation and the lateral scan area is inversely proportional to the thickness. Therefore, the scanning piezo tube should be made as thin as possible.

Sample or scanning tip is loaded at the end of the piezo tube. The mass of the load at the end of the piezo tube $m_{load}$ must be small because the resonant frequency $f$ decreases quite rapidly with increasing $m_{load}$ as shown in Fig. 1. It should be noted that even if $m_{load}$ is a half of the mass of the piezo tube, resonant frequency decreases more than 40% from unloaded one. Because thin piezo tube is light, the effect of $m_{load}$ should be taken into account.

**Figure 1.** Calculated change in the mechanical resonant frequency $f$ of the bending mode of the piezo tube as a function of the mass of the load $m_{load}$ at the end. $f$ and $m_{load}$ are normalized by unloaded resonant frequency $f_0$ and the mass of the piezo tube $m_{piezo}$, respectively.
3. Design of low-temperature and high-magnetic-field-compatible STM system

In order to use STM for spectroscopy, operating temperature should be lower than the characteristic energy scale of the phenomenon of interest. In a magnetic field, the most fundamental energy scale is the Zeeman energy. Since Zeeman splitting for free electron is only $57.9 \mu$eV/T, sub-meV energy resolution is required even at 10 T. In order to reduce the thermal broadening below sub-meV range, it is necessary to perform STM/STS below 1 K. For efficient refrigeration and to fit the small bore of the high-field magnet, STM unit should be very small.

Several types of low-temperature STM have been proposed. They are classified according to the coarse approach mechanism. So-called "Beetle type" [10] and "Pan type" [11] designs are commonly used so far. In Beetle type STM, coarse approach is made by three piezo tubes which support the mobile disk on which sample is mounted. Scanning piezo tube is located at the center of the three piezo tubes. Coarse approach is made by slip-stick motion of the mobile disk driven by the three piezo tubes. Beetle type STM is robust against the thermal drift because of its self-compensation design. Moreover, wide area can be investigated because coarse motion along the lateral direction is possible. However, it takes very long time for thermalization because contact between mobile disk and the rest of the STM is very weak. In addition, the resonant frequency of the coarse approach system is always lower than that of the scanning piezo tube because the mobile disk is much heavier than the scanning tip.

The coarse approach mechanism of Pan type STM is a linear motor driven by six shear piezo stacks. Piezo stacks hold a mobile triangular beam containing a scanning piezo tube. The linear motion is initiated by the sequential deformation of the piezo stacks. This type of STM unit is very rigid and easy to thermalize although the thermal drift effect is rather large.

For our UHV-VLT-HF STM, we have adopted Pan-type design by attaching importance to the rapid thermalization and high expected resonant frequency. The original Pan-type design is not equipped with in-situ tip exchange mechanism to eliminate additional mass at the end of the scanning piezo tube. As a result, mechanical resonant frequency is very high but turn-around time of the measurement is inevitably long. We succeeded in designing very small tip holder which can be handled by conventional vacuum manipulators and made it possible to exchange the tip in-situ without sacrificing the total performance. Tip holder is a small Cu-Be piece which can be screwed into the receptacle glued on top of the scanning piezo tube. Dimensions of the piezo tube are 6.5 mm in outer diameter, 0.6 mm in thickness and 19 mm in length. At the lowest attainable temperature of 400 mK, maximum scan area is $1 \mu m \times 1 \mu m$ with $\pm 130$ V high-voltage amplifier. The total mass of the tip-holder assembly on top of the piezo tube is about 0.5 g, giving a lowest resonant frequency of about 5.5 kHz.

