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Temperature control of a liquid helium cooled Eigler-style scanning tunneling microscope

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A procedure for operating an Eigler-style, low temperature scanning tunneling microscope (STM) at variable temperatures has been developed. A critical exchange gas pressure regime was found to allow for controlled variation of the STM temperature while it is encapsulated in a liquid helium Dewar. The sensitivity of various parameters to the ability to generate stable variable temperatures above 4 K is discussed. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1416108]

I. INTRODUCTION

Soon after the scanning tunneling microscope enabled atomic-scale views of metal and semiconductor surfaces, diffusion of adsorbates was observed.[1] To determine experimentally the energetics of such activated processes, the temperature must be varied. More recently, Eigler and his group have provided spectacular demonstrations of what could be done with a low temperature (4 K) scanning tunneling microscope (STM).[2–9] Largely motivated by this work, today numerous low temperature STMs of different designs are in operation.[10–18] Here we describe a low temperature STM that operates stably over a range of temperatures, including down to 4 K. This enables us to record the dynamics of processes over a large multiplicative range in absolute temperature.

Figure 1 shows the sophisticated design of the helium-cooled STM invented by Eigler and co-workers.[19] A pendulum is the main design feature for providing mechanical vibrational insulation. At its end, the STM head is fixed in an ultrahigh vacuum environment while the upper end is suspended from a set of stainless steel bellows, giving the STM head the freedom to swing in all directions at a frequency of about 1 Hz. Details regarding the construction of this style of low temperature STM and its operation are published elsewhere.[20] The pendulum is placed inside of an exchange gas canister, which in turn is inserted into a liquid helium Dewar. Radiation shields are welded along the length of the pendulum to ensure a gentle gradient in temperature from the top to the bottom of the pendulum. In order to cool the STM, the volume of the canister is evacuated, and then backfilled with helium gas. This exchange gas is cooled by interacting with the walls of the gas canister in contact with the liquid cryogen and to cool the STM down to 4 K, a helium gas pressure on the order of 10–1 mbar is needed. In this pressure range, both good thermal coupling and acoustic insulation from the boiling helium are obtained, yielding the low temperature desired and the high stability needed to produce high-resolution images.

Because there are also many interesting experiments to do at temperatures higher than at 4 K, e.g., studies of surface diffusion and phase transitions, we have used our Eigler-design STM[20] for performing imaging and dosing at higher temperatures by reducing the pressure of the exchange gas.[21] In this article, we describe the cooling and controlling principles.

II. COOLING PRINCIPLE

A. Basic considerations

The thermal coupling between the walls of the exchange gas canister that are at a temperature of 4 K and the STM flange is determined by the pressure p of the helium gas. The thermal conductivity of the helium gas λ is a function of the mean free path l_m and the number n_{He} of He atoms per cm^3 involved in the heat exchange process.[22] To explain the cooling mechanism, it is useful to consider two pressure regimes. One range is above a critical pressure p_c, where l_m is smaller than the mechanical dimensions, and the other pressure range is below p_c, where the l_m is larger than the mechanical dimensions. For $p \gg p_c$, the thermal conductivity is

$$\lambda \sim l_m * n_{\mathrm{He}}. \tag{1}$$

Since l_m is inversely proportional to p, and n_{He} is directly proportional to p, the thermal conductivity is approximately a constant. In the low pressure region $p \ll p_c$, where l_m is larger than the mechanical dimension, He atoms can fly without scattering between the canister wall at 4 K and the STM flange. Therefore the thermal conductivity depends only on the number of He atoms per cm^3:

$$\lambda \sim n_{\mathrm{He}}. \tag{2}$$

Since n_{He} is proportional to p, the thermal conductivity is directly proportional to the pressure $\lambda \sim p$. The mechanical dimensions are on the order of 1–5 cm and l_m on that order corresponds to a pressure $p_c \approx 1.5 \times 10^{-3}$ mbar. Unfortu-

