AIP ADVANCES **8**, 075013 (2018)

Combined *I(V)* and *dI(V)/dz* scanning tunneling spectroscopy

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(Received 11 April 2018; accepted 7 July 2018; published online 13 July 2018)

We present a method to simultaneously record *I(V)* and $\frac{dI(V)}{dz}$ spectra in a scanning tunneling microscopy measurement, where *I*, *V* and *z* refer to the tunnel current, sample bias and tip-substrate separation, respectively. The *I(V)* spectrum is recorded by ramping the bias voltage, while the feedback loop of the scanning tunneling microscope is disabled. Simultaneously the *z*-piezo is modulated with a small sinusoidal high frequency signal. The $\frac{dI(V)}{dz}$ signal is recorded using a lock-in amplifier. This method allows to simultaneously record the topography, $I(V)$, $\frac{dI(V)}{dV}$ and $\frac{dI(V)}{dz}$ in a single scanning tunneling microscopy measurement.
https://doi.org/10.1063/1.5034422

INTRODUCTION

The invention of the Scanning Tunneling Microscope (STM) by Binnig and Rohrer[1] has revolutionized our ability to visualize, study, and manipulate solid surfaces on the size scale of atoms. Besides its unparalleled spatial resolution, the STM is also capable of measuring the spatial variation of electronic properties of a surface, such as the density of states and the work function. The density of states at a particular location can be determined by measuring the tunnel current as a function of the sample bias (*I(V)*) at a constant substrate tip distance. For small sample biases the differential conductivity ($\left(\frac{dI}{dV}\right)_z$) is proportional to the local density of states. This method is commonly referred to as scanning tunneling spectroscopy (STS).[2–5]

There are, however, several other spectroscopic modes in scanning tunneling microscopy. By measuring for instance the *z*-piezo displacement as a function of the sample bias (*z(V)*) at constant tunnel current, i.e. with the feedback of the STM enabled, the local density of states can be obtained as well.[6–8] Another scanning tunneling spectroscopy mode is current-distance spectroscopy (*I(z)*).[2–5,9] In this spectroscopic mode the tunnel current is measured as a function of substrate-tip distance at constant sample bias, while the feedback loop of the STM is disabled. This method provides direct information on the height of the tunneling barrier, i.e. the averaged work function of tip (ϕ_t) and substrate (ϕ_s) and inverse decay length.[2–5,9–12]

The tunnel current is usually dominated by electronic states at, or in the vicinity of, the Γ point of the Brillouin zone of the surface of the substrate. However, if there are no states available at the Γ point, electrons have to tunnel to or from electronic states with a non-zero parallel momentum. The transition from tunneling to (or tunneling from) electronic states with a small parallel momentum to electronic states with a larger parallel momentum will lead to an increase of the inverse decay length. Since the tunnel current depends exponentially on the inverse decay length even a relatively small change in the parallel momentum can be easily detected. By simultaneously recording the *I(V)* and

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$\frac{dI(V)}{dz}$ spectra one will obtain additional information of the location of the electronic states in *k*-space. Already in 1986 Stroscio, Feenstra and Fein have shown in a seminal paper[2] that the inverse decay length can be measured by recording a series of *I(V)* curves at various set points. These experiments are experimentally very demanding, because the STM tip has to be stable throughout the whole series of measurements at different set points. The latter problem can be circumvented if the *I(V)* and *dI(V)/dz* measurements are recorded simultaneously. Since the tunnel current depends on the sample bias, *V*, and the tip-substrate distance, *z*, it seems, at least at first sight, impossible to simultaneously record the *dI(V)/dV* and *dI(V)/dz* signals. Here we solve this problem by modulating the *z*-piezo with a small, but high-frequency, sinusoidal signal. Owing to this modulation of the *z*-piezo the averaged tunnel current, $<I>$, will increase with respect to the reference tunnel current without modulation. In case that the substrate bias sampling rate is substantially lower than the *z*-piezo modulation frequency the *I(V)* and *dI(V)/dz* signals can be recorded simultaneously. The differential conductivity, *dI/dV*, can be obtained by numerically differentiating the *I(V)* signal.

