

# sub-ps Snapshot of Electron Dynamics in an Organic Thin Film Captured by THz-STM

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Atomic scale surface local structure such as steps, dangling bonds, and defects often act as electron traps due to their localized electronic state. However, electron trapping dynamics in an individual electron trap has never been observed due to a lack of time resolution in atomic resolution microscopy. Recently, THz-STM has attracted much attention as a novel technique to probe ultrafast dynamics of photo-induced phenomena with atomic scale spatial resolution<sup>[1-3]</sup>. Free space coupling of THz pulses to STM tunneling junction induces sub-ps transient voltage ( $V_{\text{THz}}$ ) across the junction, which drives ultrafast tunneling current. In this study, we have utilized THz driven ultrafast tunneling current ( $I_{\text{THz}}$ ) to capture the sub-ps snapshot of free electrons in  $\text{C}_{60}$  thin film with atomic spatial resolution. Fig. a shows the experimental setup. Excess electrons in  $\text{C}_{60}$  thin film were optically injected by IR pump pulse from Au(111) surface as illustrated in the band diagram (Fig. b.), then we probed spatial distribution of injected electrons in the thin film by mapping  $I_{\text{THz}}$ . Left top image of Fig. c shows topographic STM image of  $\text{C}_{60}/\text{Au}(111)$  surface (average  $\text{C}_{60}$  thickness  $\sim 4$  ML) and the other shows corresponding  $I_{\text{THz}}$  map taken with fixed delay time between pump IR pulse and THz pulse. Since the negative charge of injected electrons transiently modulate tunneling conductance,  $I_{\text{THz}}$  map shown in Fig. c represent of excess electron snapshot of 2 ps  $\sim$  49 ps after optical injection. These snapshots show electron accumulation occurred around the step edge and a defect indicated by the white arrow. The result represents a significant impact of atomic scale local surface structure on electron dynamics.

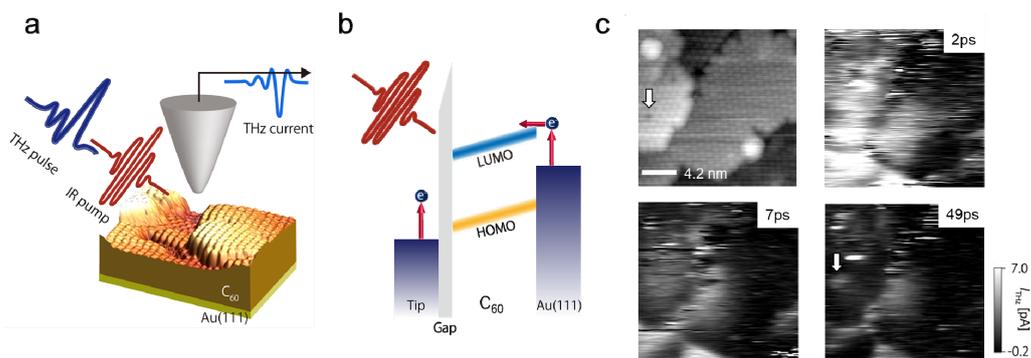


Figure. a. Illustration of experimental setup b. band diagram of STM tunneling junction under pulse excitation c. STM image of  $\text{C}_{60}/\text{Au}(111)$  surface and corresponding  $I_{\text{THz}}$  map taken with fixed delay time (2 ps, 7 ps, 49 ps)

[1] T. L. Cocker, et al., Nature photonics, 7, 620–625 (2013)

[2] T. L. Cocker, et al., Nature, 539, 263–267 (2016)

[3] S. Yoshida, et al., ACS photon, 6, 1356-1364 (2019)