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Abstract

We studied wave function dissipation (WFD) in field emission resonance (FER) by performing scanning tunneling microscopy on the highly oriented pyrolytic graphite (HOPG) and Ag(111) surfaces under two conditions: (1) the same current and FER number; (2) the same tip structure but different currents. Under the first condition, we observed that the decay rate corresponding to the WFD exhibited a larger variation on the HOPG surface than it did on the Ag(111) surface. Under the second condition, the decay rate was nearly independent of the FER electric field for the Ag(111) surface; by contrast, it was linearly proportional to the FER electric field for the HOPG surface. These remarkable differences can be attributed to the factors that the tip-induced attractive deformation caused by the electrostatic force was considerably more prominent on the HOPG surface than on the Ag(111) surface and that the deformed HOPG top layer had a unique electronic structure similar to that of single-layer graphene.

1. Introduction

An electric field of 0.1–0.3 V/Å in the junction of scanning tunneling microscope (STM) is inherently intrusive to the surface properties of materials. For example, this strong electric field can perturb electronic states such as the surface state, quantum well state, and transmission resonance, causing shifts in the state energy [1–3]. Moreover, this electric field can induce a local expansion deformation in Pb films through the STM tip [4], engendering a tip-induced attractive deformation (TIAD) caused by the corresponding electrostatic force. This TIAD is greater for the suspended graphene [5, 6]. Therefore, a TIAD can be expected on the graphite surface beneath an STM tip because of the weak van der Waals force between atomic layers of graphite. Nevertheless, this has never been detected, possibly because no tool is available for TIAD monitoring. A previous study observed giant corrugation of carbon atoms on a graphite surface by using an STM, which is due to the deformation induced by repulsive interatomic forces [7]. Moreover, through the deformation due to the contact between the atomic force microscopy tip and graphene, atomic-scale frictional characteristics of graphene can be resolved [8, 9].

Field emission resonance (FER) [10] is a quantum phenomenon occurring in an STM junction when a negative and high bias voltage is applied to the STM tip [11, 12]. Although FER originates from electrons tunneling into quantized states in a vacuum, its features such as energy, intensity, linewidth, and number, can provide information about the properties of the surface and STM tip. Consequently, FER has been a versatile technique to explore various phenomena [13–39]. For example, the FER number can reflect the sharpness of an STM tip; a higher FER number indicates a sharper tip [17, 35, 36]. In addition, previous studies demonstrated that the FER observed on the reconstructed surfaces revealed spatial variation in linewidth [32–34]. This

linewidth variation can be attributed to the local change of electron transmissivity [34]. These studies have motivated us to investigate whether electron transmissivity varies according to the TIAD on the graphite, and therefore, the linewidth is sensitive to the TIAD. For this exploration, a material whose surface does not deform easily was required. Ag(111) was selected because the metallic bond is considerably stronger than a van der Waals force. Our exploration results revealed that at the same FER number, the linewidth of first-order FER exhibited a significantly larger variation on the graphite surface than it did on the Ag(111) surface, indicating that FER can be used to monitor the TIAD of graphite. To further verify this finding, we observed the variation in linewidth with increasing current for the graphite and Ag(111) surfaces. Furthermore, our density functional theory (DFT) calculations demonstrated that the TIAD of the graphite surface could transform the electronic properties of the graphite top layer to be similar to those of the graphene, leading to variations in electron transmissivity with the TIAD level.

