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Dissipation Imaging with Low Amplitude off-Resonance Atomic Force Microscopy

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A small amplitude non-contact atomic force microscope/scanning tunnelling microscope (nc-AFM/STM) is used to study dissipative interactions at atomic resolution on Cu(100) and Si(111) surfaces. For Cu(100) atomic resolution images of phase contrast are obtained, showing energy dissipation as high as 100 meV/cycle at each atomic site during constant tunnel current scans. In contrast, the Si(111) 7×7 surface usually did not exhibit significant phase contrast during normal STM operating conditions. However, when the driving oscillation frequency was set to a sub-harmonic of the lever resonant frequency, atomic contrast in phase could be readily observed. We believe this harmonic coupling is due to the nonlinearity of the tip-sample interaction, and at these frequencies part of the energy is dissipated via the lever Q. [DOI: 10.1143/JJAP.44.5325]

KEYWORDS: Atomic force microscopy, scanning tunneling microscopy, low amplitude, force gradient, dissipation, phase, harmonic coupling

1. Introduction

Atomic force microscopy is widely used as a mechanical characterization probe as well as an imaging tool. With the advent of atom resolved nc-AFM, this capability has been extended to the atomic scale. An important resulting question is the extent to which energy is dissipated during these atom resolved measurements. In principle, for weak enough interactions, purely conservative processes and hence no dissipation should be involved. However, it has been known since the earliest nc-AFM results that significant mechanical energy is actually dissipated,[1–3] and the loss signal can even be used for imaging.[4] There have been theoretical studies proposing possible mechanisms for observed dissipation, amongst which are adhesion hysteresis,[3] atomic instabilities[5] and Brownian dissipation.[6] However, the actual origin remains uncertain. It is important, not only for a proper understanding of the interactions in imaging, but also because of its significance for atom motion and manipulation, and the relation between atom motion and local atomic and chemical structure. The type of experimental setup used is significant.[7] In this paper we present atom resolved dissipation images from an apparatus which uses very small oscillation amplitudes, and whose frequency can be varied independently of the lever resonance. This minimizes uncertainties about tip-surface separation, does not need deconvolution of interaction, and allows independent investigation of resonance effects. We show that non-linear coupling into the lever Q can be a significant dissipative effect.

2. Experiments and Results

We investigated Cu(100) and Si(111) surfaces using a UHV-AFM/STM which employs a high resolution fibre interferometer. Full details are given elsewhere.[8] The lever is oscillated with very small amplitudes (< 0.25 Å) at a frequency below its resonance, while it is scanned across the surface under tunnelling current feedback control. The changes in the oscillation amplitude are recorded via a lock-in amplifier. The oscillation amplitude gives the quantitative force gradient image directly, without deconvolution or assumptions about conservative interaction, as occur in large amplitude resonance AFM. Furthermore, the phase difference between the oscillation of the lever and the driving excitation is simultaneously recorded. This is a measure of the energy dissipation due to the tip-sample interaction.[1] In the case of very small oscillation amplitudes applied at frequencies far below resonance, the force gradient interaction between tip and sample can simply be expressed as:

$$\mathrm{d}F/\mathrm{d}z = k_0\left(1 - \frac{A_0}{A}\cos\varphi\right), \tag{1}$$

where k_0, A_0, A, φ are lever stiffness, free oscillation amplitude, the measured amplitude of the cantilever, and phase between the oscillations of the lever and its drive, respectively. The energy dissipated per cycle can be calculated, based on the model proposed by Anczykowski *et al.*,[9] using the following expression:[1]

$$E_{\mathrm{loss}} = \pi k_0 A_0 A \sin\varphi. \tag{2}$$

Home-made tungsten cantilevers with spring constants of 50 to 200 N/m were used in the experiments. The preparation and calibration of the cantilevers are reported elsewhere.[8] The Cu(100) sample, a 10 mm diameter disc of 3 mm thickness, was cleaned by repeated cycles of Ar ion bombardment at 1 kV for 30 min followed by an anneal for 30 min at 600°C. The (1×1) structure of the clean surface was verified independently by LEED investigation prior to AFM experiments. Si(111) sample was cut from P-doped, n-type wafer with 0.7–0.9 Ωcm resistivity, oriented to within 0.20° of (111) plane. It was cleaned by flash annealing to 900 and 1200 deg for a few tens of seconds following an overnight degas at about 600°C.