EXPERIMENTAL

The STM and STS experiments are performed with an Omicron ultra-high vacuum low-temperature STM. The base pressure of the ultra-high vacuum STM system was below $1\text{x}10^{-11}$ mbar. Samples and tips can be introduced via a load-lock system into a preparation system, which is equipped with facilities for sample annealing and Argon ion sputtering. The base pressures of the load-lock and preparation systems are $1\text{x}10^{-9}$ mbar and $1\text{x}10^{-10}$ mbar, respectively. The STM and STS measurements in this study are performed at 77 K. The tunnel current is converted to a voltage using a hybrid SPM PRE 4E preamplifier of Omicron with a bandwidth of 80 kHz. We have used a dual digital quad-channel high-frequency lock-in amplifier of Anfatec for our STS experiments. The *z*-piezo is modulated with an amplitude of ~0.28 Ångström and a frequency of 1831 Hz. This modulation frequency is higher than the cut off frequency of the STM feedback loop (<1 kHz). The sensitivity of the *z*-piezo depends on the temperature and varies for our tube scanner from 6.7 nm/V at room temperature to 2 nm/V at 77 K. The voltage sweep time is 35 ms per data point.

The Ge(001) samples were cut from nominally flat single-side-polished and nearly intrinsic *n*-type wafers. The samples were mounted on Mo holders and contact of the samples with any other metal has been carefully avoided during the preparation. Atomically clean Ge(001) substrates were obtained by several cycles of 500 eV Ar^+ ion sputtering, followed by annealing the sample through resistive heating at 1100 (±25) K.[13]

RESULTS AND DISCUSSION

In order to illustrate our method we make use of the well-known Simmons model. Although the Simmons model is very simplistic it mimics the main characteristics of a tunnel junction. The tunnel current, *I(V,z)*, scales linearly with the sample bias, *V*, and depends exponentially on the sample-tip distance, *z*, i.e.

$$I(V,z) = CVe^{-2\kappa z}, \tag{1}$$

where $\kappa = \sqrt{2m\phi}/\hbar$ is the inverse decay length and *C* a constant. The inverse decay length at a fixed voltage V can be obtained by measuring the dependence of the tunnel current on the tip-sample separation.[2–5,9] Since most materials have a work function, ϕ, of about 4-5 eV, κ has a typical value of about 1 $Å^{-1}$. If we add a small sinusoidal signal to the *z*-piezo the substrate-tip separation varies as $z(t) = z_0 + \hat{z}\sin(\omega t)$, where *t* is the time, z_0 the reference substrate-tip separation, $\hat{z}$ the amplitude of the sinusoidal signal and ω the frequency of the oscillation. By inserting $z(t)$ in Eq. (1) we find,

$$I(V,z) = CVe^{-2(z_0+\hat{z}\sin(\omega t))\sqrt{\frac{2m\phi}{\hbar^2}}} = I_{ref}e^{-2\kappa\hat{z}\sin(\omega t)} \tag{2}$$

where I_{ref} is the reference tunnel current without modulation. The averaged tunnel current, $\langle I\rangle$, can be found by integrating the tunnel current over a full oscillation period and dividing by an oscillation period,

$$\langle I\rangle = I_{ref}\frac{\omega}{2\pi}\int_0^{2\pi/\omega} e^{-2\kappa\hat{z}\sin(\omega t)}dt \tag{3}$$

which can be rewritten as,

$$\langle I\rangle = I_{ref}I_0(2\kappa\hat{z}) = I_{ref}\sum_{m=0}^{\infty}\frac{1}{m!\Gamma(m+1)}(\kappa\hat{z})^{2m} \tag{4}$$

where I_0 is the modified Bessel function of the first kind and of the zeroth order and Γ the gamma function. Eq. (4) reduces to,

$$\langle I\rangle = I_{ref}\left[1+(\kappa\hat{z})^2+\frac{1}{4}(\kappa\hat{z})^4+\frac{1}{36}(\kappa\hat{z})^6+\ldots\right] \tag{5}$$

For small amplitudes, i.e. $\kappa\hat{z}< 1$, $\langle I\rangle$ is slightly larger than I_{ref}. As an example, for $\kappa\hat{z}=0.5$ we find that $\langle I\rangle\approx 1.267I_{ref}$. For small amplitudes, i.e. $\hat{z}< 0.5$ Å the temporal variation of the tunnel current is approximately sinusoidal, however for larger amplitudes the peaks in the *I(V,z,t)* curve become sharper, whereas the valleys become more rounded.

