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Four-probe scanning tunnelling microscope with atomic resolution for electrical and electro-optical property measurements of nanosystems*

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We demonstrate a special four-probe scanning tunnelling microscope (STM) system in ultrahigh vacuum (UHV), which can provide coarse positioning for every probe independently with the help of scanning electron microscope (SEM) and fine positioning down to nanometre using the STM technology. The system allows conductivity measurement by means of a four-point probe method, which can draw out more accurate electron transport characteristics in nanostructures, and provides easy manipulation of low dimension materials. All measurements can be performed in variable temperature (from 30K to 500K), magnetic field (from 0 to 0.1T), and different gas environments. Simultaneously, the cathodoluminescence (CL) spectrum can be achieved through an optical subsystem. Test measurements using some nanowire samples show that this system is a powerful tool in exploring electron transport characteristics and spectra in nanoscale physics.

Keywords: four-probe STM, nanodevice, electrical measurement, manipulation, CL
PACC: 0750, 0660S, 7860H, 0600

1. Introduction

Considerable research efforts are focused on the development of low dimension nanostructured materials, due to their potential application as building blocks for nanocircuits, nanodevices and nano-optoelectronic sensors.[1−4] Simultaneously, great interest in nanoscale science and technology is focused on manipulating nanoscale materials even down to atoms or molecules and measuring the fantastic electron transport, optical and mechanical properties. Conductivity measurement by means of a four-point probe has been used in various materials for decades because of the strong suppression of contact resistance.[5,6] At the same time, four independent probes allow electrical measurement with both collinear configuration and van der Pauw configuration in which the probes are placed in square arrangement to detect the anisotropy and isotropy in conductivity of material surfaces.[7] However, the conventional four-point probe measurement in macroscale has a vital drawback of limited sensitivity in nanoscale due to the comparatively large spacing between the four points.

Scanning electron microscopes (SEM) and scanning tunnelling microscopes (STM) are essential tools chosen in the development of nanoscience and technology for high spatial resolution. Utilizing SEM and STM technology, plenty of efforts are made to reduce the distance between probes in order to improve measurement sensitivity. In recent years, several groups have reported the development of multi-probe instruments associated with SEM or STM.[8−10] One has developed linear micro-four-point probes on a single silicon chip with about 2μm electrode spacing by a conventional microfabrication technique, but the probes

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cannot be driven independently.[8] The electrode spacing of micro-four-point probes is remarkably smaller than the one of macro-four-point probes, but it is not enough for the endless pursuit of optimal local electrical measurement. Other groups reported that they had constructed multiple independently driven probes for electrical measurement in ultrahigh vacuum (UHV) associated with SEM or STM,[9,10] but the complexity of such a four-probe STM in which every probe can be independently driven usually lowers the spatial resolution far from subatomic level.

In order to perform manipulation and measurement of electron transport and optical properties on a small scale ranging down to nanometres, we developed a special UHV system composed of four-probe STM, high resolution SEM, spectrograph and molecular beam epitaxy (MBE) subsystem. Since our system is optimized to minimize the outside disturbance, each of the four tips in our system can work with the standard STM model to provide atomic-scale resolution and be controlled individually and accurately with variable spacing, which gives us an opportunity to manipulate nanoscale objects, even to manufacture artificial nanostructure conveniently. In this case, small probe spacing just limited by the probe radius can be achieved by means of coarse positioning under SEM and fine positioning using STM technology since tungsten tips with a radius of less than 10 nm have been prepared by a DC etching method.[11] Different materials, such as gold, silver and aluminium can be etched as tips for different work function.[12–14] The system also features the spectrum analysis of cathode luminescence (CL) as well as sample preparation with MBE technology. Both the sample and probes could be prepared or cleaned in the UHV environment before measurement.

2. Composition of the system

The four-tip STM nanoprobe system is shown schematically in Fig.1. The main part is the central UHV chamber for the four-probe STM stage with SEM, transferable magnet and a gas station controlled by leak valve. The UHV chambers for MBE and a fast entry lock (FEL) are connected with the central UHV chamber through gate valves in order that the sample and tips can be prepared, cleaned and exchanged without leaving UHV. The CL subsystem and electrical measurement subsystem perform spectrum and electrical analysis. Air springs are installed to reduce the mechanical noise of the whole system.

