Interplay between transmission background and Gundlach oscillation in scanning tunneling spectroscopy

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It is known that when the electronic structure beyond the vacuum level of a metal film is probed by scanning tunneling spectroscopy, both transmission resonance and Gundlach oscillation may be revealed in the tunneling spectrum. The manifestation of transmission resonance implies that there exists a transmission background involving the information about the electron transmissivity in the spectrum, superposing with the Gundlach oscillation. In this paper, it is shown that the spectral intensity of the Gundlach oscillation can vary with observed location on a Ag/Si(111)7×7 surface and a reconstructed Au(111) surface. The variation can naturally be attributed to the local change of electron transmissivity. A general phenomenon is also observed that the total intensity in the range of the Gundlach oscillation is conserved, although its intensity distribution is location dependent. By inspecting the Gundlach oscillation on the Ag(100) surface, it is demonstrated that the transmission background can be correlated to the projected bulk band structure.

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I. INTRODUCTION

Gundlach oscillation[1] observed with scanning tunneling spectroscopy (STS) is a phenomenon of field-emission resonance through standing-wave states in the tip-sample gap.[2,3] Each standing-wave state involves states of electron with momentum parallel to the surface, which has been confirmed by Wahl *et al.*[4] from observation of the interference pattern near the step. Therefore, each Gundlach oscillation represents a subband. Although Gundlach oscillation is a general phenomenon that can be observed on metal surfaces, in comparison with the steplike feature of the Gundlach oscillation in *Z-V* spectrum acquired on Ni(100),[2] it is found that the one acquired on Au(110) is much weaker.[3] This implies that the strength of the Gundlach oscillation significantly depends on the species of the metal, but, to our knowledge, its origin remains to be elucidated.

Gundlach oscillation is not the only phenomenon manifested in the tunneling spectrum when the surface of thin metal film is observed. Kubby *et al.* observed an additional signal superposed with Gundlach oscillation on a $\sqrt{3}\times\sqrt{3}$ Sn/Si(111) surface.[5] Recently, we have also detected a similar signal on Ag films grown on the Si(111)7×7 surface, and it has been identified as the transmission resonance because it reveals obvious thickness-dependent behavior.[6] Transmission resonance is a quantum phenomenon of electrons scattered by the quantum well in a metal film.[7] It originates from the interference of the electron waves, which are reflected from the film surface and the film-substrate interface. Before the work of Kubby *et al.*, the transmission resonance had in fact been observed using low-energy electron transmission spectroscopy (LEETS) (Ref. 8) for some free-electron-like metal films such as Ag and Cu films.[9,10] In LEETS and STS, electrons with energy matching levels of the transmission resonance can be of higher transmissivity and spectral intensity, respectively. Since both kinds of spectroscopy can be used to observe transmission resonance, it is natural to consider that the information about the transmissivity varied with electron energy should be involved in the tunneling spectrum. Based on this realization, we propose in this paper a viewpoint that in the tunneling spectrum there exists a transmission background which intensity is equivalent to the transmissivity,[11] superposing with Gundlach oscillation. In addition, because of the wave nature, the electron impinging on the surface of the metal, no matter if it is the thin film or the bulk, has a probability to transmit. Therefore, the viewpoint is applicable to Gundlach oscillation observed on the bulk surface as well. In this work, we will demonstrate that the spectral intensity of the Gundlach oscillation may change with observed location on the Ag/Si(111)7×7 surface and reconstructed Au(111) surface. According to the proposed viewpoint, we can explain that this location-dependent Gundlach oscillation is due to the variation of the transmission background, and is ultimately related to local electron transmissivity. In addition, in these two studied systems, we have found that the total intensity in the range of the Gundlach oscillation is conserved, although its intensity distribution is location dependent. This observation leads to the possibility of understanding why the strength of Gundlach oscillation significantly depends on the species of the metal. Furthermore, we demonstrate that the transmission background can be correlated to the density of states in the projected bulk band structure by inspecting Gundlach oscillation on the Ag(100) surface.

