

Single-molecule characterization from the perspective of optics, photonics, and optoelectronics: a review

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Abstract. A single molecule is the building block of the material world and the smallest independently stable unit. Exploring single-molecule properties using optical, photonic, and optoelectronic techniques holds great scientific significance in revealing the molecular dynamics, molecular structures, and molecular quantum properties. Nano-optical techniques, such as single-molecule photoluminescence and Raman scattering, not only enable a comprehensive analysis of interactions and conformational dynamics through spectral analysis but also provide unparalleled insights into elucidating the intricate structure of single molecules through atomic-resolution imaging. The research of photonics based on single-molecule electroluminescence has brought new ideas and limitless possibilities to the design and manufacture of photonic information devices. Single-molecule optoelectronics, which leverages photoexcitation to modulate electrical properties, has significant contributions to elucidating charge transport characteristics and optimizing the optoelectronic functions realized by single-molecule devices. Moreover, the optoelectronic characterization based on the interaction of ultrafast optical pulses with single molecules provides unprecedented opportunities for exploring their dynamic behavior and regulation laws on ultrafast time scales. We provide a timely and comprehensive overview of the latest significant advancements pertaining to the optical, photonic, and optoelectronic properties of single molecules, thereby presenting a fresh perspective for research across diverse fields, including single-molecule photophysics and photochemistry.

Keywords: single molecules; nano-optics; photonics; optoelectronics; ultrafast optics.

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1 Introduction

A single molecule represents the smallest stable unit of matter and exhibits diverse properties distinct from macroscopic aggregates, making it the focus of research in physics, chemistry, and related disciplines.¹⁻⁶ However, due to its diminutive size, there are significant challenges in identifying and delving into its physical properties.^{7,8} Among various studies, optics, photonics,

and optoelectronics are three key fields in single-molecule science.⁹⁻¹² Specifically, nano-optical methods involve detecting the emission of light or the scattering of incident light from a single molecule under the influence of external fields, thereby revealing the structure, properties, and interactions of the molecule in different environments.¹³⁻¹⁵ Due to the inherent constraints imposed by the diffraction limit, traditional optical detection methods have long been considered unable to capture optical signals from single molecules. However, the emergence of scanning tunneling microscopy (STM) has overturned this situation.¹⁶⁻¹⁸ This is because the nanotip of STM possesses

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powerful light-field localization characteristics, and the small size of this localized optical field allows optical detection to surpass the diffraction limit. Based on this, via placing single molecules within STM, related research on their luminescence has witnessed rapid and high-quality development, with the resolution of emission reaching the subnanometer scale. Furthermore, this localization feature can significantly enhance the intensity of light-scattering signals from single molecules,^{11,19,20} elevating the optical detection capabilities of single-molecule structures based on Raman scattering to the level of individual chemical bonds.

With the development of STM and single-molecule device techniques, various methods have been proposed to investigate the photonic properties of single molecules, especially electroluminescence properties.^{13,21–23} Among them, graphene-based single-molecule device platforms have garnered significant attention for the investigation of single-molecule electroluminescence.²⁴ By analyzing the electroluminescence behavior of these junctions, researchers have revealed advantages such as uniformity in both energy and polarization of emitted photons. Furthermore, these photons have been harnessed for logical operations, providing a powerful way for the development of practical single-molecule photonic devices.²⁵

Single-molecule optoelectronics aims to delve into and explore the critical photoelectric properties at the molecular scale.^{26–30} Precise optical modulation techniques have been utilized to initiate the photoexcitation process within single molecules. Then, by measuring the changes in the conductivity of single molecules, the laws governing charge transport and transmission pathways are analyzed. This is crucial for further understanding the properties of optoelectronics and revealing the physical mechanisms of light–matter interactions, supporting the design of novel optoelectronic devices. For instance, the investigation of the interaction between light and single molecules has elucidated the different mechanisms of photoisomerism and photo-conductance.^{31–34} On this foundation, the construction of optoelectronic devices such as single-molecule switches has been achieved through further design of molecular structures and devices. In addition, the optoelectronic characterization based on ultrafast optical techniques has been ingeniously applied to single-molecule studies.^{35,36} Through the interaction between ultrafast pulses and single molecules, real-time monitoring of the dynamic evolution of single-molecule structures has been achieved at the temporal resolution of picoseconds and femtoseconds, providing significant insights for ultrafast dynamics mechanisms of molecules and the construction of ultrafast-controlled single-molecule devices.

This review provides a comprehensive discussion of the latest significant advancements in optical, photonic, and optoelectronic properties of single molecules. First, several optical phenomena of single molecules and their crucial roles in revealing molecular properties are systematically outlined. Then, on the basis of electroluminescence, the photonic properties of single-photon emission and its application in information processing are discussed. Subsequently, the mechanisms and phenomena arising from the optoelectronics in single-molecule devices are detailed, as well as the optoelectronic properties based on the interaction of ultrafast optical pulses with single molecules. Finally, we focus our discussion on the emerging opportunities in the realm of combining single molecules with optical modulation methods, along with outlining potential avenues for future breakthroughs in this field.