Figure 1 shows an example of a simultaneous STM, force gradient and phase shift image, taken using an oscillation amplitude A_0 of 0.25 Å. Both phase and force gradient exhibit atomic contrast whereas the corrugation in STM topography is less than 0.1 Å. The lack of atomic scale contrast in the STM signal means we have essentially a flat tip trajectory during imaging. This is expected for low index metals, and is shown by Eigler *et al.*[10] In turn, this means that the long-range component of tip-surface interaction force[11,12] is constant throughout the measurement. Hence

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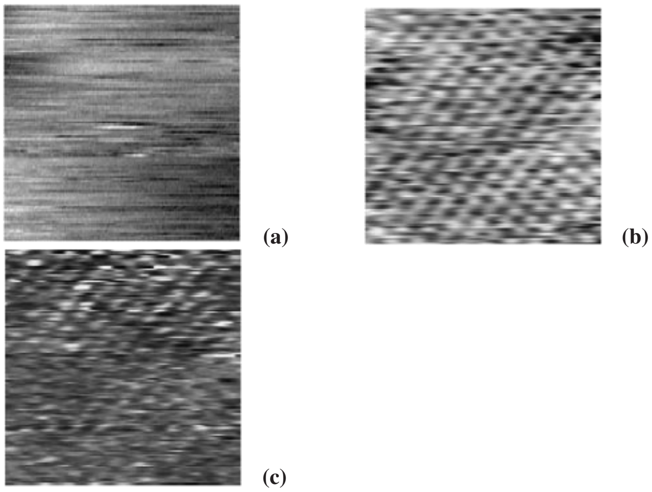

Fig. 1. Simultaneous STM (a), force gradient (b) and phase (c) images of Cu(100). Image size is $32 \times 32\,\text{Å}^2$. $V_{tip} = 20\,\text{mV}$, $I_T = 16\,\text{nA}$, $A_0 = 0.25$ Å and $k_0 = 55\,\text{N/m}$. Black to white scale corresponds to 541.2 N/m (force gradient) and 1.53 Å (STM). STM corrugation amplitude is <0.1 Å.

the force gradient and phase scans are mapping the truly local interactions and dissipations at the surface. This particular scan showed a corrugation at the single atom periodicity of about 16 deg in phase, which corresponds to an energy loss of about 100 meV/cycle. Another scan taken with a larger tip-sample separation showed a phase corrugation of 1.6 deg corresponding to about 10 meV dissipated energy. The measured values of energy loss are expected, since at usual STM operation separations on metal surfaces there are already strong forces between the tip and sample which readily result in atomic instabilities and relaxations on either side.[13,14] This atomic scale contrast in phase images suggests a significant energy loss at every atomic site. Elsewhere[1] we have proposed a simple mechanism that explains the energy dissipation in the tip-surface region. An atom or defect in the vicinity of the tip, which can assume two energy states undergoes a transition between the two states induced by the interaction. Within the mechanism of a bistable atom that successfully accounts for the observed dissipation, the tip is able to preserve its integrity and this allows the stability of the AFM images despite the presence of significant dissipation.

For semiconductor surfaces, the electronic properties and surface geometry result in different STM and force imaging conditions from those of metals. Significant STM topographic contrast is available at moderate currents. On Si surfaces the onset of tunnel current is experimentally known to occur before the start of stronger short-range force interactions.[12,15] Unlike metals, therefore, it is possible to obtain strong contrast STM images of Si with relatively small (but still measurable) force gradients present. We find that during almost all our Si(111) experiments, the phase signal showed either weak or no measurable atomic scale contrast. This is broadly consistent with the fact that during STM operation at typical tunnel currents there is a relatively weak force gradient of interaction.[16] However, whilst varying the lever drive frequency, we noted that at certain frequencies, significant atomic contrast in phase could be

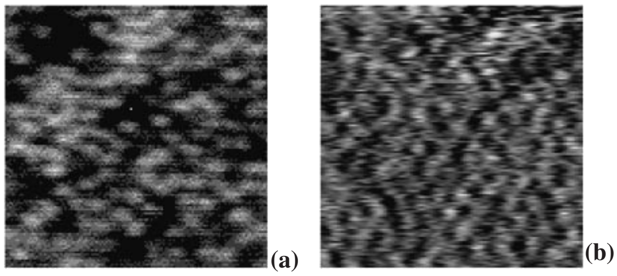

Fig. 2. Simultaneous STM (a) and phase (b) images of Si(111). Image size is $100 \times 100\,\text{Å}^2$. $V_{tip} = -2\,\text{V}$, $I_T = 0.5\,\text{nA}$, $A_0 = 0.5\,\text{Å}$, $k_0 = 98\,\text{N/m}$, $f_{res} = 15094\,\text{Hz}$, $f_0 = 7470\,\text{Hz}$.