2. Experiment and calculation details

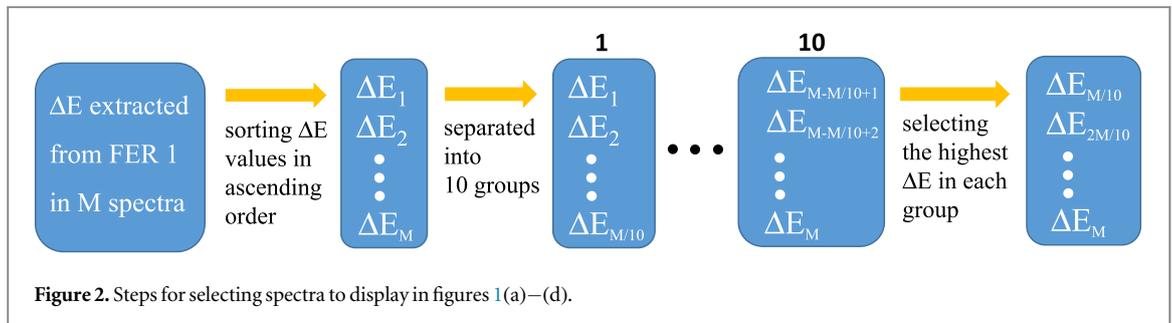
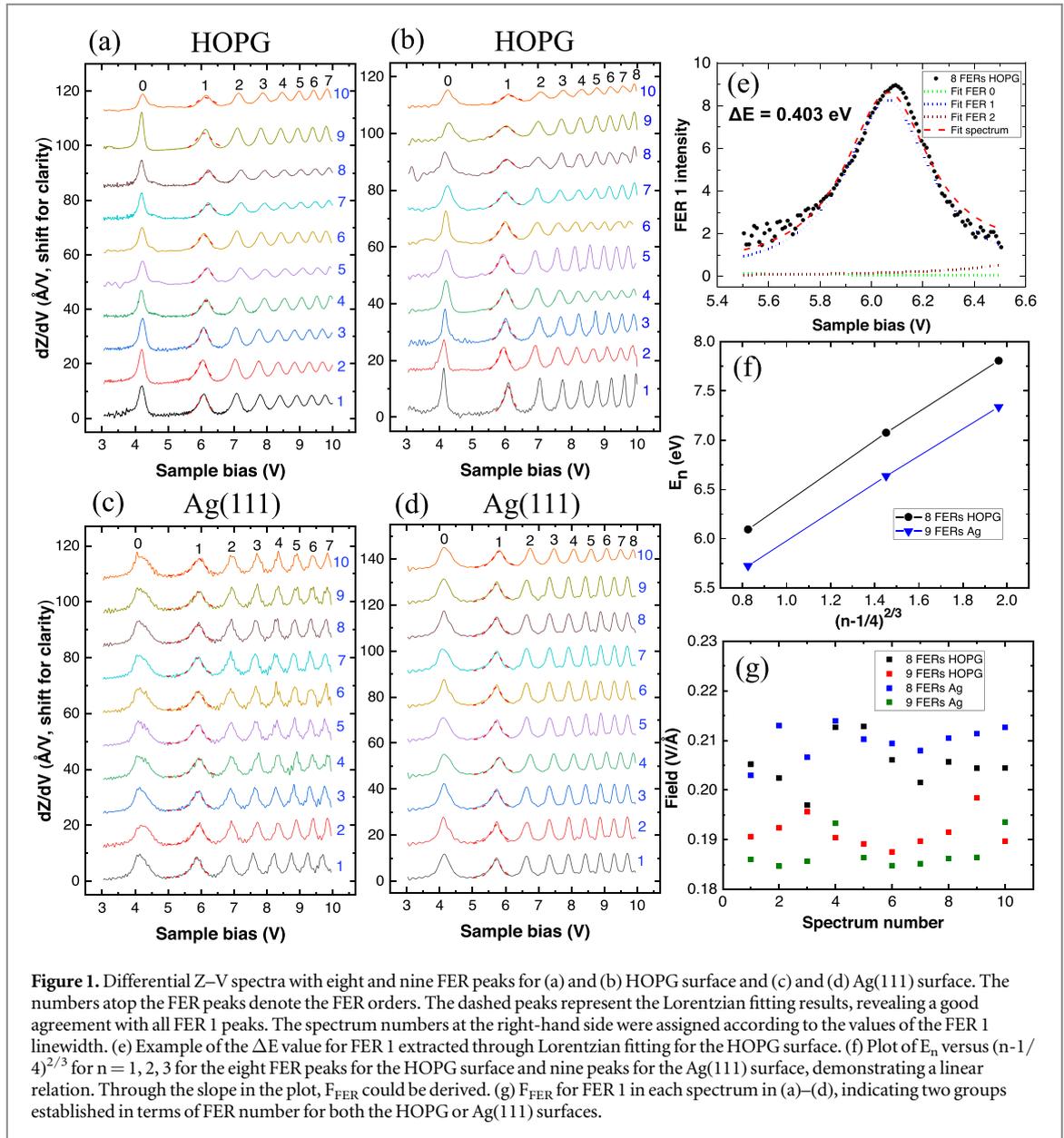
A clean graphite surface was prepared by cleaving the highly oriented pyrolytic graphite (HOPG) in air by using adhesive tape. A clean Ag(111) surface was prepared using ion sputtering followed by annealing at 600 °C for several cycles. An ultra-high-vacuum STM operated at 78 K or 5 K was used to observe FER on HOPG and Ag(111) surfaces. The FER detection was performed in Z–V (distance–voltage) spectroscopy by using Pt–Ir tips. For a Z–V measurement, the variation of the distance between the tip and surface was recorded with an active feedback, while the bias voltage was ramped from 3 to 10 V. The acquired Z–V spectra were differentiated using a numerical method to reveal the FER peaks.

The electronic structures were calculated using the projector augmented wave approach, as implemented in the Vienna *Ab initio* Simulation Package based on DFT. The Perdew–Burke–Ernzerhof form of the generalized gradient approximation was used for the exchange–correlation functional. The energy convergence threshold was set as 10^{-4} eV in self-consistent field calculations, and the energy convergence threshold was set as 10^{-3} eV for the structure optimizations. The lattice constant for Ag was determined to be 4.16 Å by minimizing the energy of the cell volume. Regarding the calculations performed for Ag, a $12 \times 12 \times 12$ k-point sampling grid was used to achieve energy convergence to within 1 meV/atom. To achieve high accuracy, we used a $24 \times 24 \times 24$ k-point grid to calculate the density of states (DOS) along the band symmetry points of Γ and L. Because the lattice constant for HOPG could not be obtained by minimizing the energy of cell volume, we set the experimental lattice constant a_0 to 2.46 Å for both HOPG and graphene and set c_0 to 6.78 Å for HOPG, in accordance with a previous study [40]. Regarding the calculations performed for HOPG, an $8 \times 8 \times 8$ k-point sampling grid was used to achieve energy convergence to within 1 meV/atom, and a $15 \times 15 \times 15$ k-point grid was used to achieve high accuracy in the DOS calculations. For the graphene calculations, an $11 \times 11 \times 1$ k-point sampling grid was used to achieve convergence. Moreover, a $15 \times 15 \times 1$ k-point grid with a vacuum thickness of 40 Å was used to achieve high accuracy in the DOS calculations.