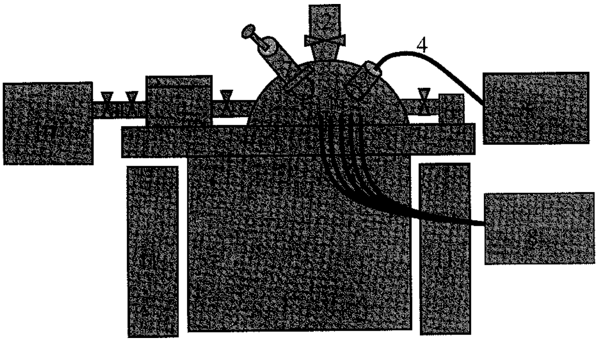

Fig.1. Overview of the system. (1) central UHV chamber for four-probe STM, (2) SEM, (3) collector lens for CL, (4) fibre, (5) the spectrograph, (6) leak valve and gas station, (7) column with transferable magnet, (8) electrical measurement subsystem, (9) fast entry lock, (10) UHV MBE chamber, (11) air springs and (12) system bench.

The central UHV chamber for nanoprobe stage (Omicron) is the core of our system. Figure 2 shows an inside view of the central UHV chamber in our system. From the picture, you can clearly see that the stage is equipped with four independent STM probe stations. Standard UHV STM works without interference because every probe is driven to allow fine positioning in the x, y, and z directions with ultimate accuracy by independent piezo-motors and current control units. It is important that all four probes can employ STM technique to provide clear STM imaging down to atomic scale in order to minimize the interference from the environment. More specifically, the four independent STM probes provide not only the conventional working methods under the control of a feedback circuit but also a completely manual mode in which the movement resolution is 2 nm in the x and y directions and 1 nm in the z direction, enabling each of the four tips to be easily manipulated and manufactured in nanoscale. (We will show the results later.) The navigation of each probe on a sample surface is designed to proceed in two steps. In the first step, probes hop toward the object using slip/stick effects in the x, y, z ranges of 10 mm, 10 mm, and 5 mm respectively, which is for coarse positioning. Secondly, the probes are located at the accurate position using STM technology. The tip held by little magnet on each probe module can be easily exchanged for another in UHV environment using a wobble-stick. A carousel, which can hold ten tips or samples, is placed beside the nanoprobe stage for sample and tip exchange. In Fig.2 the four tips are gold ones for better electrical contact with the nanoscale sample and devices because of its comparatively low work function, moreover, the tips etched with tungsten, silver and other materials can be conveniently exchanged in our experiment.

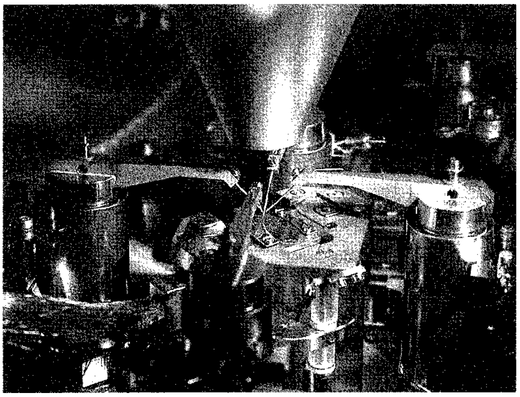

Fig.2. Four independent nanoprobes equipped in the central UHV chamber. The cone over the tips is SEM.