The simplest experiment to check the effect of a sinusoidal *z*-piezo modulation on the tunnel current is a measurement of the tunnel current with the feedback loop of the scanning tunneling microscope disabled. If the parameters are chosen well, i.e. sufficiently high modulation frequency, small *z*-piezo modulation and sufficiently large tunnel current integration time, the averaged tunnel current, *<I>,* will increase with increasing amplitude of the *z*-piezo modulation (here we have taken a modulation frequency of 1831 Hz, a *z*-piezo modulation <0.1 nm and a tunnel current integration time of 10 ms). In Figure 1 we shown the variation of *<I>* as a function of the amplitude of sinusoidal z-piezo modulation. The amplitudes are 0.28 Å, 0.56 Å, 0.84 Å, 1.04 Å and 1.32 Å, respectively. Alternatively, the feedback loop of the scanning tunneling microscope can also be enabled. In this case the tunnel current remains fixed at a pre-defined set point current, but the *z*-piezo retracts with increasing amplitude of the *z*-piezo modulation in order to keep the averaged tunnel current constant. In the latter case the frequency of the *z*-piezo modulation should be substantially larger than the cut off frequency of the feedback loop of the scanning tunneling microscope electronics. Both experiments reveal that the tunnel current and/or *z*-piezo displacement alters upon an increase of the amplitude of the sinusoidal *z*-piezo modulation. In addition, these measurements also show that a conventional *I(V)* curve can be recorded, while a small sinusoidal modulation is added to the *z*-piezo, provided at least that the frequency of the modulation is sufficiently high (the integration time in the tunneling current measurement should involve several oscillation periods of the *z*-piezo).

In the next series of experiments we recorded a conventional *I(V)* measurement, while a small high frequency sinusoidal modulation was added to the *z*-piezo. Simultaneously we recorded the *dI(V)/dz* signal using a lock-in amplifier. In Figure 2(a) an *I(V)* spectrum of a Ge(001) surface at 77 K is displayed. The *I(V)* curve was recorded while a sinusoidal signal with a root mean square amplitude of 10 mV (rms), which corresponds to an amplitude of the z-piezo modulation of 0.28 Å (at 77 K), and a frequency of 1831 Hz was added to the *z*-piezo. The *dI/dz* signal, which was recorded simultaneously with the *I(V)* signal using a lock-in amplifier, is displayed in Figure 2(b). In order to remove the offset of the *IV*-converter, the *I(V)* curve was set to zero at *V=0*. Since the *dI/dz* signal is

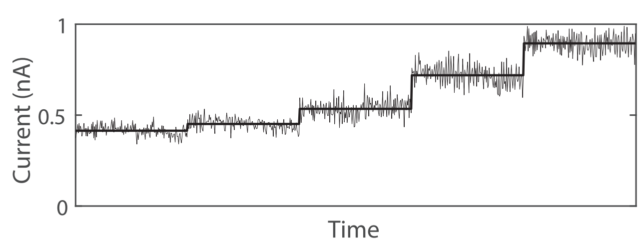

FIG. 1. Tunnel current as a function of the amplitude of the sinusoidal z-piezo displacement. The z-piezo amplitudes (from left to right) are 0.28 Å, 0.56 Å, 0.84 Å, 1.04 Å and 1.32 Å, respectively. Modulation frequency 1831 Hz, sample bias -1.5 V and temperature 77 K.

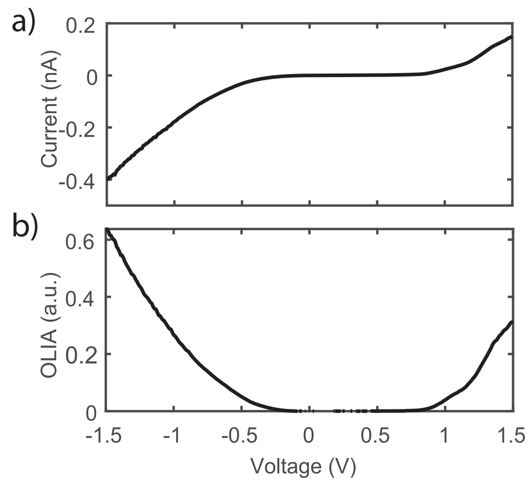

FIG. 2. (a) I(V) curve recorded on Ge(001) at 77 K. Setpoints I= 0.4 nA and V= -1.5 V. (b) dI(V)/dz curve recorded simultaneously with the I(V) curve. Modulation amplitude z-piezo 10 mV and a frequency of 1831 Hz.