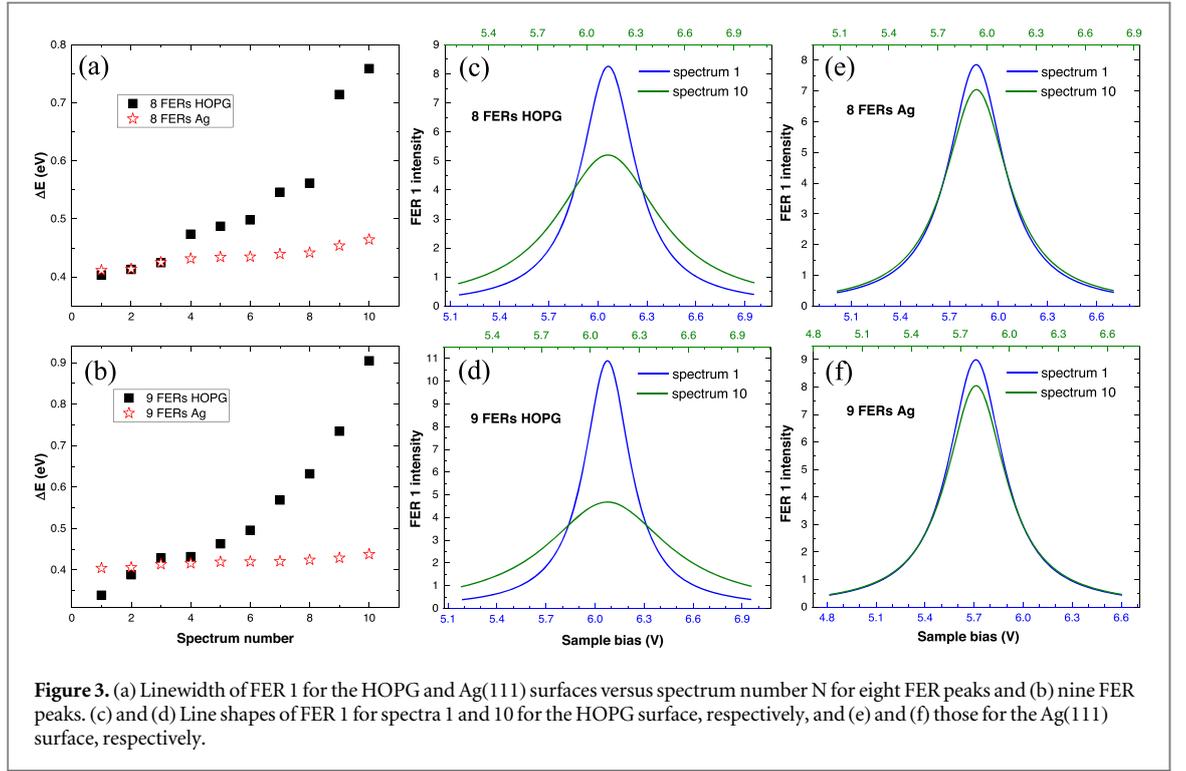
3. Results and discussion

In the experiment, FER spectra were first acquired by repeatedly performing Z–V spectroscopy at 78 K and 30 pA on the HOPG and Ag(111) surfaces. Because a high voltage of up to 10 V was applied, the sharpness of the tip may change, causing variations in FER number. Moreover, the repeated execution of Z–V spectroscopy could lead to the destruction of the tip apex, resulting in the disappearance of FER peaks. When this situation occurred, voltage pulses were applied to sharpen the tip or replacing it with a new one. Thus, spectra with different FER numbers were obtained.

Because the work function of Ag(111) (4.74 eV) is close to that of HOPG (4.7 eV) [41], the same FER number indicates that the sharpness levels of STM tips on HOPG and Ag(111) were close. Therefore, we collected spectra of the same FER number on HOPG and Ag(111) surfaces. Figure 1 displays differential Z–V spectra with eight and nine FER peaks for HOPG [figures 1(a) and (b), respectively] and Ag(111) [figures 1(c) and (d), respectively]. The numbers indicated above peaks denote the FER orders. We focused on the linewidth ΔE of first-order FER (named FER 1 hereafter), which was extracted through Lorentzian fitting, as an example of FER 1 on the HOPG shown in figure 1(e). The dashed peaks in figures 1(a)–(d) are Lorentzian fittings, showing a high level consistency with all the FER 1 peaks. In our analysis, as illustrated in figure 2, the extracted linewidth values of FER 1 in spectra with the same FER number were sorted in ascending order and then separated into 10 groups. In each group, the spectrum with the highest ΔE value for FER 1 was selected to be displayed in figures 1(a)–(d). The numbers on the right-hand sides of the spectra were assigned according to the highest ΔE values in 10 groups. A larger number N in figures 1(a)–(d) corresponds to a higher ΔE value.