A high resolution SEM (FEI—Model 2LE-Column) in UHV is equipped on the top of the nanoprobe stage to enable tip navigation on complex structures for quick positioning. The cone shown in Fig.2 is just the bottom of SEM that leaves a relatively large working distance (more than 25 mm). At typical imaging condition (1 nA probe current, 25 mm working distance and 25kV beam voltage), the smallest distance that our SEM can distinguish is about 20 nm. The samples can be easily exchanged without affecting the vacuum of the SEM chamber because an isolation valve option separates it from central UHV chamber. Simultaneously, the electron beam of the SEM can be used as the induced source of the CL subsystem. As shown in Fig.1, the cathode luminescence induced by electrons with the energy of several tens of keV is focused by collector lens fixed outside the viewport of central UHV chamber. Quartz is chosen as the material of both the viewport and lens because of its high transmission coefficient especially for ultraviolet. The spectra are analysed by a spectrograph (Acton—model Inspectrum-300-122B), which can cover the range from 200 nm to 900 nm with resolution of 0.2 nm, connected to collector lens by a one-metre-long fibre. The temperature of the charge-coupled device (CCD) detector in the spectrograph is set to -20 degrees C in order to reduce the noise. Since the electron beam is focused on a single nanostructure, the spectra of that can be recorded by our CL subsystem *in situ*. Except for CL spectra, photoluminescence (PL) spectra and electroluminescence (EL) spectra can also be researched in our system after a slight alteration in induced source.

The nanoprobe stage allows measurement and application of both voltage and current with up to four probe modules. We chose Semiconductor Characterization System 4200 (Keithley—model SCS-4200) as our electrical measurement subsystem, which is connected to the four tips with low-noise cables. Three independent source-measure units (SMU) in SCS-4200 and the common ground terminal allows electrical measurements with the four-point probe method. It should be noticed here that the four-point probe method gives us correct resistances irrespective of Ohmic or non-Ohmic contacts at the probe contacts. The voltage supplied by every SMU is in the range from −20V to 20V with the resolution of 2μV. The current resolution of SMU is as high as 1fA. In our experiment, the electrical noise of the whole system of four-probe STM is less than 30 fA while applying one-volt voltage. It is obvious that system composed of nanoprobe stage and SCS-4200 offers us an ideal environment for electrical measurement. In addition, the voltage on the sample platform can be applied controllably, which gives us more choices to fabricate and study the characteristics of nanodevices, for instance, field-effect transistor.

A transferable magnet that can provide a magnetic field of about 0.1T is installed in central UHV chamber. The column with magnet is separated from the central UHV chamber by an isolation valve. It is necessary that we choose Mu-metal as the material of the column to avoid unwanted effects on SEM from the magnetic field. When we want to apply an appropriate magnetic field in our experiment, it is easy to transfer the magnet into the central UHV chamber and the intensity of the magnetic field can be approximately controlled by the distance between the magnet and sample. Also, a variable leak valve (Varian—model 9515106) is equipped with a central UHV chamber to give us accurate controllability of the pure gas pressure such as oxygen, nitrogen and so on. The minimum controlled leak rate is 10^{-9} torr·l/sec.

The chamber of FEL is connected to the central UHV chamber by the transfer tube and gate valve. Another carousel, which can be loaded with up to ten samples or probes, and one wobblestick is equipped in FEL to transfer samples and probes into the central UHV chamber. A heater, which provides resistive heating and direct current heating, is installed in the chamber of FEL. The maximum temperature allowed in the former mode is 1170K, and the latter is about up to 1400K depended on the flowing current (maximum heating current 5A). It is highly advantageous to clean the tungsten tip by means of taking a tip to approach silicon stick heated by direct current with wobblestick and evaporating impurity that is attached to the tungsten tip during etching, also the possible tungsten oxides.

The standard MBE chamber is equipped with evaporators to deposit metal and organic thin films. A quartz crystal microbalance (Sycon—model STM-100) is used to monitor thin film growth. Several surface science tools, including a reflection high energy electron diffraction (RHEED), a molecular beam deposition associated with low energy electron diffraction (MBD-LEED, Kentax), an ion sputter gun and a gas dosing system, are equipped with MBE chamber. There are two gate valves between the MBE chamber and FEL, so the UHV chamber for MBE could be dismantled in order to avoid the mechanical noise after a sample is completed and transferred into FEL.

In our system, the temperature of the sample platform in the central UHV chamber is able to vary from 30K to 500K under the control of liquid helium flowing and solid-state heating element below the stage. If liquid nitrogen is employed, the ultimate temperature is about 120K. A temperature controller (Lakeshore—model 331) provides temperature measurement and adjustment.