proportional to I (see eq. (1)), also the dI/dz signal has to be set to zero at $V=0$. In Figures 3(a)–(b) the differential conductivity, $dI(V)/dV$, and the normalized differential conductivity, $(dI(V)/dV)/(I/V)$, are shown. The normalized differential conductivity is, as expected, 1 at zero bias. Electronic states are found at 0.5 eV below the Fermi level and 0.3 eV, 0.9 eV and 1.2 eV above the Fermi level. Although all the electronic states are also found in other experimental studies, the filled dimer state, which is located at -1.3 V, is not very well resolved in our spectrum.[14–16] The latter can be ascribed to several effects, such as the doping of the Ge substrate or the electronic structure of the scanning tunneling microscope tip. Since the aim of this paper is to introduce a new scanning tunneling spectroscopy mode we will not elaborate on the exact electronic structure of our sample.

As will be shown below the $dI(V)/dz$ signal can be used to determine the inverse decay length. First we derive an expression for the output of the lock-in amplifier, O_{LIA}. The output of the lock-in amplifier is given by,

$$O_{LIA} = \frac{1}{T}\int_0^T S_{ref} S_{input} dt \tag{6}$$

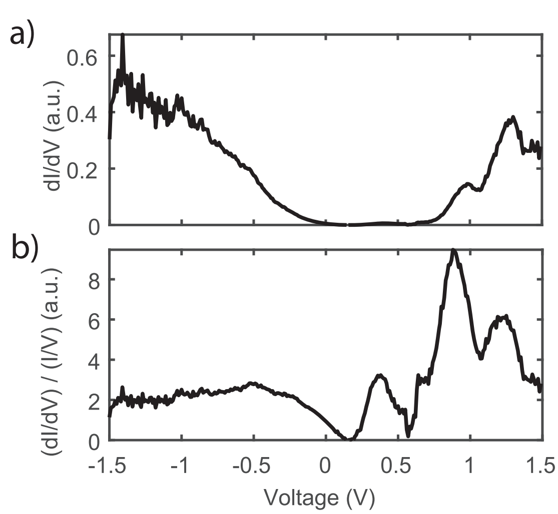

FIG. 3. (a) Differential conductivity, dI(V)/dV, versus voltage of a Ge(001) surface at 77 K. (b) Normalized differential conductivity versus voltage of a Ge(001) surface at 77 K.

The reference signal, S_{ref}, and input signal, S_{input}, are given by respectively,

$$S_{ref} = \hat{z}\sin(\omega t) \tag{7a}$$

and

$$S_{signal} = CVe^{-2\kappa(z_0+\hat{z}\sin(\omega t))} \tag{7b}$$

If we integrate eq. (6) over a full period, i.e. $T = 2\pi/\omega$, we find,

$$O_{LIA} = \frac{\omega}{2\pi}\int_0^{2\pi/\omega} \hat{z}\sin(\omega t)CVe^{-2\kappa(z_0+\hat{z}\sin(\omega t))}dt = \frac{\hat{z}CVe^{-2\kappa z_0}}{2\pi}\int_0^{2\pi}\sin(y)e^{-2\kappa\hat{z}\sin(y)}dy \tag{8}$$

Eq. (8) can be simplified to,

$$O_{LIA} = \hat{z}I_{ref}I_1(-2\kappa\hat{z}) \tag{9}$$

where I_1 is the modified Bessel function of the first kind and the first order. Eq. (9) reduces to,

$$O_{LIA} = \hat{z}I_{ref}(-\kappa z)\sum_{k=0}^{\infty}\frac{\left(\kappa^2\hat{z}^2\right)^k}{k!(k+1)!} = -\kappa\hat{z}^2 I_{ref}\left(1 + \frac{\kappa^2\hat{z}^2}{2} + \frac{\kappa^4\hat{z}^4}{12} + \frac{\kappa^6\hat{z}^6}{144} + ..\right) \tag{10}$$

The ratio $O_{LIA}/\langle I\rangle$ is given by,

$$\frac{O_{LIA}}{\langle I\rangle} = -\kappa\hat{z}^2\left[\frac{1 + \frac{1}{2}(\kappa\hat{z})^2 + \frac{1}{12}(\kappa\hat{z})^4 + \frac{1}{144}(\kappa\hat{z})^6 + ..}{1 + (\kappa\hat{z})^2 + \frac{1}{4}(\kappa\hat{z})^4 + \frac{1}{36}(\kappa\hat{z})^6 + \ldots.}\right] \tag{11}$$

For small amplitudes, i.e. $\kappa\hat{z}< 1$, the normalized signal of the lock-in amplifier, $O_{LIA}/ < I >$, is proportional to the inverse decay length, κ, and the squared value of the amplitude of the sinusoidal modulation.

