Determining the Film Thickness and Probing the Interface Structure with Characteristic Scanning Tunneling Spectroscopy

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(Received January 19, 2006)

Structural and electronic properties of atomic-scale flat Ag films grown on Si(111)7×7 are investigated with scanning tunneling microscopy and spectroscopy. The spectrum for each film thickness reveals features of transmission resonance and standing-wave states. The spectral intensity or energy levels of these features vary with the film thickness, causing the spectrum for each thickness to exhibit a unique fingerprint-like aspect. The spectra can thus be used to discern the film thickness. In addition, the spectrum reveals a slight intensity variation with the change of location on the film. This variation is due to a phase change in the electronic reflection at the buried interface. Using this effect, the buried interfacial structure can be probed as well.

PACS numbers: 73.21.Fg, 68.37.Ef, 73.61.At

The growth of thin metal films on semiconductor surfaces is a subject of great interest in science as well as in technological applications. It involves two main issues: (i) how to grow a metal film with atomic-scale flatness, and (ii) understanding the interfacial structure and electronic properties such as the Schottky barriers at the metal/semiconductor interface. For the first issue, some recent studies have demonstrated that the quantum size effect can be a driving force for forming flat metal films on semiconductor surfaces [1–5]. In these studies, scanning tunneling microscopy (STM) is a useful technique for investigating the flatness, the electronic structure, and the preferred thickness of the metal films. However, determining the film thickness with STM requires the exposure of a bare part of the semiconductor substrate or the metal wetting layer. In other words, as the film covers the entire substrate it becomes very difficult to precisely determine the film thickness. In this report, we demonstrate that this difficulty can be overcome by performing scanning tunneling spectroscopy (STS). We acquired the spectra of flat Ag metal films grown on Si(111)7×7 at the energy range of 2∼9 eV above the Fermi level. The spectrum for each film thickness reveals the features of both transmission resonance [6] and standing-wave states in the tunneling gap [7, 8]. Because the spectral intensities or energy levels of these features vary significantly with the film thickness, these spectra exhibit unique characteristics and can be viewed as the fingerprints of the film thickness. This allows us to determine the film thickness unambiguously even when the film covers the entire substrate. Recently, Upton *et al.* have demonstrated that the absolute thickness of Pb film grown on an Si(111) surface can also be precisely determined by observing the signals of quantum-well states

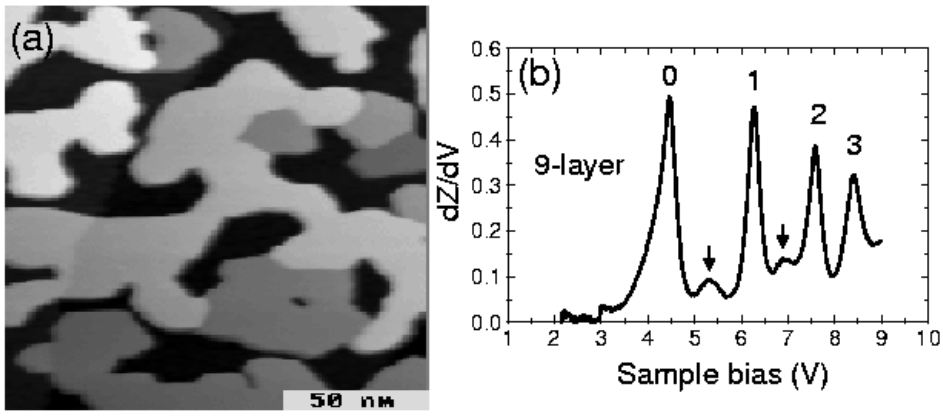

FIG. 1: (a) The growth of flat Ag films on Si(111)7×7 surface at room temperature at the coverage of 3.5 ML. The image size is 150×150 nm^2. (b) dZ/dV-V curve measured on the 9-layer Ag film. Peaks 0∼3 are standing-wave states. Bump features indicated by arrows are transmission resonances.

in photoemission spectroscopy [9]. In addition, we have also observed that the spectral intensity changes slightly with the measured location on the film. The spatial mapping of the spectrum intensity exhibits the 7×7 periodicity of the substrate, indicating that local variations of the spectral intensity originate from the interface properties. In other words, we can utilize this phenomenon to probe the structure of the metal/semiconductor interface. Although previous studies have demonstrated that STS can be used to study the metal/semiconductor interface, the signal used comes from the quantum-well states below the vacuum level [10–12]. Our emphasis here is to use signals from the transmission resonance and standing-wave states above the vacuum level.