In order to achieve a high-resolution STM/SEM image and accurate electrical measurement of weak current, both the mechanical and electrical noise have to be reduced as far as possible. Therefore, we chose air springs (Integrated dynamics engineering—model PD301 H) with a resonance frequency of 1Hz as our external passive vibration isolation system that can effectively absorb vibrations with a frequency above its resonance frequency. The bench holding the whole system is suspended by air springs. All cables from the bench of the vacuum system are supported in a U-like bend to keep no contact to floor, which is good practice to prevent mechanical noise pick-up.

3. Experimental results

We have report that there are four independent STM probes in the system. Connected to four control units respectively, each probe can scan the surface simultaneously with outstanding resolution performance by the help of air springs. As shown in Fig.3, we get atomic scale resolution image of highly oriented pyrolytic graphite (HOPG) surface. The STM tip is tungsten, and every tip of the four can provide the ability reproducibly.

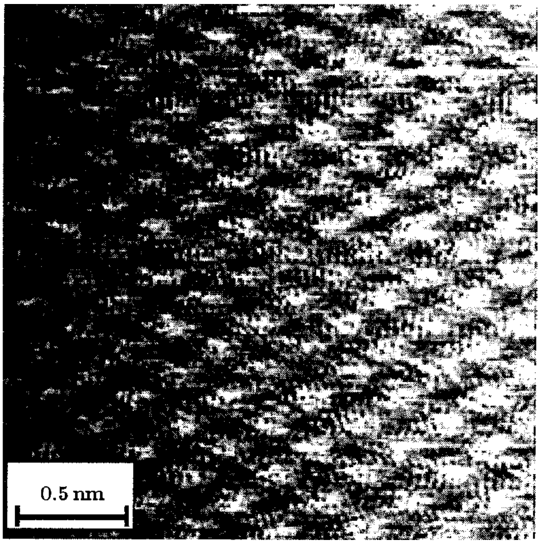

Fig.3. STM image of HOPG acquired by one of the four probes. $I_{\text{setpoint}}=500\,\text{pA}$, $V_{\text{gap}}=70\,\text{mV}$.

It is possible for us to position probes on some 1D or 2D nanostructures with STM technique, which can provide higher resolution than SEM does. We put a sample dispersed with Boron nanowires into the UHV chamber and scanned it in STM mode. As shown in Fig.4, crossed nanowires were found. Then we focused on the upper left corner of the cross (Fig.4(b)). In this case, we can position the STM tip on an appointed place. Moreover, in the same way we can position the other tips on different branches of the cross, so it provides a smart approach to do some I-V measurements to investigate the properties of the nanowires and/or the junction.

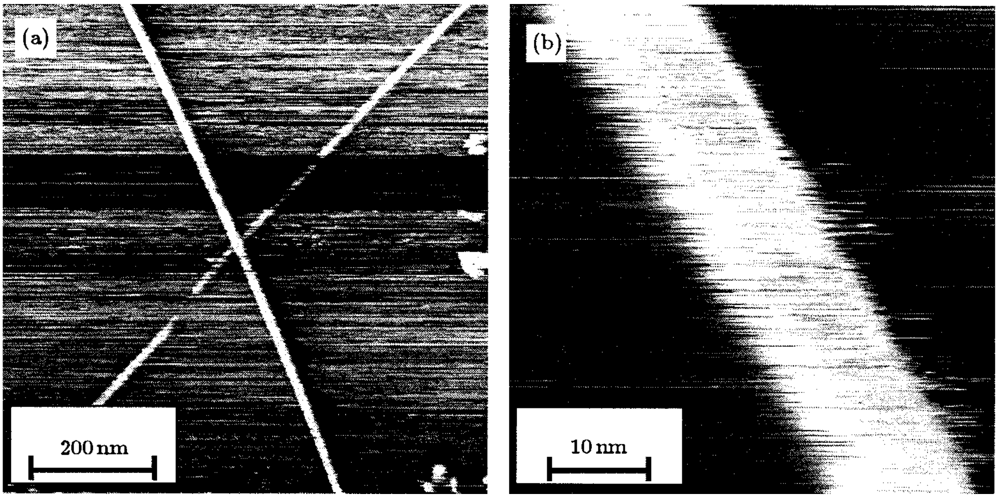

Fig.4. STM image of Boron nanowires. (a) $I_{\rm setpoint}$=500 pA, $V_{\rm gap}$=300 mV. (b) The image is the upper left corner of the (a), $I_{\rm setpoint}$ and $V_{\rm gap}$ are the same as (a).