Figure 2 shows a schematic illustration and a photograph of the unit. Scanning piezo tube (3) with a tip holder assembly (4) is located inside of a beam (5) which is supported by six shear piezo stacks (6). Two piezo stacks out of six are mounted on a plate loaded by two Cu-Be leaf springs. Rest of the stacks are directly glued on the body (7). (Not shown in Fig. 2.) Reliability of the linear motion is very sensitive to the strength of the leaf spring and the contact surface between the beam and the shear piezo stacks. Sample is placed on a Cu-Be sample stud (2) which is screwed into a Cu-Be receptacle (1). A thermometer and a heater are mounted on the receptacle for temperature monitor and control. Diameter of the unit is about 40 mm. Still there is a margin to reduce the size of the unit.

The choice of the materials is very important for the STM working under combined extreme conditions. All the materials must be UHV compatible (low outgassing and bakeable) and non-magnetic. In addition, in order to reduce the thermal drift, it is important to use materials which have thermal expansion coefficient similar to that of scanning piezo tube. Various engineering ceramics meet this criterion. Among them, we chose AlN as a body (7) material because of its very high thermal conductivity. The mobile beam is made of SiC which is a very hard material.
Figure 2. Schematic illustration and photograph of the STM unit. In the photograph, connection assembly to the $^3$He pot is also shown. 1. Sample stud receptacle. 2. Sample stud. 3. Scanning piezo tube. 4. Tip holder assembly. 5. Mobile triangular beam. 6. Shear piezo stack. 7. Body.

and has relatively high thermal conductivity.

4. Operation of the UHV-VLT-HF STM

The STM unit is designed to be compatible with the unit of the commercial UHV-STM system (Unisoku USM-1300) with $^3$He refrigerator option. The system consists of load-lock chamber, preparation chamber and low temperature insert. The preparation chamber and the low temperature insert are evacuated together all the time by an ion pump. We do not bake the insert but UHV condition of the order of $10^{-10}$ Torr is routinely achieved at the preparation chamber. The vacuum level at the STM unit should be much better because of the cryo-pumping effect.

It is often referred that the needle valve at the He inlet and/or 1 K pot of the refrigerator produces a large vibration noise. After some try and error with the needle valve, we found that everything is quiet at relatively high He flow rate in our system. The noise spectra of tunneling current and tip-sample distance are shown in Fig. 3. The current noise floor as low as $10 \text{ fA}/\sqrt{\text{Hz}}$ is achieved with an $5 \times 10^9 \text{ V/A}$ trans-impedance preamplifier (FEMTO LCA-1K-5G). There are characteristic noises at several frequencies but the noise peak intensity does not exceed $0.5 \text{ pA}/\sqrt{\text{Hz}}$. Accordingly, the distance noise is also very small. The noise floor as low as $10 \text{ fm}/\sqrt{\text{Hz}}$ is achieved.

The lowest attainable temperature measured at the sample stud receptacle is about 400 mK. With 30 litters of $^3$He gas, the base temperature can be kept for more than 50 hours. By using the heater, variable temperature measurements can be performed. The temperature stability better than a few mK is necessary to reduce the noise caused by thermal expansion/contraction. Even during the measurements at elevated temperatures, 1 K pot is always pumped to provide the counter cooling power. Temperature of the 1 K pot increases with increasing sample temperature but continuous operation is possible up to about 60 K. Keeping the 1K pot cool is also helpful
Figure 3. Noise spectra of the STM unit. The current and z-axis distance noise spectra were taken with feed-back loop opened and closed, respectively. Data were taken at sample bias voltage of 400 mV and tunneling current of 8 pA. The sample is Ca$_{2-x}$Na$_x$CuO$_2$Cl$_2$.

to maintain the UHV environment at elevated temperatures.

Strong magnetic field perpendicular to the sample surface can be applied by an 11 T superconducting magnet. At 11 T, the tip shifts about a few nm laterally from the zero-field position and tip-sample distance changes a few tens nm. Since these amounts of shifts are small compared to the maximum scan size, it is easy to scan the exactly same area at any applied field.