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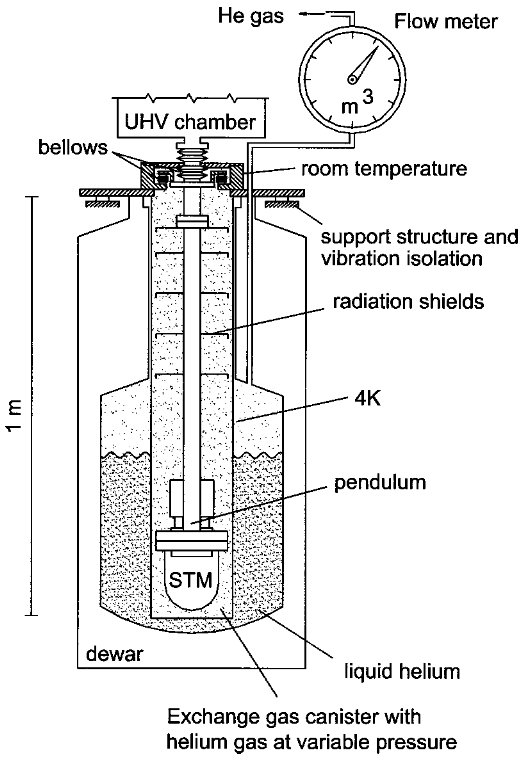

FIG. 1. Illustration of the basic design principle for the Eigler-style low temperature STM. The STM is mounted at the end of a bellows-supported pendulum, which is encapsulated in an exchange gas canister inside of a liquid helium Dewar.

nately, we cannot measure the pressure at the 4 K site but only at room temperature, so we have to consider a reduction factor of 300/4=75.

Figure 2 shows the STM flange temperature as a function of the helium pressure in the exchange gas canister, with the pressure being measured at room temperature with a cold cathode gauge, PKR251 (Pfeiffer Vakuum). The curve is relatively steep indicating that the temperature changes rapidly with small variations in helium pressure. The time constants associated with varying the temperature as a function of helium pressure make this measurement very difficult. To measure one data point, the He pressure is set to a value where the temperature falls slowly. Then, the pressure is incrementally reduced by pumping cycles until the temperature gradient falls to zero. The time to measure one data point is about 3–4 h. This plot, however, gives a first impression of the critical pressure range. This exchange gas pressure regime allows us to operate the microscope at variable temperatures by changing the local helium pressure with the introduction of a heater at the STM flange. The design aspects of the heater control will be discussed in the following sections. It can be seen in Fig. 2 that the desired pressure for operating over a range of temperatures is on the order of $2-7\times10^{-4}$ mbar. Again, this value is the pressure measured at the room temperature top of the pendulum, which was corrected for helium by a factor of approximately 5.9, because the gauge is calibrated for nitrogen.

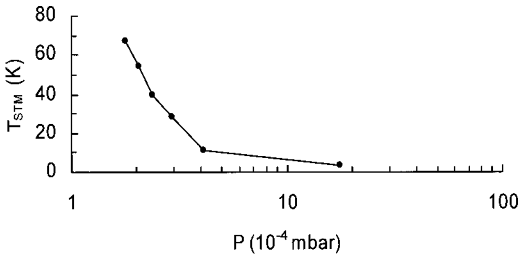

FIG. 2. Temperature of the STM as a function of He pressure inside the exchange gas canister. Changes in temperature are accessible with only small variations in the local helium pressure at the STM flange.

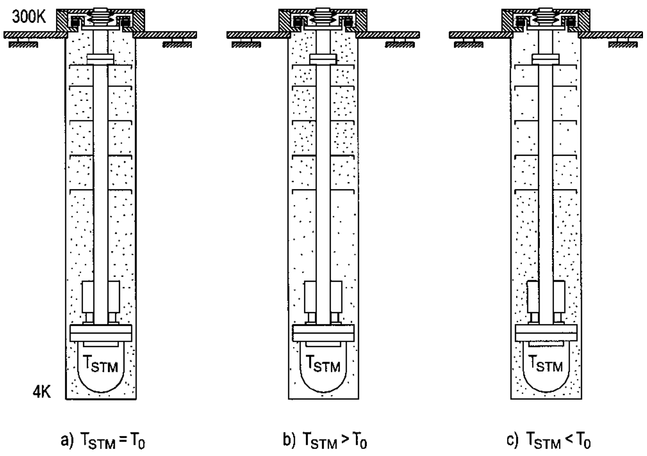

FIG. 3. (a) Distribution of helium atoms for a given STM temperature. The distribution has been exaggerated for the illustration. At temperatures significantly lower than 300 K, the vast majority of the helium atoms will be at the STM site. (b) If T_{STM} is raised, then the helium atom distribution changes, shifting the population to the 300 K end of the pendulum. The thermal conductivity is capable of dropping to a point where the temperature cannot be stabilized. (c) For a lower T_{STM}, the helium atom distribution shifts to the STM and increases the cooling power to the STM.