II. EXPERIMENT

In our experiment, Gundlach oscillation was studied for three kinds of metal surface. To prepare the Ag film on a Ag/Si(111)7×7 surface, silver was deposited onto the surface at room temperature with a flux of 0.26 ML per minute. After deposition, the sample was transferred to a homebuilt scanning tunneling microscope (STM) operating at 109 K. To prepare clean Au(111) and Ag(100) surfaces, samples were treated with cycles of ion beam sputtering and annealing to 600 °C. The Au(111) and Ag(100) surfaces were then studied using another homebuilt STM operating at 4.3 K.

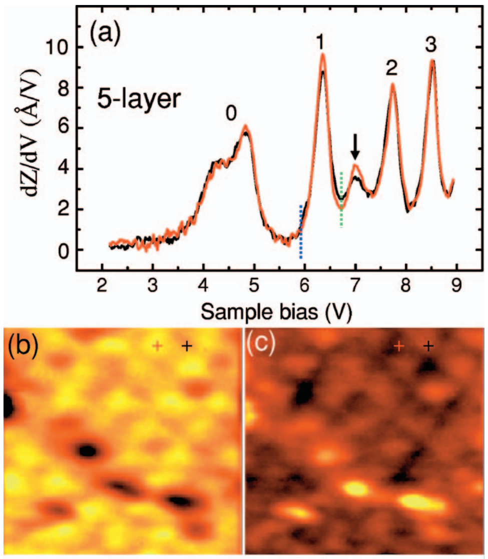

FIG. 1. (Color) (a) Spectra acquired at two locations on the five-layer film, revealing visible differences. The blue and green dashed lines mark the onset and end of peak 1 in the black and red curves, respectively. The arrow marks the transmission resonance. (b) and (c) show the mappings at the apex (6.35 eV) and the end (6.72 eV) of peak 1, respectively. Crosses mark the locations where the spectra were acquired. The image size is 120×120 Å^2.

Gundlach oscillation was observed using *Z*-*V* spectroscopy. For *Z*-*V* measurement, the tip trajectory was recorded with an active feedback, while the sample bias was ramped from 2 to 9 volts. The dZ/dV-V spectrum was obtained by numerical differentiation of the *Z*-*V* spectrum. The spatial mapping of intensity of dZ/dV-V spectrum at a specific energy was acquired by the numerical method or by combining the lock-in amplifier with a 30 mV bias modulation at 5 kHz. The frequency is too high for feedback loop to react, and therefore the output quantity from the lock-in amplifier is dI/dV. It does not correspond to dZ/dV but it can be representative of dZ/dV. Hence, the peak positions of Gundlach oscillation and transmission resonance in a spectrum obtained by lock-in amplifier are the same as those in the dZ/dV-V spectrum.

III. RESULTS AND DISCUSSION

In our previous study,[6] we have shown that both Gundlach oscillation and transmission resonance can be revealed in the tunneling spectrum taken on the Ag film grown on the Si(111)7×7 surface. Figure 1(a) shows two dZ/dV-V spectra acquired at two locations on a five-layer Ag film. The signal indicated by an arrow is the transmission resonance. The distinct peaks marked by numbers are the Gundlach oscillations, and depending on the potential well for forming these peaks, they can be classified. The potential well in the tunneling gap is a superposition of the image potential and the electrostatic potential.[2] The actual shape of this well varies with the applied voltage while the *Z*-*V* spectroscopy is being performed. When the Fermi level of the tip is close to the vacuum level of the sample, contribution of the image potential is significant. The superposition of the image potential and the electrostatic potential forms a specific potential well, and peak 0 is the Gundlach oscillation related to a standing-wave state in this well. When the Fermi level of the tip is higher than the vacuum level of the sample, the image potential becomes negligible, and the potential well can be approximated to be of triangular shape. Referring to the work function of the Ag film on Si(111) being 4.41 eV,[12] peaks 1, 2, 3 are Gundlach oscillations above the vacuum level and can be classified into a group. In this paper, we focus only on Gundlach oscillation above the vacuum level because the correlation between its intensity and the transmissivity of the free electron is concerned. Since each Gundlach oscillation is a subband, its onset and end can be accurately determined provided that there is no overlap with a nearby Gundlach oscillation. Peak 1 in Fig. 1(a) is such a Gundlach oscillation, its onset and end are marked by blue and green dashed lines, respectively.