Although the overall potential in the STM junction along the surface normal is not linear [36], a previous study demonstrated that the linear potential is a good approximation for the formation of FER quantized states of order $n = 1, 2, 3$ so that the electric field F_{FER} is constant for these three states [38]. The corresponding FER energies E_n can be expressed with the energies of quantized states in the triangular potential well [42] as follows:



$$E_n = E_{\text{vac}} + \alpha F_{\text{FER}} \frac{2}{3} \left(n - \frac{1}{4} \right)^{\frac{2}{3}}, \quad (1)$$

where E_{vac} is the vacuum level and

$$\alpha = \left(\frac{\hbar^2}{2m} \right)^{\frac{1}{3}} \left(\frac{3\pi e}{2} \right)^{\frac{2}{3}}. \quad (2)$$

Thus, the plot of E_n versus $(n-1/4)^{2/3}$ is linear for $n = 1, 2, 3$, as displayed in figure 1(f) for 8 FER peaks on HOPG and 9 FER peaks on Ag(111). F_{FER} can be obtained from calculating the slope in the plot divided by α to the power of $3/2$. Figure 1(g) presents the F_{FER} values for FER 1 in the spectra displayed in figures 1(a)–(d); as indicated in this figure, two separate groups were observed in terms of FER number for both the HOPG and Ag(111) cases. On average, F_{FER} value for the eight FER peaks was higher than that for the nine peaks and at the same FER number, the F_{FER} value for the HOPG case was similar to that for the Ag(111) case. Figure 3(a) exhibits that for the spectra with eight FER peaks, the FER 1 linewidths on both HOPG and Ag(111) increased with spectrum number N ; nevertheless, the ΔE variation between spectra 10 and 1 for HOPG was considerably larger than that for Ag(111). This difference was also observed in the spectra with nine FER peaks [figure 3(b)]. Consequently, the line shapes of FER 1 in spectra 1 and 10 for HOPG were clearly different [figures 3(c) and (d)], but those for Ag(111) only had slight changes [figures 3(e) and 3(f)].

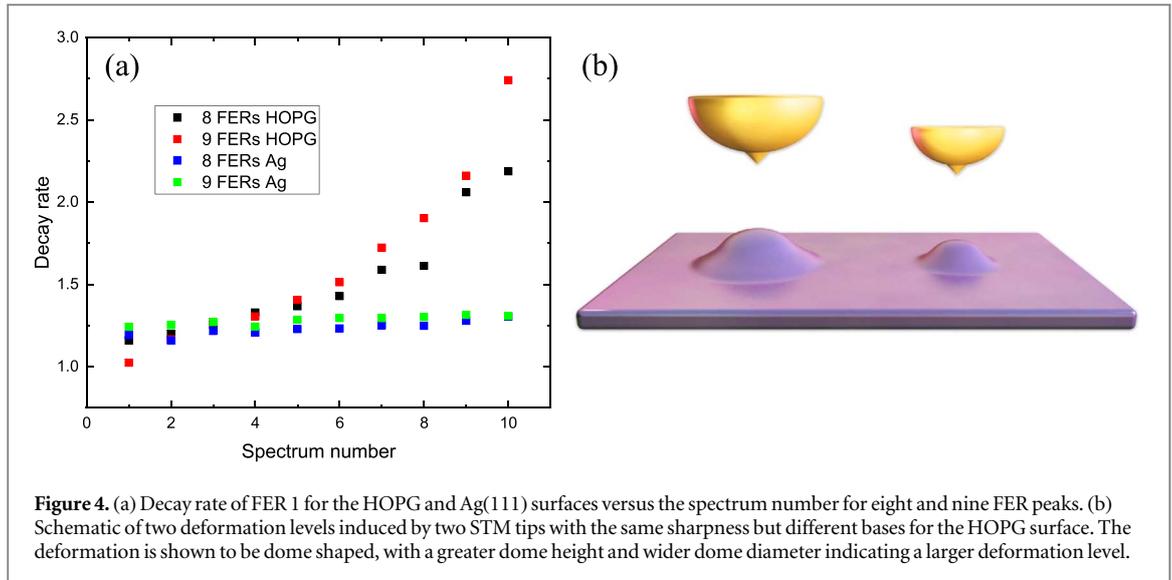
Previous studies revealed that FER electrons can leave the quantized state through light emission [16, 17] or surface transmission [34], which indicates that the wave function of FER electrons is not static but can be dissipated. In spite of wave function dissipation (WFD), before leaving FER state, the electron moves to and fro in round trips in the STM junction. Due to WFD, the probability $P(i)$ that electrons remain in the quantized state may decay with the number i of round trips at a rate of D which is the sum of the electron transmissivity Γ_t and the decay rate per round trip due to light emission Γ_l . $P(i)$ can be defined as follows

$$P(i) = (1 - D)^i. \quad (3)$$

Equation (3) indicates that the probability that a resonant electron stays in FER state for $(i-1)$ round trips but exits FER state in the i th round trip is $P(i-1) - P(i)$. The average number of round trips $\langle i \rangle$ of all resonant electrons is as follows:

$$\langle i \rangle = \sum_{i=1}^{i=i_{\text{max}}} i [P(i-1) - P(i)], \quad (4)$$

where i_{max} is the maximum number of round trips for which a resonant electron can stay in FER state, depending on the number of all resonant electrons. Using equations (3) and (4), $\langle i \rangle = 1/D$ is obtained (see supplementary



material (available online at stacks.iop.org/JPCO/6/075010/mmedia). Resonant electrons move back and forth within a distance $s = (E_n - E_{vac})/eF_{FER}$ between the surface and the classical turning point. As a result, the round-trip time t for resonant electron can be calculated using $2s = eF_{FER} t^2/m$. By combining this equation with equations (1) and (2), $t = \beta \frac{(n - \frac{1}{4})^{\frac{1}{3}}}{F_{FER}^{\frac{2}{3}}}$ is obtained, where $\beta = \left(\frac{3m\hbar\pi}{e^2}\right)^{\frac{1}{3}}$. The mean lifetime of resonant electrons in FER state, proportional to $1/\Delta E$, can be defined as $\langle i \rangle t$. Consequently, ΔE is proportional to D . This relation can explain that on reconstructed Au(111) surface, FER has a larger ΔE at the ridge area with a higher $\Gamma_t(D)$, leading to a spatial variation of FER peak intensity [34] (see supplementary material).