B. Unstable passive system

To avoid any mechanical vibration during tunneling, a valve shuts the exchange gas canister, and the turbo pump which pumps the helium gas is switched off. Now we have a closed system, where the number of He atoms N_{He} is constant but the density of He atoms n_{He} involved in the cooling process changes with the STM flange temperature. Figure 3(a) schematically shows a distribution of He atoms for a certain flange temperature $T_{\mathrm{STM}}=T_0$. Of course in reality the He atoms are not distributed equally as shown for didactic reasons in Fig. 3(a), but many more atoms are at the 4 K site than at the 300 K site.

The thermal loss caused by radiation and the thermal conductivity of the pendulum is balanced by the thermal conductivity of the He gas. At first sight, the system appears stable because in the case of a small temperature rise of the STM flange, the cooling power increases because of the increase in the temperature difference between the STM flange and the surrounding 4 K walls. This would only hold true for a source of constant thermal conductivity, which would lead to a cooling power that is proportional to ΔT. For the case of helium gas as the source of cooling, there is another effect to consider. When the temperature of the flange rises somewhat, the local distribution of He atoms inside the exchange gas canister changes as shown in Fig. 3(b). More He atoms are then located at the room temperature site and less He atoms

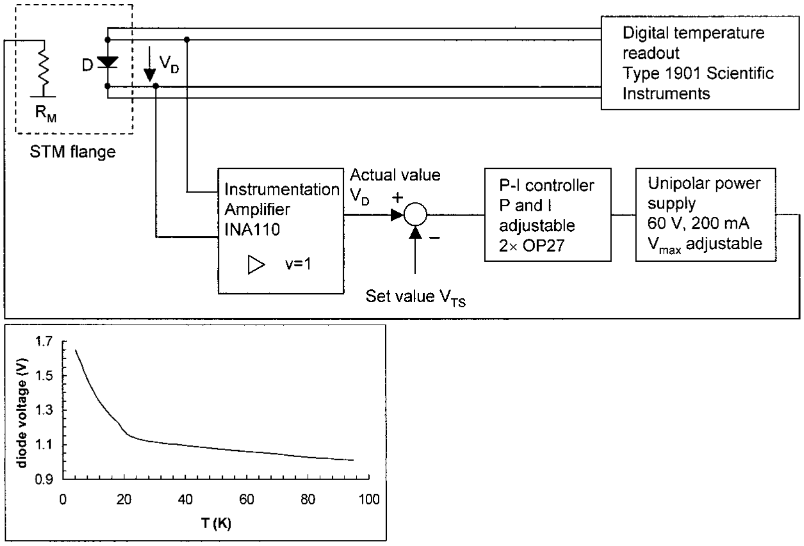

FIG. 4. Temperature control circuit used for stabilizing elevated temperatures. The inset plot relates the output diode voltage V_D to changes in temperature.

per cm^3 are involved in the cooling process, i.e., the thermal conductivity λ of the He gas decreases and the flange temperature can rise even further. For the reverse case as shown in Fig. 3(c), the behavior is similar; the temperature goes down as a result of an increased λ. This effect is further amplified by the falling curve of the pendulum's stainless steel heat conductivity between 300 and 4 K, which changes by nearly 2 orders of magnitude between 2×10^{-3} and 1.5×10^{-1} W/cm K.[22] These effects make the system unstable, i.e., the temperature of the STM cannot be kept constant without a temperature controller.

III. STM FLANGE TEMPERATURE CONTROLLER

It is not possible to stabilize the temperature by controlling the pressure of the exchange gas because the mechanical vibrations from the turbopump would be transmitted to the STM. Also, the time constant of such a process would be very large. The easiest way to control the system is to put a heater (wire-wound resistor) on the STM flange and to control this power to stabilize the temperature.