The spectra shown in Fig. 1(a) display visible intensity differences at the transmission resonance, the end and the apex of the peak 1. In order to find the origin of the local variation of the spectra, we performed the spatial mapping of the spectral intensity using the lock-in technique. Figures 1(b) and 1(c) show such mappings at the apex and the end of peak 1. Figure 1(c) has a reversal contrast of Fig. 1(b) because the spectral intensity of two curves at the end of peak 1 is reversed. Crosses mark the locations where the spectra were acquired, and their colors denote the corresponding spectra in Fig. 1(a). Interestingly, it can be seen that Fig. 1(b) exhibits a hexagonal lattice with a period of about 27 Å, in agreement with that of the Si(111)7×7 reconstruction. Therefore, the variations of spectral intensity with locations essentially originate from the Ag/Si(111)7×7 interface property. According to the proposed viewpoint, this indicates that the electron transmissivity at the transmission resonance may change with the location due to the modulation of interface property. That is, the local variation of potential barrier affects the electron reflection phase at the buried interface, causing the transmittivity of electrons to vary with the location. This local variation of transmissivity also correspondingly emerges in the transmission background of peak 1. The intensities at the onset of peak 1 in both curves are the same but the intensity at the end of the peak 1 in the black curve is larger than that in the red curve. It is plausible to expect that the transmission background in the range of peak 1 in the red curve cannot be higher than that in the black curve. In addition, we measured the total intensity of peak 1 for both curves, and the result indicates that the total intensity of peak 1 is conserved though its intensity distribution is different in each curve. The intensity distribution of the peak is the superposition of the transmission background and Gundlach oscillation. Since the total intensity of the transmission background of peak 1 in the black curve is larger than that in the red curve, the total intensity of the Gundlach oscillation of peak 1 in the red curve should be larger due to conservation of the total intensity. This results in the apex of peak 1 in the red curve having a greater intensity. Therefore, the total intensity of the transmission background is complementary with that of the Gundlach oscillation. Next, we will show that this complementary phenomenon should be general because it also appears on the Au(111) surface.

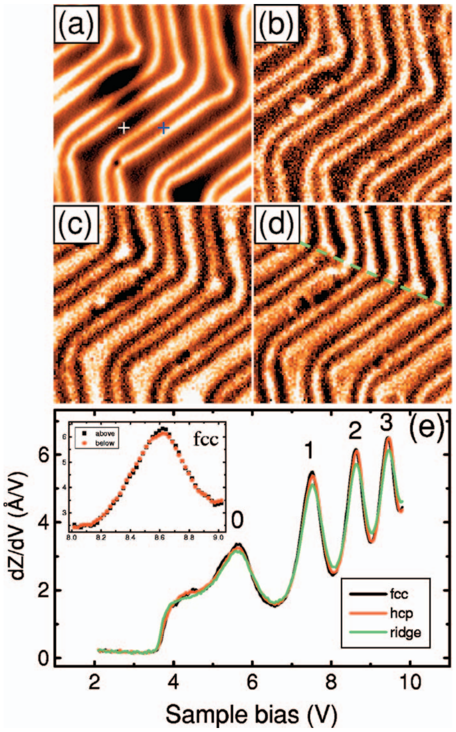

FIG. 2. (Color) (a) Topographical image of a reconstructed Au(111) surface. The image size is 286×286 Å^2. (b) Mapping at the valley (8.06 eV) between peak 1 and peak 2 in (e). (c) and (d) are mappings at the apexes of peak 1 (7.53 eV) and peak 2 (8.64 eV) in (e), respectively. (e) The average dZ/dV-V spectra of the fcc, hcp, and ridge regions. The inset shows peak 2 at fcc regions above and below the green dashed line in (d). The pixels of topographical image in (a) are 500×500. The pixels of mappings in (b), (c), and (d) are 100×100. The mappings are smoothed by the interpolation.