In our experiment, a Si(111)7×7 surface was obtained by annealing the sample to 1200°C followed by slow cooling to room temperature. Silver was deposited onto the Si(111)7×7 surface at room temperature with a flux of 0.26 ML per minute. The vacuum during the deposition was kept below 2×10^{-10} torr. After deposition, the sample was transferred to a homebuilt STM in which the sample was cooled to 109 K. The electronic structures of the Ag films were probed by using Z-V spectroscopy for which the feedback is active; the tip trajectory was recorded while the sample bias was ramped from 2 to 9 volts. The spatial mappings of the spectral intensity were carried out by using a lock-in technique with a modulation of the sample voltage of 30 mV at a frequency of 5 KHz.

It is known that flat silver films with a (111) face can be grown on Si(111)7×7 at room temperature [13]. Figure 1(a) shows a typical STM topography image of the Ag film grown at room temperature with a coverage of 3.6 ML. At this coverage, about 0.35 ML of Ag was consumed to wet the Si(111)7×7 and the rest grew into a flat film on the (111) surface. The growth of the film is not layer-by-layer, thus several kinds of thickness can often be observed, and certainly the thickness increases with coverage. Among the crystalline films, the wetting

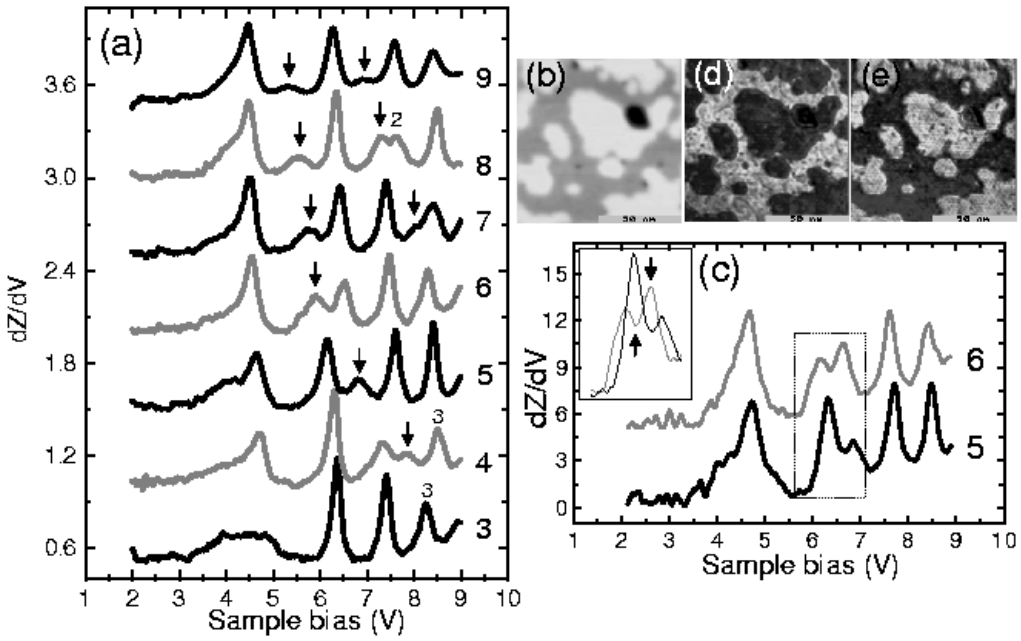

FIG. 2: (a) Spectra acquired on Ag films of different thickness. Numbers at the right side of the spectra mark the atomic layers of thickness. Arrows mark the transmission resonance. (b) Typical STM image of atomic-scale flat Ag films grown on Si(111) 7×7 at 110 K at the coverage of 5.5 ML followed by annealing to RT. Image size is 1000×1000 Å^2. (c) Gray and black curves are spectra acquired on light gray and dark gray regions in (b), respectively. The normalized curves in the inset are part (marked by a dashed rectangle) of both curves. The upward and downward arrows in the inset mark the biases for spatial mappings of the spectral intensity in (d) and (e).