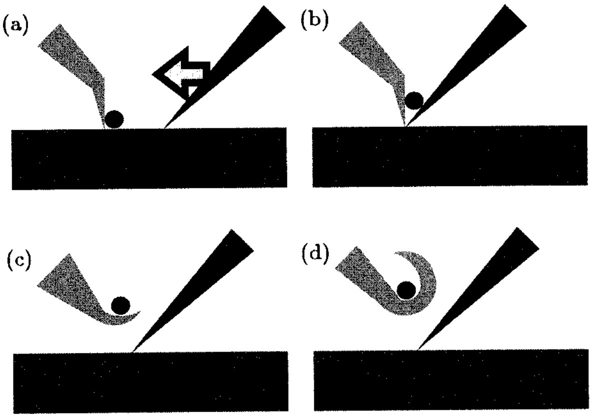

Fig.5. Process of hooking a nanorod. (a), (c), and (d) show A- type, B-type, and C-type hooks respectively.

To manipulate and manufacture things in the nanometre scale, even at the scale of atoms and molecules, is the ultimate goal of nanoscale science and technology. The outstanding maneuverability of our system makes it possible to manipulate or manufacture some low dimension nanostructures. While one probe is pushed against the substrate or against another probe controllably and gingerly with the navigation of SEM, a hook, which has a special shape, may be formed at the top of the probe. Figure 5 shows three types of hooks (A, B, C) and the process of hooking a nanorod. At first, we use A-type hook to stop the nanorod moving. Then we use a normal probe to push the nanorod in order to cause the nanorod to go upwards. Simultaneously use another probe with a B-type hook to hook the nanorod and lift the nanorod. Finally, considering the easily dropping from B-type hook, we use a C-type hook to hook the nanorod firmly. In this way, we can suspend a nanorod with the help of several probes, and move the nanorod to any place we want. We can even move or exchange the substrate while the nanorod is suspended.

What we have demonstrated is an unabridged process of hooking a nanorod. Usually, under some conditions, we can abridge some steps in the process. As shown in Fig.6, there are two nanorods and one is crossed over the other. We can directly use a C-type probe to hook the nanorod. Figure 6(c) clearly shows the nanorod is hooked firmly. Figure 6(d) shows one nanorod is suspended from four probes in another experiment. We can do some four-terminal or two-terminal *I-V* measurements during suspension, eliminating the influence of substrate that includes leak current, capacitance, and so on.

Figure 7 shows a perylene nanostick being suspended with two probes while measuring its electrical characteristics. It is clearly shown that the leak current is about 30 fA when voltage is one volt. The very low leak current and noise should be attributed to the good shielding of the system and the cable. The vacuum chamber made of stainless steel is just like a screen shielding the nanostructure and probes from electrostatic or magnetic interference. Triax cable and connector provide very high leak resistance and very low cable-capacitance.