obtained. Interestingly, these frequencies were found to be around half the fundamental resonance frequency of the cantilever. Figure 2 shows simultaneous STM and phase images of a Si(111) surface taken with a lever whose free resonant frequency was 15094 Hz. The oscillation amplitude and frequency were 0.25 Å and 7470 Hz respectively. Phase scan, as well as STM topography, shows atomic contrast. In another image with the same lever, the drive frequency was changed to 7501 Hz, whilst keeping all other imaging parameters the same as the 7470 Hz scan. The resulting STM topography and phase images are shown in Fig. 3. The change of frequency by only about 30 Hz resulted in a considerable decrease in atomic corrugation amplitude in phase image. We then scanned the surface with various other frequencies around half of the resonance frequency of the lever, and observed that the corrugation height in phase scans changed in a distinct manner. Figure 4 shows phase image corrugation height values corresponding to various drive oscillation frequencies. The corrugation height peaks

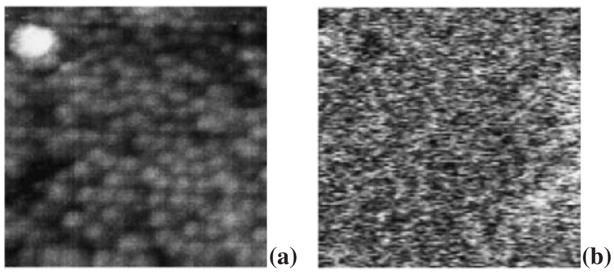

Fig. 3. Simultaneous STM (a) and phase (b) images of Si(111). Image size is $100 \times 100\,\text{Å}^2$. $V_{tip} = -2\,\text{V}$, $I_T = 0.5\,\text{nA}$, $A_0 = 0.5\,\text{Å}$ and $k_0 = 98\,\text{N/m}$, $f_{res} = 15094\,\text{Hz}$, $f_0 = 7501\,\text{Hz}$.

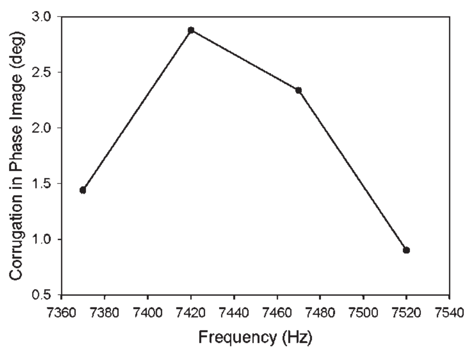

Fig. 4. Corrugation height in phase scans vs lever oscillation frequency.

at a frequency very close to half of the lever resonance frequency.

An interesting point is that the contrast in the simultaneous STM scans did not show any frequency dependence. This indicates that the change with frequency of phase corrugation cannot be tip trajectory dependent. It will therefore also not be related to changes in the overall interaction force. The fact that the peak occurs close to half the lever resonant frequency strongly suggests harmonic coupling into the lever resonance, and therefore into the macroscopic dissipation processes associated with the finite Q of the lever. This mechanism of course requires a non-linearity in the interaction—such a non-linearity is clearly present in these experiments from the (very non-linear) short range interatomic forces. An initial trial simulation based on models of AFM dynamics,[17,18] suggested that this non-linear coupling is a plausible mechanism to explain our results. More quantitative simulations are in progress. At this stage, therefore, we are not able to say how much of the measured phase signal is local, and how much coupling into the lever Q. However, it seems likely from the contrast away from half resonance that there is some finite, but small local component to the dissipation.

It is interesting to note that even with our oscillation amplitudes as small as 0.25 Å, the nonlinearity of the atomic interaction could give harmonic coupling. With larger amplitudes, much stronger coupling, and into significantly higher harmonics is possible. Care may be required in interpreting images, particularly of dissipation, if there are a significant number of resonances present in a large amplitude nc-AFM.

3. Conclusion

In summary, we obtained atomic resolution damping images of a Cu(100) surface using extremely small amplitudes. The use of small amplitudes means that the energy input to the interacting tip-sample system is relatively low and hence the perturbation is minimal. The images revealed energy dissipation values of up to 100 meV per cycle at atomic sites. The experiments on Si(111) surface showed that the frequency of operation affects the phase images, with a peak in phase corrugations at around the lever half resonance. The non-linearity of the tip-surface interaction can give this harmonic coupling. Avoiding sub-harmonic frequencies may be necessary in low amplitude off-resonance technique, if the goal is to measure local dissipative interactions. However, the fine control of this behaviour of phase at certain frequencies may also provide a new means of imaging in off-resonance AFM.

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