Development of ultralow temperature scanning tunneling microscope cooled by a dilution refrigerator

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An ultralow temperature scanning tunneling microscope (STM) was developed by installing an STM head in a dilution refrigerator. Using the apparatus, STM images and the differential tunneling conductance spectra were taken at the sample temperature of 90 mK on a cleaved surface of 2H-NbSe₂. The atomically-resolved and the CDW-modulated STM images, and the superconducting gap as well as the CDW gap in the tunneling conduction (dI/dV) spectra were observed. [DOI: 10.1380/ejssnt.2004.151]

Keywords: Scanning tunneling microscope; Dilution refrigerator; NbSe₂; Charge density wave; Superconducting gap

I. INTRODUCTION

The invention of scanning tunneling microscopy (STM) [1], with its capability of producing atomically-resolved surface images, revolutionized in surface science and created nano science and technology. With an intention to expand its versatility, lots of STM apparatus working under various conditions have been developed. At very low temperature attained with a dilution refrigerator (< 300 mK), a variety of exotic and interesting physical phenomena, such as superconductivity and charge/magnetic ordering, are observed. In order to study these in a nanometer scale spatial resolution, several STM setups combined with a dilution refrigerator have been developed [2-5]. On the other hand, it has been difficult to probe surface properties at such low temperatures using conventional surface science techniques since these techniques are not basically compatible with the condition of the very low temperature. The cooled parts are often shielded to avoid thermal radiation from outside, and thus conventional surface science tools such as electron gun and energy analyzer are not accessible. In the present study we constructed a compact STM unit which can be installed and operated in a dilution refrigerator, mainly for surface science studies. The sample can be cooled down to 90 mK in the STM unit. In this paper we report on STM observation and tunneling spectroscopic measurements performed on a cleaved 2H-NbSe₂ surface.

II. EXPERIMENTAL

The ultralow temperature STM was constructed by attaching a home-made STM head on a mixing chamber of a commercial dilution refrigerator (Kelvinox 25, Oxford Instruments). The STM head is compact with ca. 30 x 30 x 50 mm³ in size, so that it can be fitted into an internal vacuum chamber (IVC) which contains the cooled parts of the refrigerator (Fig. 1). A piezo-driven linear motor motion similar to the one developed by Pan et al. [6] is used for coarse approaching of the tip attached on a scanning tube piezo actuator (outer diameter: 3 mm, inner diameter: 2 mm, length: 9 mm) toward the sample. The sample holder has six electrodes available for applying sample bias voltage, measuring resistance of temperature sensor, and other purposes. The holder can be transferred between the STM head and a UHV preparation chamber in which one can do basic surface preparation of samples, such as annealing, sputtering, metal deposition, gas adsorption and so on.

During an operation of the dilution refrigerator, three pumping systems have to be operated; a combination of turbo molecular pump (TMP) and rotary pump (RP) for IVC pumping, another TMP and RP combination for circulating ³He for the dilution process, and RP for pumping 1K pot, which pre-cools the circulating ³He gas. In order to avoid vibrations induced by these pumping systems to disturb the STM operation, all RPs are isolated into a remote box and the pumping lines are tightly clamped to the wall and floor. The STM apparatus including the dilution refrigerator with the STM head and the preparation chamber equipped with ion/titanium sublimation pumps are installed on a vibration isolation table (OSD-type, Showa Science). Details of the system will be pub-

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lished elsewhere [7].

In order to test its performance, we carried out STM and tunneling spectroscopic studies on a cleaved surface of 2H-NbSe₂. The surface is ideal for a test of low-temperature STM since it is inert and easily prepared, and the material shows charge-density-wave (CDW) and superconducting transitions at 33 K and 7.2 K, respectively. A sample was cleaved in air after setting it on the sample holder and set into the STM head. The sample temperature was measured with a resistive temperature sensor (Cernox, CX-1030, Lakeshore) attached to the sample holder adjacent to the sample. A Pt(90 %)-Ir(10 %) tip sharpened by electrochemical etching with CaCl₂/HCl solution was used as a probe.