The scanning tip is made from a tungsten wire by electro-chemical etching. We have developed a computer-controlled tip-etching system by which very sharp tip can be reproducibly produced without any practices. Before the measurement, atomic arrangement near the tip apex is imaged by field-ion microscope (FIM) attached on the preparation chamber. Furthermore, the atoms near the tip apex can be field evaporated one by one with the FIM. Finally, we can leave only one atom at the apex. Such a well-characterized atomically-sharp tip is very important to obtain highly reproducible data. The sample surfaces are prepared by cleaving either at room temperature or at low temperatures. Surface treatment tools (heating stage etc.) will be installed in near future.

Measured vibration noise amplitude is less than 1 pm and tunneling spectroscopy with energy resolution of the order of 10 µV is possible with conventional lock-in technique. We found that the energy resolution is not limited by electronic noise but by thermal broadening at 400 mK. Therefore, higher resolution may be achieved if the STM unit is combined with a dilution refrigerator.

5. UHV-VLT-HF STM study on Sr$_3$Ru$_2$O$_7$ [12]
We have applied the newly developed STM system to the study of SCES, especially paying attention to the electronic evolution near the magnetic transition. We have chosen Sr$_3$Ru$_2$O$_7$ as a sample because of its unique intrinsic critical nature. Sr$_3$Ru$_2$O$_7$ is one of the Ruddlesden-Popper series ruthenates whose general chemical formula is given by Sr$_{n+1}$Ru$_n$O$_{3n+1}$. The $n = 1$ compound Sr$_2$RuO$_4$ is a 2-dimensional paramagnetic metal which shows spin-triplet superconductivity below 1.5 K [13]. The other end member SrRuO$_3$ ($n = \infty$) is a 3-dimensional
ferromagnetic metal. Sr$_3$Ru$_2$O$_7$ ($n = 2$) has an intermediate dimensionality and is expected to have a character between the two. Indeed, Sr$_3$Ru$_2$O$_7$ is a enhanced paramagnet very close to the magnetic instability. Small uniaxial pressure drives the system into ferromagnetic phase [14] and an antiferromagnetic insulating phase is induced by small amount of impurity doping [15]. Another interesting feature is a metamagnetic transition at around 8 T for applied magnetic field $H \parallel [001]$ and 5.5 T for $H \perp [001]$. Especially, novel field-tuned quantum-critical behavior has been found near the metamagnetic transition for $H \parallel [001]$ [16]. We have performed STM/STS on Sr$_3$Ru$_2$O$_7$ to make clear the change in LDOS near the magnetically-tuned quantum critical point.

Crystal structure of Sr$_3$Ru$_2$O$_7$ is shown in Fig. 4(a). The structure can be viewed as a stacking of a double layer of RuO$_6$ octahedra along [001] axis. Since the crystal cleaves between the double layers, clean and flat surfaces which are necessary for STM/STS are easily obtained.

An STM topograph of the cleaved surface of Sr$_3$Ru$_2$O$_7$ taken at 400 mK is shown in Fig. 4(b). No image processing is made except the plane subtraction. Line profile of the topograph shown in Fig. 4(c) indicates that the amplitude of the noise is smaller than 1 pm.

In Fig. 4(b), square atomic lattice is clearly imaged with several single atomic defects. The vicinity of the defects are imaged as dark, which indicates that the defect alters the electronic states of the surrounding area. Lattice constant of the observed square lattice is 0.4 nm being consistent with the Ru - Ru (apical O - apical O) or Sr - Sr distance. At this stage, it is not clear which atomic site is imaged as a bright protrusion. Weak c(2×2) superstructure, which appear in Fig. 4(c) as an alternate height modulation, is observed. This superstructure becomes clear