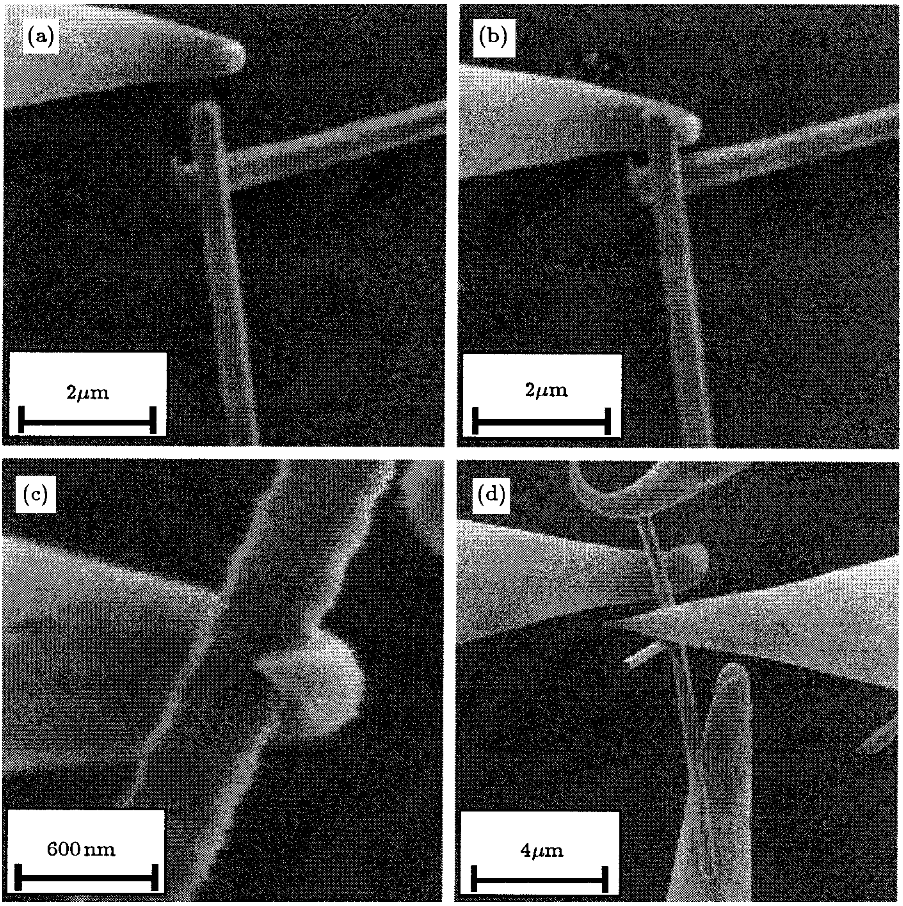

Fig.6. (a) to (c) The probe with a hook at the end was hooking a nanostick. (d) A nanostick was suspended with four probes.

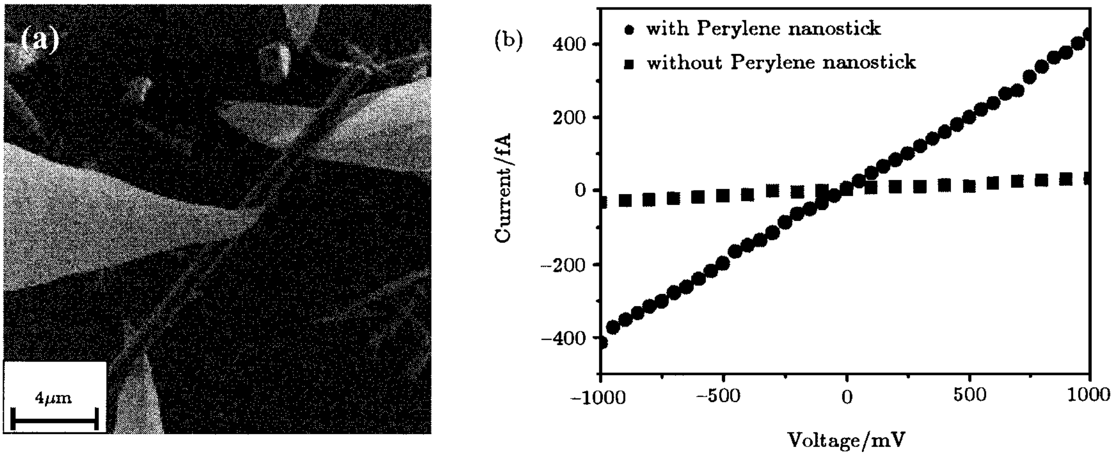

Fig.7. (a) A perylene nanostick was suspended with two probes. (b) I-V curve (●) of the perylene nanostick measured by the two probes; I-V curve (■) of the leak current.

Electrical measurements can be carried out at different temperatures. With liquid nitrogen flowing, the temperature varies from 120K to room temperature. We measured electrical conductivity of a kind of semiconductor at different temperatures. The electrical conductivity-temperature curve in Fig.8 shows the exponential characteristic.