In its most simplest form the inverse decay length is given by $\kappa = \sqrt{2m\phi}/\hbar$. A slightly more refined approach that involves (1) a tip and substrate which have different work functions, (2) an applied bias voltage, *V,* between tip and substrate and (3) tunneling to or from electronic states with a non-zero parallel momentum ($k_{parallel}$) gives the following expression for the inverse decay length,

$$\kappa = \sqrt{\frac{2m}{\hbar^2}\left(\frac{\phi_t + \phi_s}{2} - E + \frac{eV}{2}\right) + k^2{}_{parallel}} \tag{12}$$

where E is the energy relative to the Fermi level, V the sample bias voltage and $\phi_{t,s}$ the work function of tip and substrate, respectively. The smallest value of κ is obtained for electronic states of the substrate that are located at the Γ point of the surface Brillouin zone, i.e. $k_{parallel}$=0. If no states are available at the Γ point, electrons will tunnel to or from electronic states in the vicinity of the Γ point. The tunneling current is usually dominated by the electronic states with the smallest parallel momentum. In Figure 4 the inverse decay length of the Ge(001) substrate is depicted. Since the lock-in amplifier signal is only proportional to *dI(V)/dz* we still have to normalize the inverse decay length. In Figure 1 the change of the tunnel current is shown as a function of amplitude of z-piezo modulation at a sample bias of -1.5 V. By using Eq. (5) we can extract the value of the inverse decay length form the measured values of $<I>/I_{ref}$. We find an inverse decay length of 0.8 $Å^{-1}$ at a sample bias of -1.5 V. This value has been used to scale the inverse decay length in Figure 4.

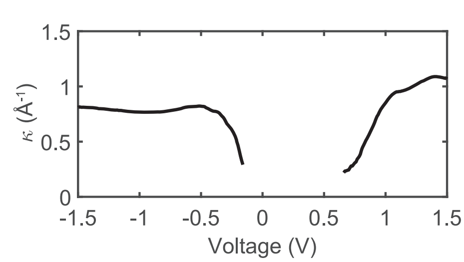

FIG. 4. Inverse decay length versus voltage.

Since the *I(V)* and *dI(V)/dz* signals are both very close to zero at low sample bias we have omitted the voltage range where the tunnel current is below a threshold value of 2 pA (0.5% of the set point current). This value of 2 pA is a worse-case estimated error bar in the tunnel current. It is clear from the inverse decay length plot that the smallest value of the inverse decay length is obtained in or near the band gap range of the Ge substrate. It should be pointed out here that the surface of Ge(001) is always slightly *n*-type owing to the dangling bonds, which lead to a band bending of -0.3 eV. The increase of the inverse decay length for voltages smaller than -0.5 V and larger than 1 eV reveals that tunneling occurs to or from electronic states with a non-zero parallel momentum.

CONCLUSIONS

We have developed a scanning tunneling spectroscopy mode that allows to simultaneously record the topography, $I(V)$, $\frac{dI(V)}{dV}$ and $\frac{dI(V)}{dz}$. At each pixel of a pre-defined grid an $I(V)$ curve is recorded while the feedback loop of the scanning tunneling microscope is switched off and the z-piezo is modulated with a high frequency small sinusoidal signal. Using a lock-in amplifier the $\frac{dI(V)}{dz}$ signal is simultaneously recorded with the $I(V)$ signal. The $\frac{dI(V)}{dV}$ signal is obtained by numerically differentiating the $I(V)$ signal. This new dual spectroscopic mode is not only easy applicable, but it is also very advantageous since the requirements regarding the stability of the STM tip are less severe as compared to the case that the *I(V)* and *dI(V)/dz* curves are recorded separately. This novel method not only provides information on the electronic states in energy space, but it also gives information regarding the dispersion in k-space.

ACKNOWLEDGMENTS

The authors thank Paul de Boeij and Bene Poelsema for stimulating discussions. C. C, R. van B and H.J.W. Z thank Dutch organization for scientific research NWO for financial support (FOM PR3237 and 14TWOD07).

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