Figure 2(a) shows a typical topographical image of an Au(111) surface with the well-known $22 \times \sqrt{3}$ herringbone reconstruction.[13] In this structure, the bright ridge lines separate the surface into the hcp and the fcc regions, as marked by blue and white crosses in Fig. 2(a), respectively. Recently, Chen *et al.* have demonstrated that the electronic structure of the hcp region differs from that of the fcc region. They observed that there is a striking intensity difference between the two regions in the tunneling spectra near the surface-state band edge.[14] This observation prompted us to investigate whether the transmission background of two regions is different. We used STS to acquire Z-V spectra and topographical image simultaneously in order to obtain the spectrum at a precise position. Figure 2(e) shows the average dZ/dV-V spectra taken at the fcc, hcp, and ridge regions. In each spectrum, there are four peaks of Gundlach oscillation and we will focus on peaks 1, 2, 3. Local variations of spectral intensity are exhibited in these peaks, but this differs from the result of Chen *et al.*, where the spectral intensity of the ridge region was found to be significantly different from that of the fcc and hcp regions. Due to the fact that adjacent peaks overlap, the onset and the end of each peak cannot be determined as the peak 1 in Fig. 1(a). In our measurements, the energy levels of the valleys between peaks and apexes of the peaks

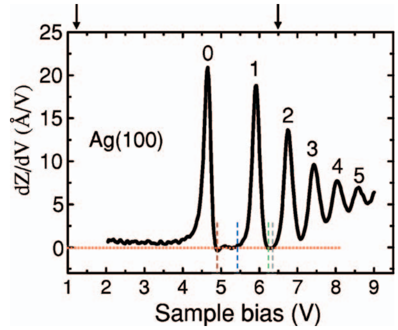

FIG. 3. (Color) The dZ/dV-V spectrum acquired on an Ag(100) surface. Downward arrows mark the range of the band gap. Red, brown, blue, green, and gray dashed lines mark the level of zero intensity, the end of peak 0, the onset of peak 1, the end of peak 1, and the onset of peak 2, respectively.

for the three regions are the same. However, the intensities at the valleys for the ridge region are all higher than those for the fcc and hcp regions. This implies that the transmission background of the ridge region is always higher than those of the fcc and hcp regions. According to the proposed viewpoint, we can infer that the ridge region has the highest electron transmissivity on the reconstructed Au(111) surface. This local variation of transmissivity can reflect on the mapping of the spectral intensity, which is simply the spatial variation of the numerical value of dZ/dV at a specific bias. Figure 2(b) shows the spectral mapping at the valley between peak 1 and peak 2. It turns out that the bright area (higher intensity) in Fig. 2(b) corresponds to the ridge region in Fig. 2(a). Figure 2(e) also reveals the same phenomenon as in Fig. 1(a): the peak with higher transmission background has lower intensity at the apex, and vice versa. This indicates that complementary phenomenon also manifests on the Au(111) surface. Figure 2(c), the spectral mapping at the apex of peak 1, thus clearly exhibits a reversal contrast of Fig. 2(b) as expected. Moreover, Fig. 2(c) reveals that the contrast at the fcc region is slightly brighter than that at the hcp region, consistent with the spectral intensities of two regions at the apex of peak 1, shown in Fig. 2(e). Careful identification of the intensities at the valleys around peak 1 indeed verifies that the transmission background in the range of peak 1 at the hcp region is slightly larger than that at the fcc region, again showing the complementary phenomenon. Figure 2(d) shows the spectral mapping at the apex of peak 2. It exhibits that fcc and hcp regions above the green dashed line are brighter than those below it. This can also be verified by the spectra provided in the inset of Fig. 2(e), where only spectra at fcc regions are shown. It implies that transmissivity also changes with the orientation of the herringbone reconstruction, which may result from the inhomogeneous relief of local strain field on the Au(111) surface. We have also observed the complementary phenomenon on the 9×9Ag/Cu(111) (Ref. 15) and 11×11 Pb/Si(111) (Ref. 16) superstructures, and therefore believe that the complementary phenomenon is a general one.