2 Optical Properties of Single Molecules

The study of optical properties of single molecules is of great significance in revealing the physical mechanisms of the microscopic world. By utilizing advanced nano-optical technologies, single-molecule optical behavior can be conveniently observed. In particular, STM is the most important nano-optical technique platform for studying single-molecule optical phenomena. The use of STM helps to elucidate the mechanism of molecular electronic structures and energy conversion, revealing a wealth of information about electron spin modulation by light, interactions between surface plasmon and single molecules, and single-molecule fluorescence.³⁷ In the following, we will provide a detailed overview of the research progress, mainly including single-molecule photoluminescence and Raman scattering.

2.1 Single-Molecule Photoluminescence

The photoluminescence of single molecule is a process where, upon absorption of photonic energy by the electrons within the molecule, the electrons transition to an unstable excited state, followed by a radiative transition back to the ground state. This process ultimately releases energy in the form of light, with the emitted photons exhibiting a wavelength different from the absorbed photons. Single-molecule photoluminescence can be measured through scanning near-field optical microscopy (SNOM). The main principle of SNOM is to utilize the localized enhancement of the light field at the nanoscale, which can break the optical diffraction limit and achieve spatial resolution at the nanoscale. Taking advantage of the extremely small near-field optical probe in SNOM, the laser is precisely focused on a single molecule and excites it. The excited single molecule emits fluorescence, which is collected and transmitted to the detection system. By precisely adjusting the proximity of the light source to the sample, the photoluminescence signals with subnanometer spatial resolution can be obtained.

STM is a pivotal representative of SNOM imaging, exhibiting remarkable utility in the spectroscopic and imaging characterization of single molecules. Extensive studies have elucidated the behavior and properties of single molecules adsorbed on STM substrates, encompassing the understanding of molecular dynamics,^{38,39} chemical reactions,^{40,41} and intermolecular interactions.^{42,43} The investigation of single-molecule photoluminescence based on STM is usually conducted under ultrahigh vacuum and low-temperature conditions, which can effectively avoid the interference of thermal noise and chemical reactions on the emission spectrum. Two primary challenges emerged in exploring single-molecule photoluminescence for the STM platform. First, the fluorescence emitted by a single molecule is exceedingly weak, falling significantly below the detection threshold of detectors. Second, upon direct contact between the molecules and the electrodes, charge and energy transfer occur, resulting in fluorescence quenching. To overcome the issue of weak fluorescence intensity, an effective approach is to employ nanocavity plasmons (NCPs) to achieve resonance-enhanced fluorescence. This requires the selection of appropriate tip materials and precise control of plasmonic nanocavity structures, especially the fabrication and control of atomic-level structures of the tip, to achieve plasmon resonance modes. Usually, plasmon-active metal tips (such as silver) can be utilized, and an atomic scale protrusion structure is constructed through the tip modifications; then the distance between the tip and the sample is controlled at the subnanometer scale.

At this point, within the nanocavity between the tip and the metal substrate, the plasmon resonance and the lightning rod effect of the protrusion structure jointly induce a highly localized and significantly enhanced electromagnetic field below the tip. When the plasmon resonance mode effectively matches the photon energy of the incident laser and molecular luminescence, the emission intensity of single molecules can be significantly increased by several orders of magnitude. In addition, fluorescence quenching can be addressed by constructing a decoupling layer on the substrate, which not only increases the spatial distance between the molecules and the electrodes but also exhibits excellent insulating properties, effectively reducing the rate of charge and energy transfer from the molecules to the substrate. Common decoupling layers include molecules, oxides, and inorganic salts, among which multilayer sodium chloride (NaCl) is the most widely used.^{44–51} Based on this, an in-depth investigation of single-molecule photoluminescence in STM can be realized. For instance, photoluminescence imaging with subnanometer resolution has been achieved for a single zinc phthalocyanine (ZnPc) molecule placed on a substrate coated with three layers of NaCl [Fig. 1(a)].¹¹ The image of a single ZnPc molecule exhibits a ring-like pattern with four emission maxima at the lobes and a dark feature at the center [Fig. 1(b)]. Further analysis of the photoluminescence intensity reveals a spatial resolution of 0.8 nm [Fig. 1(c)]. This work breaks the limitations of fluorescence imaging resolution and significantly

improves the spatial imaging resolution of photoluminescence to the subnanometer scale.

In addition, STM-based photoluminescence imaging not only achieves subnanometer level resolution but also offers unprecedented possibilities for manipulating local molecular environments and elucidating complex interactions between light and matter. For instance, this approach has been employed to drive the photopolymerization of free-base phthalocyanine (H_2Pc), enabling the manipulation of photochemical reactions.⁵² As shown in the upper part of Fig. 1(d), the two tautomeric images of H_2Pc exhibit two similar double symmetric modes. The formation of tautomers is attributed to the switching of hydrogen atoms within the H_2Pc molecule [the lower part of Fig. 1(d)]. Upon illumination by a laser, the image reveals a fourfold symmetric pattern with noisy lines, indicating the continuous and rapid switching between the two tautomeric states of the molecule. Visualization of the switching process is achieved using tunneling current, where the current fluctuations can reflect different conductance states corresponding to the two distinct configurations and continuous switching. Furthermore, the rate of intramolecular hydrogen atom transfer can be controlled through the incident laser wavelength, thereby regulating the conversion rate between molecular isomers. These results demonstrate the manipulation of phototautomerization in a single H_2Pc molecule, validating the possibility of controlling light-induced excited-state-driven photochemical reaction