layer is still exposed, thus allowing us to determine the film thickness. STS is used to take a precise Z-V spectrum on films of different thickness. The curve shown in Fig. 1(b) is a dZ/dV-V spectrum which is differentiated from the Z-V spectrum (not shown) measured on a film of 9-atomic layer thickness above the silicon substrate. The number of atomic layers is determined in terms of an interspacing of 2.7 Å [2]. Here we assume that the wetting layer under the film has been crystallized to an atomic layer. Because the work function of the thin Ag film on the Si(111) is 4.41 eV [14], the bump features indicated by arrows in the spectrum are above the vacuum level; they resulted from the transmission resonance [6]. In addition, peaks indicated by the numbers 0∼3 are the well-known standing-wave states in the tunneling gap of STM [7, 8]. These two kinds of signals originate from the fact that electrons impinging on a surface in free space have a chance to be transmited or to be reflected, which should also occur in the STM configuration. For transmission electrons, the quantum well in the silver film can significantly affect their transmissivity [15]. According to quantum mechanics, the probability for an electron transmitting through

a square potential well obeys the equation [16]

$$1/T = 1 + V^2 \sin^2(kt)/4E(E+V)\,, \tag{1}$$

where T is the transmission probability, E is the energy of the incident electrons, V is the depth of the potential well, t is the width of the well, k is the wave vector of an electron in the well, and

$$\hbar^2 k^2/2m = E + V\,. \tag{2}$$

It is obvious that T can be one when kt is equal to an integral multipe of π. At this condition, electrons can penetrate the film completely. This quantum phenomenon is termed transmission resonance, which is the origin of the bump features in the spectrum. For reflected electrons, they are unable to return to the vacuum in STM as they are in free space, since the potential well between the tip and the sample would confine them in the tunneling gap causing the wave vector of the reflected electrons in the surface normal direction to obey the Bohr-Sommerfeld quantization rule and form standing-wave states. That causes the observed spectrum to exhibit peak features. Therefore the spectrum of a 9-layer thick film contains information of two kinds of quantum phenomena, and they also appear in the spectra of other thicknesses. Fig. 2(a) shows the dZ/dV-V spectra acquired on the Ag film of 3∼9 layers. The numbers at the right hand side of each curve mark the film thickness. In each spectrum, except for the spectrum of 3-layer thick film, there appears a transmission resonance marked by arrows. By comparing the intensity distribution of each spectrum, it is obvious that each one is of a unique aspect. The factors for forming these characteristic spectra are the following. First, the transmission resonance moves toward the vacuum level with increasing film thickness. Furthermore, in the spectra of 7∼9-layer thick films, another transmission resonance of higher energy is revealed. Second, there are four standing-wave states in each spectrum. However, the intensity of each standing-wave state is not always much higher than that of the transmission resonance, as is the situation in the spectrum of 9-layer thick film. For example, the intensity of the second standing-wave state, as marked by 2 in the spectrum of 8-layer thick film, is obviously much smaller than that of other states but is comparable to that of the transmission resonance [17]. In our observation, this is due to the intensity of the standing-wave state decreasing with decreasing separation between it and the transmission resonance. Third, Kubby *et al.* have demonstrated that the appearance of transmission resonance can displace the higher order standing-wave states to higher energy [6], which also appears in our spectra. For example, because of the appearance of the transmission resonance in the spectrum of 4-layer thick film, the energy level of the third standing-wave state as marked by 3, is higher than that in the spectrum of 3-layer thick film. These factors influence the distribution of the spectral intensity, thus Fig. 2(a) can be used as a data base of the fingerprints for determining the film thickness. Therefore, besides measuring the film thickness directly from the height distribution of STM images, one can also distinguish the thickness by acquiring dZ/dV-V spectra for films and comparing them with the spectra in the data base. This is especially useful for Ag films covering the entire wetting layer. Previous studies have shown that Ag

can be grown into a complete film on the Si(111)7×7 by low temperature deposition followed by annealing to room temperature [2]. Fig. 2(b) shows an STM image of Ag film formed in this way. Using STM, we can only determine that the height difference between the light gray and the dark gray region is one atomic layer, but cannot determine the absolute thickness of both regions above the substrate. The black and gray curves in Fig. 2(c) are the spectra acquired on dark gray and light gray regions, respectively. From a comparison with the spectra of Fig. 2(a), it is easy to tell that the black and gray curves are the spectra of 5-layer and 6-layer thick film, respectively. The thickness of the dark gray and light gray regions can thus be unambiguously identified. The inset in Fig. 2(c) shows the normalized curves, which are the parts (marked by a dashed rectangle) of both curves. In the inset, the intensity of the 5-layer curve (black) at 6.3 V as indicated by upward arrow is higher than that of the 6-layer curve (gray), resulting in the spatial mapping of the spectral intensity at 6.3 V for the 5-layer thick area in Fig. 2(b) exhibiting a bright region, as shown in Fig. 2(d). On the other hand, Fig. 2(e) shows a reverse contrast, because the voltage of the mapping is 6.7 V at which, as indicated by downward arrow, the intensity of the 6-layer curve is higher than that of the 5-layer curve in the inset. Although the spectra exhibit a thickness-dependent behavior, actually their aspects can be tuned by adjusting the tunneling current, i.e., the electric field between the tip and sample. The main alternation in the spectra is that the amount of the standing-wave states varies with the electric field. In addition, the acquired spectra by different tips may not be alike at the same tunneling current. In our observation, however, the spectra always can be tuned to be almost identical to that of Fig. 2(a) at the criterion of the appearance of four standing-wave states. Therefore, the goal of identifying film thickness by the spectra still can be achieved.