For $n = 1$, D can be represented by a quantity $(0.75)^{1/3} \Delta E / F_{FER}^{2/3}$. Thus, given ΔE and F_{FER} , we can calculate D for each FER 1 peak in figure 1. Figure 4(a) displays the variation of the D values for FER 1 for HOPG and Ag(111) versus N for the spectra with eight and nine FER peaks. Although the D value for Ag(111) (D_{Ag}) remained nearly unchanged as N varied, the D value for HOPG (D_{HOPG}) rapidly increased with N for $N > 5$. We suggest that this difference could be attributed to the TIAD caused by the electrostatic force in STM junction and D proportional to the TIAD level (discussed later). The TIAD was more prominent on the HOPG surface because the metallic bond in Ag crystals is considerably stronger than the van der Waals force between atomic layers in HOPG. Therefore, D_{HOPG} value exhibited a larger variation. However, the sharpness levels of the STM tip were close because FER number was the same. The sharpness level was unable to explain why the D_{Ag} value was nearly constant but the D_{HOPG} value varied. It is well accepted that an STM tip is composed of a base with a radius of tens of nanometers and a protrusion whose open angle determines the tip sharpness. Therefore, STM tips may have the same sharpness but different base radii. According to a theoretical study [43], when the distance d between the tip and surface is smaller than the base radius R , the electrostatic force F between them is as follows:

$$F = \pi\epsilon_0 V_0^2 R/d, \quad (5)$$

where V_0 is the applied bias voltage. Therefore, a tip with a larger base radius can engender a stronger electrostatic force, causing a larger deformation level on the HOPG surface, and vice versa [figure 4(b)]. The TIAD is similar to be dome shaped. A wider dome diameter and greater height are noted to be associated with a higher TIAD level. Because Ag crystals do not deform easily, D_{Ag} is insensitive to the base radius. Using equation (5), the electrostatic force can be estimated by assuming $R = 30$ nm and $d = 2$ nm for FER 1 whose energy is 6 eV, which is approximately 15 nN.

The TIAD probed by the FER linewidth demonstrated above was performed at the same current under different tip bases. Actually the electrostatic force can be tuned by changing the current. Accordingly, we also observed FER 1 ΔE values for HOPG and Ag(111) at 5 K under different currents. Figure 5(a) displays the FER spectra for HOPG at 0.01–1 nA, and figure 5(b) shows those for Ag(111) at 0.03–10 nA. As exhibited in both figures, when the current increased, the energy of the FER 1 peak (denoted by 1) moved toward high energy [39], indicating that the F_{FER} of FER 1 increased with the applied current. Figure 5(c) depicts smooth plots of F_{FER} versus current, implies that the tip structures on both surfaces were unchanged as the current increased. Figure 5(d) exhibits that FER 1 ΔE values enlarged with the current for both cases. On the basis of the results illustrated in figures 5(c) and (d), we calculated D_{HOPG} and D_{Ag} as a function of F_{FER} , and figure 6(a) presents a plot of the calculation results. Figure 6(a) reveals a considerable disparity that D_{HOPG} was linearly proportional