Figure 4 shows the block diagram of the temperature control circuit. A silicon diode temperature sensor (DT-470-SD-12, LakeShore Cryotronics) is mounted on the STM flange. The diode voltage V_D from the temperature readout instrument (Type 1901, Scientific Instruments) is fed into an instrumentation amplifier (INA110, Burr–Brown) and then compared with the adjustable preset voltage V_{TS}. A *PI* controller (*I* is time constant and *P* is amplification factor) is driven by the voltage difference V_D-V_{TS}, and its output voltage drives a unipolar voltage controlled power supply (60 V, 200 mA). Only when the voltage difference V_D-V_{TS} is positive does the output of the *PI* controller turn positive and the output voltage of the power supply heat the wire-would resistor R_M. The reverse sign of the voltage difference is caused by the characteristic of the diode voltage temperature dependence (see Fig. 4 inset). To avoid a large temperature overshoot, the maximum power of the power supply is adjustable.

The parameters of the controller are adjustable. This is necessary because the time constant of the system varies with temperature. The specific heat coefficient of the flange material (mainly stainless steel and copper) ranges approximately from 10^{-4} J/g K at 4 K to 10^{-1} J/g K at 80 K. This means the system reacts much faster at lower temperatures than at higher ones. Experiments have shown that the control circuit gives a reasonably small temperature error without an integral path. The proportional amplification factor has to be optimized and should be approximately 70% of the value at which the system oscillates. Figure 5 shows the control characteristic after V_{TS} was lowered from 1630 to 1610 mV to raise the temperature from 4.9 to 5.3 K. The temperature is stabilized within 10 min and the period of the damped oscillation is about 4.7 min.

The control circuit is very simple, but the operating parameters must be chosen very carefully. When operating the STM at a temperature higher than 4 K, the thermal coupling between the flange and the canister allows heat to be transmitted to the liquid helium in the Dewar. Liquid helium consumption through boil off is measured by a helium flow meter (see Fig. 1) and this value acts as a sensitive gauge for the degree of coupling between the STM and the walls of the canister. For helium consumption of ~0.5 l/h, the flange temperature gradient is zero. Figure 6 shows the temperature gradient as a function of helium consumption.

At a helium flow rate below 0.5 l/h the thermal coupling between the 4 K walls and the microscope flange cannot

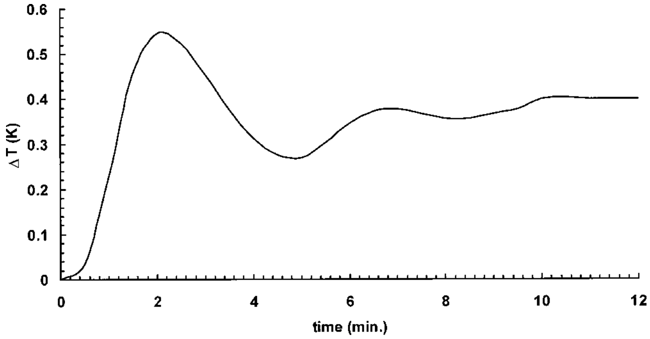

FIG. 5. Controller characteristic at 5 K after a change of the preset value to raise the temperature from 4.9 to 5.3 K.

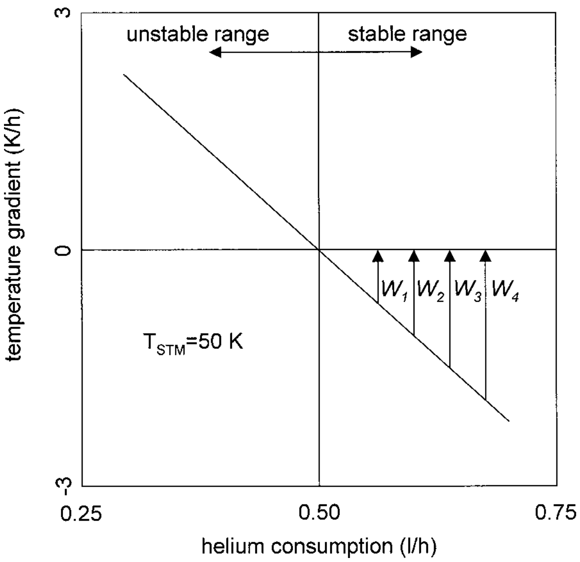

FIG. 6. Helium consumption rate plotted against rate of temperature change for stabilizing a STM flange temperature of 50 K. Power settings to the STM flange heater are listed where $W_1 = 150$ mW.