Figure 3 demonstrates the dZ/dV-V spectrum acquired on a Ag(100) surface, in which six peaks of Gundlach oscillation were observed, as marked by numbers. All peaks are above the vacuum level except peak 0, referring to the work function of 4.64 eV on the Ag(100) surface. Peak 1 is the one we are interested in because the spectral intensities are zero at its onset and end, marked by blue and green dashed lines, respectively. This indicates that the intensity of the transmission background is also zero in the distribution of peak 1, and therefore peak 1 is a pure Gundlach oscillation without transmission background. Brown and gray dashed lines mark the end of peak 0 and the onset of peak 2. It can clearly be seen that the spectral intensity is zero in the range between the brown and blue dashed lines, as well as in the one between the green and gray dashed lines. According to the proposed viewpoint, we can therefore conclude that the electron transmissivity is zero in the energy range between brown and gray dashed lines. This region without transmission background is in the band gap of the projected bulk band structure of the Ag(100) face.[17] Two downward arrows in Fig. 3 mark the range of the band gap. Previous studies have demonstrated that the transmissivity of the free electron impinging on some crystal surfaces is reduced when the electron energy coincides with the energy level of the band gap in the band structure.[18] This implies that the density of states would be the channel for electron transmission because it is zero in the band gap, causing the decreasing of the transmissivity. Therefore, the transmission background with zero intensity in the spectrum essentially originates from the band gap. In other words, it is plausible to consider that the transmission background can directly reflect the electronic density of states at the surface. Based on this opinion, the transmission resonance may originate from the existence of an oscillatory distribution of the density of the states in the electronic structure of the metal film. The transmission resonance corresponds to the energy level at which the density of states is at a maximum. Peaks 3, 4, 5 are above the band gap, and thus each of them has a nonzero transmission background. Moreover, we note that the intensities at the apex of peaks 3, 4, 5 are much smaller than that at the apex of peak 1. This reveals the complementary phenomenon again, i.e., the peak without transmission background has the strongest intensity at the apex. Since the transmission background can be correlated to the density of states of the surface, it is why the strength of the Gundlach oscillation significantly depends on the species of the metal.

Although the density of states can be probed using STS, that can be well described by the models of metal-vacuum-metal tunneling[19,20] only when the applied bias is in the range between −2–2 volts. In our experiment, we have applied the sample bias above the vacuum level of the sample to probe the transmission background. The energy range is beyond the range that those models are applied to. In addition, Becker *et al.*[3] has mentioned that if the applied bias exceeds the work function of the sample, field emission will appear on the tip. Our recent study also shows that the electron tunneling starts to enter the field emission regime when the sample bias exceeds 3.5 V.[21] Therefore, the manifestation of the transmission background in the tunneling spectrum cannot be described by the model of metal-vacuum-metal tunneling. A more sophisticated model including the factors of field emission is needed.

IV. CONCLUSIONS

In summary, based on the fact that the transmission resonance can be probed using the STS, we propose a viewpoint that there exists a transmission background which intensity is equivalent to the electron transmissivity, superposing with the Gundlach oscillation. Utilizing this viewpoint, we explain that the spectral intensity of the Gundlach oscillation varied with the location on a metal surface is due to the local variation of electron transmissivity. Gundlach oscillation is the quantized state existing in the vacuum, i.e., in the tip-sample gap. Intuitively, its density of states should be independent of the density of states associated with the surface. However, our experimental results reveal a general phenomenon that the total spectral intensity of the Gundlach oscillation is complementary with that of the transmission background. In addition, we also show that the transmission background can be related to the projected bulk band structure. This implies that the intensity of Gundlach oscillation still conveys the details of local electronic structure on the surface. Therefore, it seems that there exists the counterpart providing the density of states for the Gundlach oscillation in the electronic structure. We hope these observations can inspire the theorist to investigate this counterpart with a more fundamental approach.

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