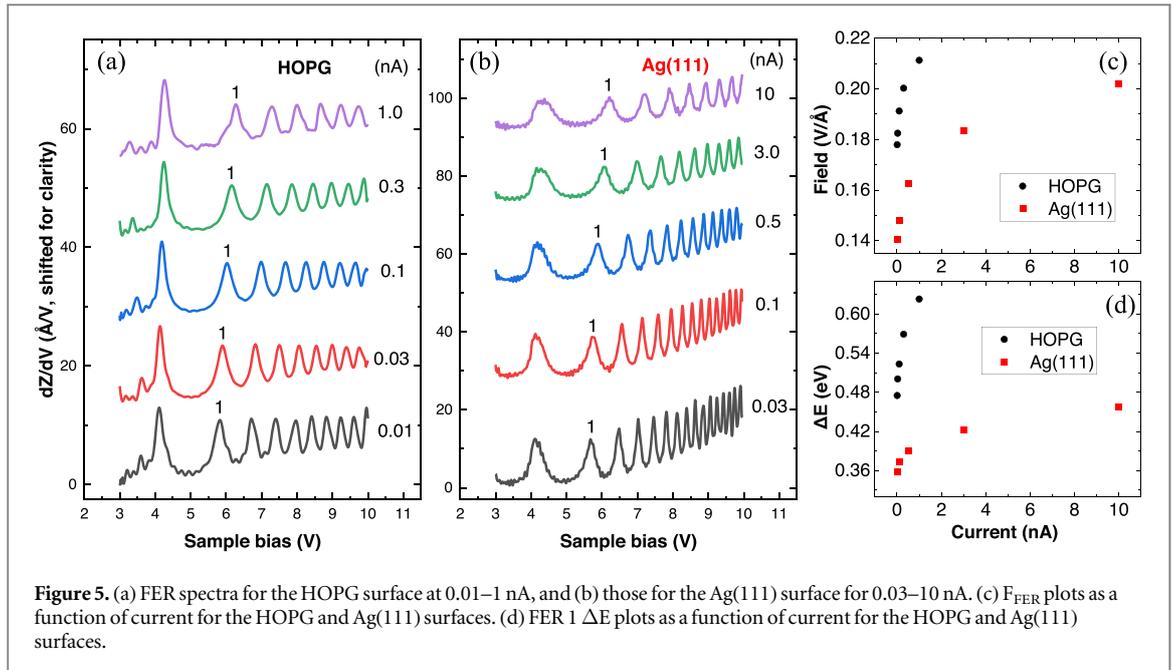


Figure 5. (a) FER spectra for the HOPG surface at 0.01–1 nA, and (b) those for the Ag(111) surface for 0.03–10 nA. (c) F_{FER} plots as a function of current for the HOPG and Ag(111) surfaces. (d) FER 1 ΔE plots as a function of current for the HOPG and Ag(111) surfaces.

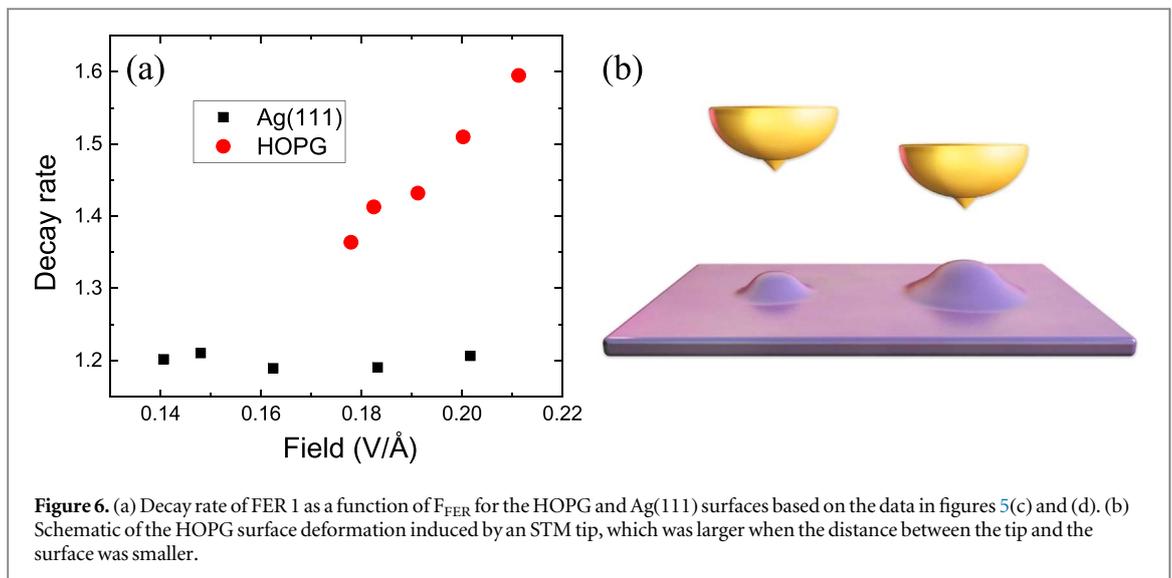
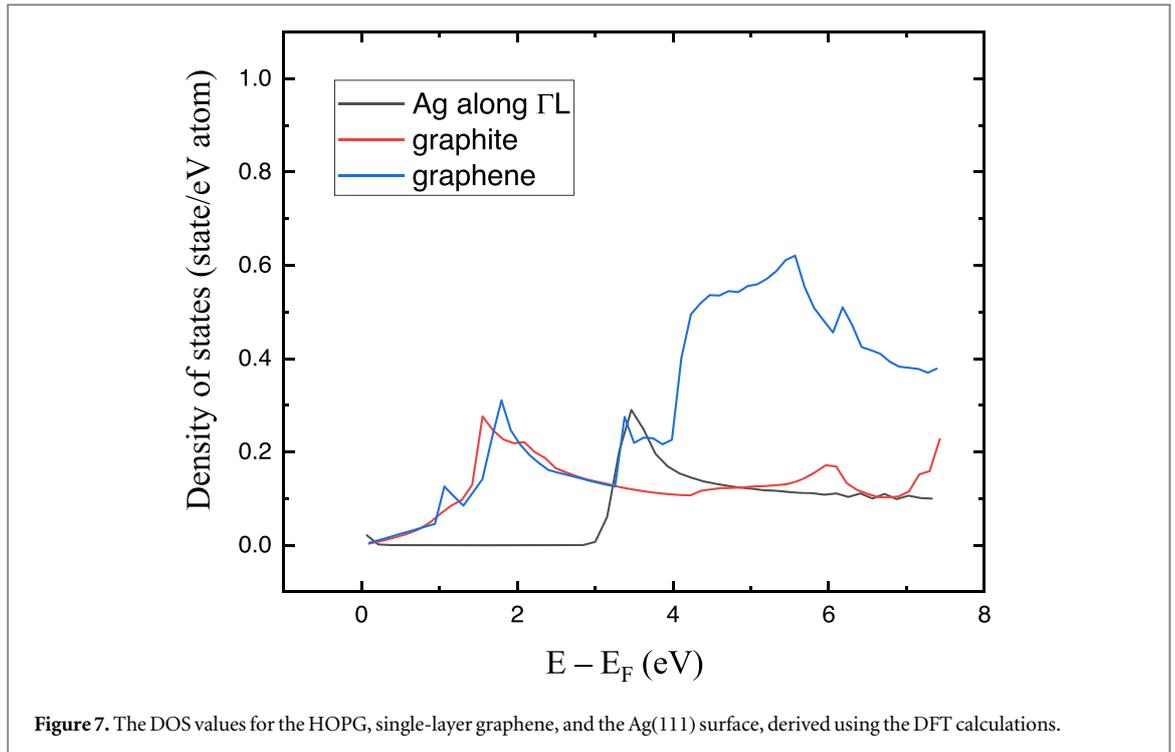


