In this study, we achieved the first atomic-scale visualization of photocurrent channels in a single molecule, and a description of the mechanism of photon-to-electron energy conversion with an ultimate precision based on the molecular orbitals [1-3].

Photoinduced electron transfer (PET) plays an essential role in light energy conversion processes, such as photocurrent generation, photocatalysis, and photosynthesis. The quest for an atomic-scale comprehension of PET has been a longstanding pursuit. However, the spatial resolution of conventional microscopic techniques for photocurrent measurement has previously been insufficient to resolve individual molecules, leaving the detailed mechanism obscured at the atomic scale.

Overcoming the limitations of the conventional methods imposed by the diffraction limit of light, we dedicated many years to developing Photon-STM, a scanning tunneling microscope combined with laser irradiation and photon detection systems. Our instrumentation has opened up a previously unexplored research frontier in exciton nanoscience [1-6].

Recently, we upgraded Photon-STM by installing a narrow-width tunable laser, enabling single-molecule laser nanospectroscopy with micro-electron volt energy resolution [5-6]. Applying this new technique to efficiently excite a molecule below the STM tip, we have recently succeeded to enhance the spatial resolution in photocurrent measurement, which lead to achieve the long-standing goal of visualizing photocurrent channels in a single molecule with atomic resolution [1-3].

In ref. [1], Photocurrent imaging and theoretical analysis reveal that the generation of photocurrent is decisively governed by the individual frontier molecular orbitals. Moreover, our technique has allowed us to discover that photocurrent flows locally through multiple counterflowing photocurrent channels, even at a voltage where the averaged photocurrent is zero. We also demonstrated the controllability of the quantum efficiencies of photocurrent generation and competing photoluminescence by tuning the molecule-electrode distance. These results clearly indicate that selective promotion and suppression of a specific de-excitation process is possible by an atomic-scale design of a molecular interface. Our findings provide a new design strategy for improving the efficiency of light energy conversion with organic devices. In addition, our method provides a technical basis for unprecedented atomic-scale visualization of excited states, which has the potential to revolutionize our fundamental understanding of functional energy conversion processes in the excited states.

We believe that these results will captivate a wide range of scientists across various disciplines, including organic devices, molecular biology, molecular spectroscopy, nanoscience and technology, quantum optics, and surface science.

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