When we acquired the spectra of a film, we often observed that the spectral intensity changes slightly with the measured location but this does not affect the determination of film thickness. Fig. 3(a) shows two spectra acquired at two locations on the 5-layer thick film, revealing visible intensity differences. In order to find the origin of the local variation of the spectra, we performed a spatial mapping of the spectral intensity. Fig. 3(b) shows the mapping for 6.35 V at which, as marked by a dash line, the intensity distributions of the two spectra in Fig. 3(a) are different. It is interesting to see that Fig. 3(b) exhibits a hexagonal lattice with a period of $\sim$ 27 Å, in agreement with that of the Si(111)7×7 reconstruction. Therefore, the variations of the spectral intensity with location essentially originated from the Ag/Si(111)7×7 interface properties. In other words, we can utilize this location-dependent spectrum to probe the interface structure nondestructively. For example, the periodic pattern in Fig. 3(b) can be correlated to a change of the electronic reflection phase at the buried interface, which is ultimately related to the local variation of the Schottky barrier. Therefore it is possible to extract the spatial variation of the Schottky barrier from location-dependent spectra combined with a theoretical calculation. In addition, Fig. 3(b) also exhibits non-periodic structures, as marked by the arrows. They may represent some defects or metal clusters formed during deposition. These structures would scatter electrons and affect the transport behavior of electrons between the metal film and the semiconductor. Thus, using the location-dependent spectra, one can monitor the interface quality and understand its influence on the electron transport.

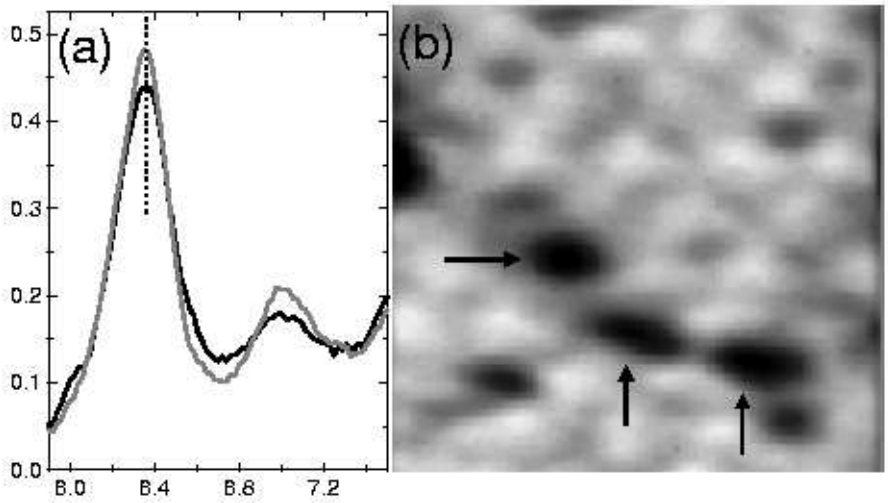

FIG. 3: (a) Spectra revealing a visible difference, acquired at two locations on the 5-layer thick film. (b) The spatial mapping of intensity of 6.35 V at which, as marked by a dashed line, the intensities of the two spectra in (a) are different. A hexagonal lattice with a period of ~ 27 Å and non-periodic structures, as marked by arrows. Image size is 12×12 nm^2.

In summary, we have demonstrated that STS spectra acquired on flat Ag films grown on Si(111)7×7 exhibit some thickness-dependent and location-dependent characteristics. They allow us to unambiguously determine the film thickness and also to probe the Ag/Si(111)7×7 interfacial structure. We believe that these characteristics can be applied to most flat metal films on semiconductor substrates. Therefore, the techniques we present here should be useful in technological applications.

The authors would like to acknowledge C. S. Kuo, C. Y. Lin, and C. H. Hsieh for supporting the construction of STM. This work was supported by the National Science Council and the Academia Sinica and the project of Academic Excellence of Education Ministry of Taiwan.

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[17] This transmission resonance can be distinguished by tuning the energy level of the standing-wave state 2, which can done by adjusting the tunneling current. When its energy level is changed, its spectral intensity would be larger than that of the transmission, allowing us to identify both of them.