Figure 6. (a) Decay rate of FER 1 as a function of F_{FER} for the HOPG and Ag(111) surfaces based on the data in figures 5(c) and (d). (b) Schematic of the HOPG surface deformation induced by an STM tip, which was larger when the distance between the tip and the surface was smaller.

to F_{FER} , whereas D_{Ag} was nearly independent of F_{FER} . Because the electrostatic force enhances with increasing F_{FER} , figure 5(a) reflects that the TIAD on the HOPG surface was considerably larger than that on Ag(111), consistent with the results in figure 4(a). For increasing the current, the tip was controlled through the STM feedback, moving it closer to the surface. Accordingly, the electrostatic force was enhanced, attracting and displacing the HOPG surface upward. As a result, the TIAD level was higher (lower) at a shorter (longer) tip-surface distance, as visualized in figure 6(b).

Because HOPG and Ag(111) have no energy gap above their vacuum levels, the Γ_t fluctuation caused by quantum trapping effect [38] is weak. Consequently, D was determined to be dependent only on Γ_t in this study. A previous study suggested that Γ_t is proportional to the DOS [34], signifying that the difference between D_{HOPG} and D_{Ag} can be explained by the DOS. Figure 7 depicts DOS values derived through DFT calculations for the HOPG, single-layer graphene, and Ag(111) surface. The DOS values derived at the FER 1 energy of approximately 6 eV were comparable for HOPG and Ag(111), explaining that D_{HOPG} and D_{Ag} values are close at $N < 5$ in figure 4(a). Therefore, the situation can occur on HOPG that the tip base is too small to induce the deformation.

As mentioned, the D_{HOPG} value exhibited larger variations [figure 4(a)] and were linearly proportional to F_{FER} [figure 6(a)], indicating that the DOS increases with the TIAD level. As displayed in figure 7, at approximately 6 eV, the DOS value of graphene was considerably greater than that of the HOPG surface. This notable DOS difference suggests that the electronic structure of the top HOPG layer gradually approaches that of