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**Figure 4.** (a) Crystal structure of Sr$_3$Ru$_2$O$_7$. Spheres denote Sr atoms. Ru and O atoms are located at the center and apexes of octahedra, respectively. Dotted arrows indicate cleavage planes. Broken line denotes the unit cell. (b) STM topograph of the cleaved surface of Sr$_3$Ru$_2$O$_7$ taken at 400 mK. Sample bias voltage and tunneling current were +50 mV and 50 pA, respectively. (c) Line profile taken along the line shown in (b).
when the STM topograph is taken at a sample bias voltage below a few tens mV. Since there is an alternate rotation of RuO$_{6}$ octahedra around the [001] axis [17], the surface crystal structure has a c(2×2) symmetry. However, even in the presence of the rotation, all the octahedra are electronically equivalent. Since the superstructure is strongly bias-voltage dependent, we infer that observed c(2×2) superstructure is not caused by the atomic displacement but is electronic in origin. Since such a superstructure has not been reported by bulk probes such as x-ray diffraction, it may be an electronic order induced at the surface. Similar c(2×2) superstructure has been observed in the single-layer (n=1) compound Sr$_{2}$RuO$_{4}$ [18]. However, in the case of Sr$_{2}$RuO$_{4}$, the superstructure is clearly imaged even at very high bias voltage of -750 mV, which suggests that atomic displacement plays a role. Therefore, the superstructures in the two ruthenates may be different in origin.

Next we performed spectroscopic measurements. Zero-field tunneling spectrum $dI/dV$ (∝ LDOS) taken on the cleaved surface is shown in Fig. 5(a). There is a suppression of LDOS or "pseudogap" below a few tens mV. Since c(2×2) superstructure mentioned above becomes clear below this energy scale, we believe that the pseudogap is related to the superstructure formation. At the bottom of the pseudogap, two LDOS peaks are observed around ±4 mV. The width of the peaks is very narrow, only a few meV. Such narrow peaks are difficult to be explained in terms of the conventional band picture.

We performed spectroscopic measurements at different locations and found that overall features shown in Fig. 5(a) are commonly observed over the surface although there is a measurable small change in the spectrum from site to site [19].

In order to relate the observed LDOS spectrum to the metamagnetic instability, we examined the magnetic field dependence of LDOS. Figure. 5(b) shows a evolution of the tunneling spectrum up to 11 T. All the spectra are taken at the atomically same position. Therefore, detailed field effect can be discussed. With increasing magnetic field, the spectral weights of the peaks at -4 mV and +4 mV are transferred to the sub-gap features at -1 mV and +2 mV, respectively. Above the metamagnetic transition field of 8 T, LDOS near the Fermi energy suddenly increases.

**Figure 5.** (a) Tunneling spectrum of Sr$_{3}$Ru$_{2}$O$_{7}$ taken at 560 mK. Sample bias voltage and a tunneling current were +50 mV and 50 pA, respectively. (b) Magnetic-field dependence of the tunneling spectra near the Fermi energy. Sample bias voltage and a tunneling current were +7 mV and 50 pA, respectively.
The coincidence of the metamagnetic transition and the onset of increase in LDOS at the Fermi energy strongly suggests that the observed narrow peaks near the Fermi level are responsible for the metamagnetic transition. The evolution of the spectra is apparently different from the Zeeman splitting of the uncorrelated band structure because no splitting is observed and a energy range in which spectral weight transfer occurs is much wider than that expected from the Zeeman effect. Together with the very narrow width of the LDOS peaks, it is most plausible that many-body effect plays a role for the formation of the LDOS peaks and metamagnetism.

In conclusion, we have built an STM/STS system which works under combined extreme conditions of ultra-high vacuum, very-low temperature and high magnetic field. The newly-developed STM unit has high mechanical resonant frequency of 5.5 kHz and is capable of the in-situ tip/sample exchange. The measured vibration noise amplitude is smaller than 1 pm and a spectroscopic measurements with energy resolution of the order of 10 µV is possible. We applied this system to the study of metamagnetic transition in strongly-correlated metal Sr₃Ru₂O₇. We found two sharp LDOS peaks below and above the Fermi energy and revealed that LDOS between the two peaks increases drastically above the metamagnetic transition field. Our results demonstrate that high-field STM/STS provides rich information which can not be obtained by other probes and is useful for the research of strongly correlated electron systems.

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