III. RESULTS AND DISCUSSION

Figure 2(a) shows an STM image taken at the sample temperature of 90 mK. Atomic arrangement of the surface with a unit distance of 0.35 nm can be clearly observed. A slight modulation in height contrast with a periodicity of 3 times unit cell, which are marked with green circles in Fig. 2(a), corresponds to the CDW [8]. Fast Fourier transformed (FFT) pattern of the image is shown in Fig. 2(b). In addition to the regular 1×1 spots, fractional 1/3 spots are visible in the pattern.

The CDW formation is confirmed by an energy gap at Fermi energy ($E_F$) appearing in the tunneling conductance ($dI/dV$) spectrum (Fig. 3(a)), which was taken at 4.2 K (cooled by liquid He). It has been known that the differential conductance is basically proportional to the density of states (DOS) of the sample. The conductance was measured by the modulation method; applying a modulated voltage (770 Hz) on the sample bias voltage and measuring the same frequency component in the tunneling current with a lock-in amplifier. The gap width of ca. 35 meV is consistent with the previous reports [2]. In a narrow energy range, a gap sandwiched by two peaks, which is characteristic for DOS of superconductor, is observed (Fig. 3(b)). The shape of the spectrum is fitted with the superconducting DOS formulated by Bardeen-Cooper-Schrieffer (BCS) theory convoluted with a metallic DOS (Fermi-Dirac function) of the tip as is shown below [9]:

$$\frac{dI}{dV} \propto \int_{-\infty}^{+\infty} \frac{|E|}{\sqrt{E^2 - \Delta^2}} \left( \frac{1}{k_BT} \exp\left[\frac{E + eV}{k_BT}\right] + 1 \right)^{-1} dE,$$

where $k_B$ and $T$ are Boltzmann constant and temperature, respectively. $\Delta$ is a half the gap width of the superconductor. The previously reported value of $\Delta$ (1.1 mV at 4.2 K [10]) was assumed. By taking the smearing effect induced by a finite ac modulation voltage (0.8 mVpp) into account, we found a good agreement between the experimental (red) and theoretical (green) spectra in Fig. 3(b).

At 90 mK, we obtained a spectrum shown in Fig. 3(c),
obviously different from that of Fig. 3(b). It has a gap wider approximately twice than that of Fig. 3(b). A small peak is found at $E_F (V = 0)$. We believe that these features of the spectrum are due to tunneling between tip and sample both of which are superconducting. The tunneling spectrum between two superconducting electrodes can be given basically by a convolution of two BCS gap spectra, having an energy gap of $2(\Delta_1 + \Delta_2)$ and peaks in the gap width at an energy of $\pm 2|\Delta_1 - \Delta_2|$, where $2\Delta_1$ and $2\Delta_2$ are a superconducting gap of the two electrodes [8,9]. Since the measured spectrum is symmetric with respect to $E_F$ and the peak in the gap is located around $E_F$, the gap width of the tip should be same as that of the sample. Pt-Ir alloy, which is a material we used for a tip, does not become superconducting at the temperature ($T_C < 10 \text{ mK}$). Surface segregation of Ir would make the tip apex superconducting, but its superconducting gap is $\sim 0.02 \text{ meV}$, too small to account for the spectrum shape. The most probable candidate for the tip material is NbSe$_2$, same as that of the sample. Probably, a piece of NbSe$_2$ was picked up by the tip during scanning or approaching and the tunneling current flew at an apex of the piece. Since coherent length of the material is 7.7 nm, the piece has to be larger than the length to be superconducting.

IV. CONCLUSIONS

In conclusion, we have developed an ultralow temperature STM by attaching a STM head to a dilution refrigerator. Using the setup, STM images and $dI/dV$ spectra were taken at the sample temperature of 90 mK on a cleaved surface of 2H-NbSe$_2$. The atomically-resolved and CDW-modulated STM images demonstrate high spa-
tial resolution of the setup. The superconducting gap of the material as well as the CDW gap was observed in the differential tunneling conductance spectra.

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