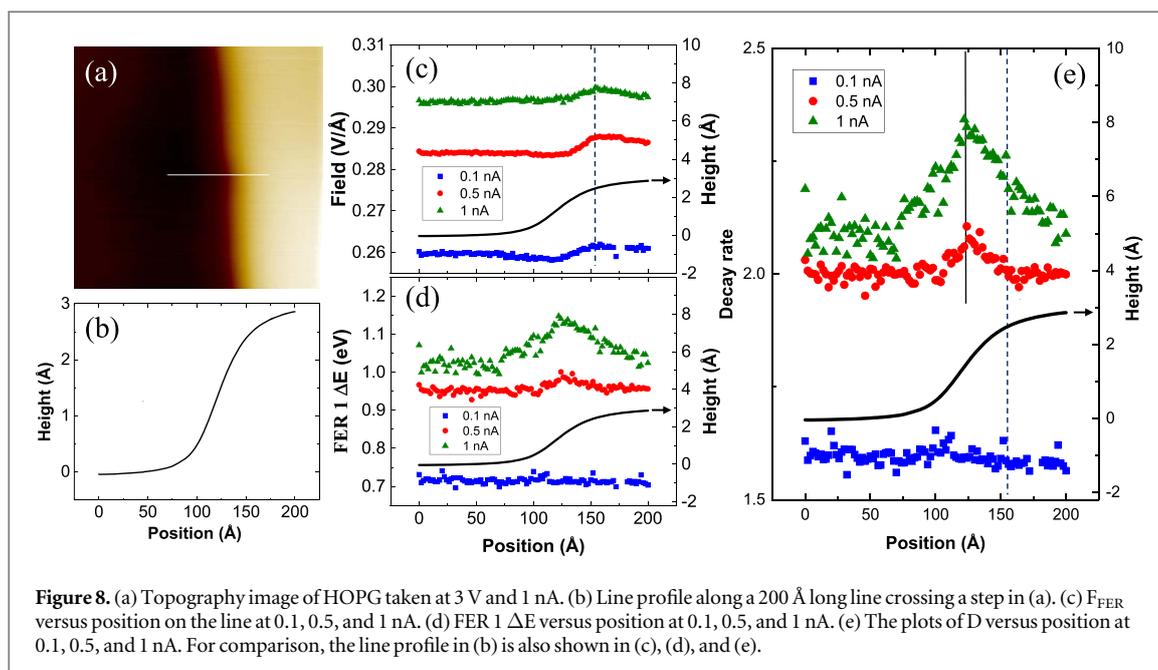


the graphene as the TIAD level increased, thus increasing the corresponding DOS value. Moreover, because HOPG is a semimetal, it might not be able to behave like a metal to wholly block the penetration of the electric field in STM junction into deeper layers, but the top layer can effectively reduce the electric field. Consequently, the electrostatic force on deeper layers would be weaker than that on the top layer. The TIAD levels of deeper layers would be lower than that of top layer. The distances between the top layer and deeper layers are larger than those in HOPG without electrostatic force. Therefore, the electrostatic force in the STM junction locally attracts the top layer to be away from the second layer, which could effectively produce a quasi-graphene layer beneath the tip on the HOPG surface.

We measured the distance change (ΔZ) in the Z - V spectrum corresponding to the differential spectrum of $N = 1$ in figures 1(a)–(d) for a bias voltage ranging from 3 to 10 V. The results show that distance changes are 30.0 and 34.1 Å for 8 and 9 FER peaks in the HOPG case, and 30.4 and 35.4 Å for 8 and 9 FER peaks in the Ag case, respectively. Therefore, for the same FER number, ΔZ on HOPG is nearly the same as that on Ag(111), indicating that the TIAD level on HOPG was not increased while the bias voltage increased, i.e. the electrostatic force could be constant when observing FER. Thus, FER peaks were measured on a stable dome-shaped surface.

Figure 8(a) is a topography image of HOPG taken at 3 V, 1 nA and 5 K, in which the surface consists of a high terrace (bright area) and a low terrace (dark area) due to a step. Figure 8(b) shows a line profile along a 200 Å-long line crossing a step in figure 8(a), revealing that the step height is close to one atomic layer. We took an FER spectrum along the line every 2 Å from left to right under 0.1, 0.5, and 1 nA (see supplementary material), and measured F_{FER} and FER 1 ΔE in each spectrum. Figure 8(c) presents F_{FER} versus position on the line at three currents. Although the STM feedback was on, F_{FER} still had a slight increase when the tip crossed the step, which is approximately 1.4% of F_{FER} on the low terrace for all three cases in figure 8(c). According to this F_{FER} increase, the position of the step edge can be determined from the maximum of F_{FER} , which is nearly identical for three cases in figure 8(c), marked by a dashed line at the position of 154 Å. Figure 8(d) displays FER 1 ΔE versus position at three currents. With the values in figures 8(c) and (d), we depicted the plots of D_{HOPG} versus position, as displayed in figure 8(e). For comparison, the line profile in figure 8(b) is also shown in figures 8(c), (d), and (e).

At 0.1 nA, the plot shows that D_{HOPG} is nearly constant. The plot at 0.5 nA reveals that D_{HOPG} is nearly constant at most of positions, but presents a triangle shaped variation for some positions; it increases from the position of 104 Å and reaches maximum at 124 Å then decreases. Because the step edge is at 154 Å (dashed line), the increase of D_{HOPG} occurs at the low terrace. Moreover, except the triangle shaped variation, there is no difference for D_{HOPG} at the low and high terraces. Therefore, the step can elevate the height of TIAD on the low terrace as the tip is close to a step. However, F_{FER} should exceed 0.26 V Å to detect this step-induced height elevation, according to figure 8(c). The triangle shaped variation reflects that the van der Waals force is weaker in the vicinity of a step due to less symmetry. The plot at 1 nA demonstrates that the triangle shaped variation becomes more pronounced and the positions at which D_{HOPG} is nearly constant shrink to a range between 0 to



74 Å, displaying that a stronger electrostatic force can extend the area of step-induced height elevation. Moreover, the average D_{HOPG} on the high terrace (154–200 Å) is greater than that on the low terrace (0–74 Å), indicating that due to less symmetry, at the high terrace, the van der Waals force near the step is weaker than that away from the step, but is stronger than that near the step at the low terrace because F_{FER} should be as high as 0.295 V Å to observe this difference, according to figure 8(c). In addition, because the triangle shaped variation results from the step, the lateral position of the highest value in D_{HOPG} is the same as that in the plot at 0.5 nA, as marked by a solid line.

4. Conclusions

We demonstrated that FER can be used as a tool to probe the TIAD of an HOPG surface. By measuring FER energies to derive F_{FER} and by extracting FER linewidth, we derived D_{HOPG} to monitor the TIAD of the surface. A higher D_{HOPG} value represents a higher TIAD level. For comparison, we also observed FER on the Ag(111) surface. The derived D_{Ag} was insensitive to the electrostatic force in STM junction, implying that the TIAD on Ag surface is negligible. DFT calculations demonstrated that the DOS of the graphene was considerably higher than that of graphite at the FER I energy, signifying that the TIAD rendered the top layer analogous to graphene because the decay rate is proportional to the DOS. It is well known that the electronic structures of two-dimensional (2D) materials are sensitive to the strain [44, 45]. Strain-induced change of the DOS above the vacuum level in 2D materials is expected, implying that FER may be a tool to probe the strain distribution on 2D materials through measuring the decay rate of its wave function.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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