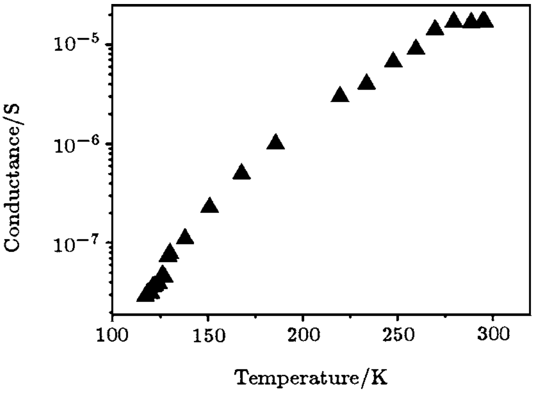

Fig.8. The exponential curve shows the electrical conductivity-temperature characteristics of the sample.

Optical characteristics are another important set of properties of nanoscale materials. In order to acquire the CL spectrum of a ZnO nanostick, we first took an SEM image of the ZnO nanostick, and then focused the electron beam on a selected part of the ZnO nanostick. Using a set of lenses to collect the light emission from the ZnO nanostick, we obtained the CL spectrum from the spectrograph. In the PL spectrum (Fig.9), a near-band-edge (NBE) ultraviolet emission peak, at around 385 nm, is clearly observed. This system can also be used to take the PL spectrum if a light source is employed. For example, using a He–Cd laser line of 325 nm as the light source, we can get the PL spectrum of the ZnO nanostick.

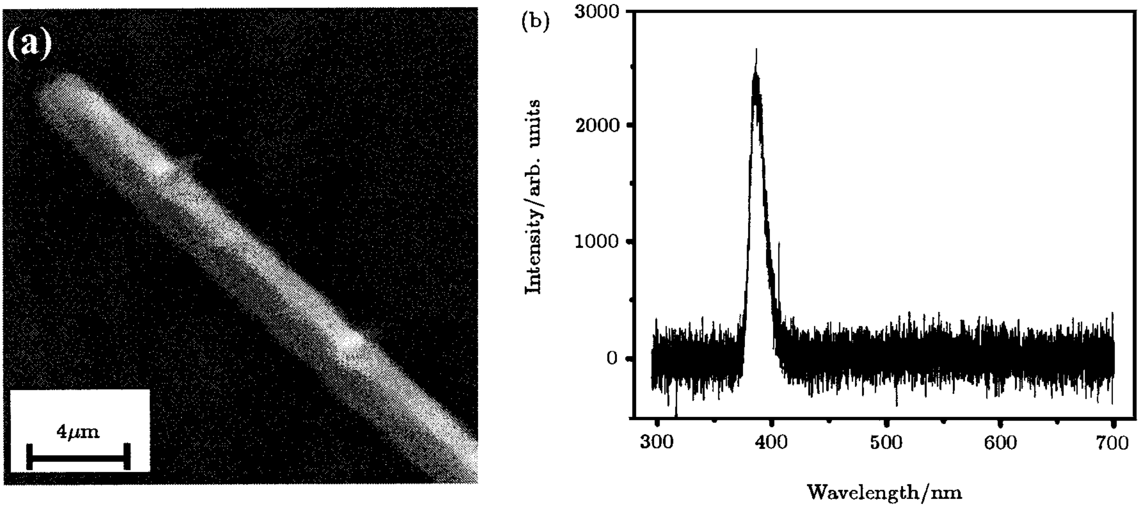

Fig.9. SEM image (a) of ZnO nanostick and CL spectrum (b).

4. Future work

It is clear by now that the four-probe STM system is a very powerful tool for manipulating some low dimensional nanostructures and measuring their electrical, magnetic and optic properties. We will focus research on low dimensional nanostructures, especially 1D nanostructures, such as functional molecules, nanotubes, nanorods, nanowires, nanojunctions, etc.[15–17] With the help of the MBE subsystem, we also can take advantage of the nanoprobe to investigate the properties of 2D nanostructures, even some fractal structures.[18]

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