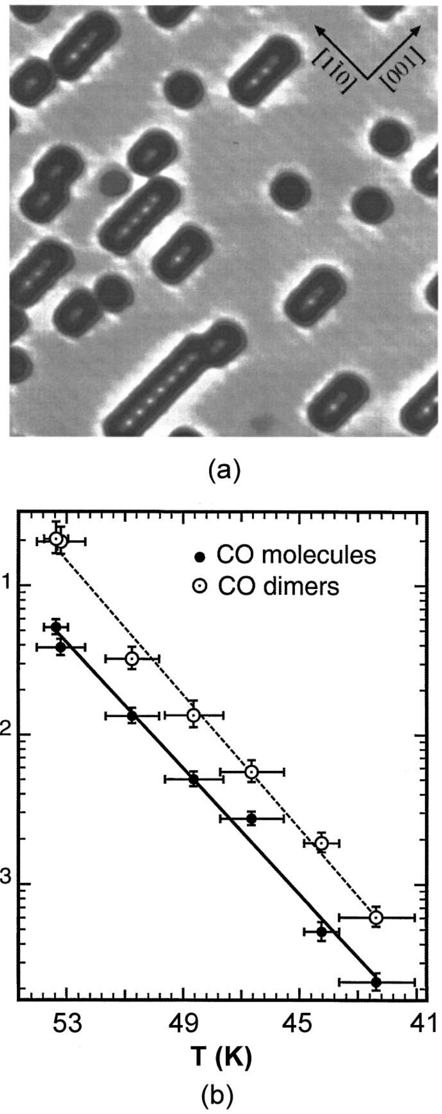

FIG. 7. (a) 100 Å×100 Å image of CO molecules on Cu(110), $V_b = -20$ mV, $I_t = 10$ nA, $T = 5$ K. Molecules were dosed at a STM temperature of 53 K. (b) Arrhenius analysis illustrating the difference in activation energy for hopping between single CO molecules and dimers.

balance the incoming heat by radiation and conduction from the 300 K site. Therefore, the flange temperature rises and there is no way to control the system by heating the resistor. The consequence is that there is no stable operating point for a helium consumption below 0.5 l/h. The pressure in the gas exchange canister has to be adjusted such that the helium flow is slightly above 0.5 l/h. The cooling power of the increased helium flow can then be compensated by the electrical power W_i to keep the flange temperature constant. It is best to choose an operation point close to 0.5 l/h to keep helium consumption low. In addition to indicating whether a stable elevated temperature can be accessed, measuring the helium consumption rate determines the ability to perform experiments over many hours. Various STM studies involve imaging or acquiring tunneling spectra over long periods of time[23,24] and the consumption rate must be kept low enough so that the need to replenish helium does not reduce its ability to operate for long times at elevated temperatures.

In the limit of small temperature changes close to 4 K, i.e., 4–10 K, the perturbation in local helium pressure at the STM does not significantly change the degree of thermal coupling to the canister walls. However, when attempting to stabilize temperatures above 10 K, as high as 60 K, the system needs to be tuned very carefully with regard to the pressure of helium in the exchange canister. Since it is not possible to measure the helium pressure at the STM, the best indicator of that pressure is the rate of liquid helium consumption through boiloff. The heater power and helium pressure in the canister can then be tuned to achieve a manageable helium consumption rate.

IV. APPLICATION EXAMPLE

As an example of variable temperature operation, Fig. 7(a) shows adsorbed CO molecules on a Cu(110) surface. Measurements of the diffusion of CO and CO dimers were performed between 42 and 53 K. Figure 7(b) illustrates the difference in activation energy associated with the hopping frequency of the two different adsorbed species.[25] In order to perform experiments over a given temperature range in a serial fashion, it was found to be more efficacious to start a set of experiments at the high temperature end of the range and then to reduce temperature. For the inverse experiment, the sample was more susceptible to contamination as the entire STM was warmed up. As shown in Fig. 4, careful control of the temperature in the STM enabled quantitative simultaneous measurements of diffusion of adsorbed molecules.

V. DISCUSSION

The pendulum suspension makes the Eigler-style scanning tunneling microscope one of the most stable instruments for low temperature operation. We have shown that by reducing the helium gas pressure in the exchange gas canister, this STM can also been used at higher temperatures to perform temperature-dependent measurements without degrading its high stability. We have also shown the design of a temperature controller and how to choose the operating parameters to keep the temperature stable while minimizing the helium consumption.

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