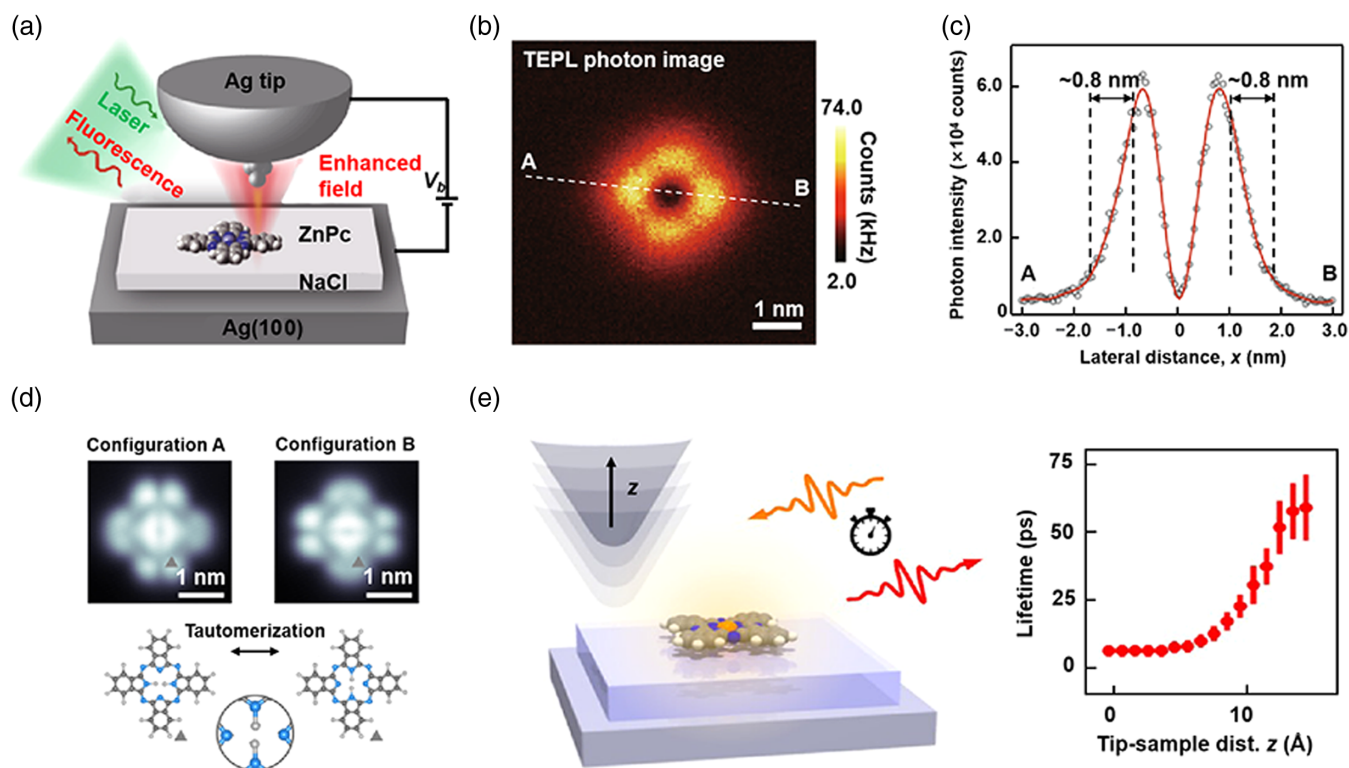


Fig. 1 Single-molecule photoluminescence. (a) Schematic diagram of the experimental setup of photoluminescence. (b) Photon image of single ZnPc molecule. (c) Corresponding photon intensity profile. (d) Phototautomerization and visualization of H_2Pc molecules. (e) Determination of the excitation lifetime of a single molecule in NCP through the time-dependent single-photon counting technique. [(a)–(c) Reproduced with permission from Ref. 11. Copyright 2020, Springer Nature Limited. (d) Reproduced with permission from Ref. 52. Copyright 2024, Springer Nature Limited. (e) Reproduced with permission from Ref. 53. Copyright 2024, American Chemical Society.]

processes at the single-molecule level. Furthermore, by combining the time-dependent single-photon counting technique, the excitation lifetime of a single molecule within the NCP can be determined [Fig. 1(e)].⁵³ In the absence of the NCP, H₂Pc molecular assembly exhibits nanosecond lifetimes. However, with NCP, the excitation lifetime of the H₂Pc molecule is reduced to the picosecond level due to the coupling effect between the tip and the NCP when the STM tip approaches the single H₂Pc molecule. This method effectively circumvents the limitations inherent in estimating the lifetimes of single molecules based solely on the widths of their emission spectra.

2.2 Single-Molecule Raman Scattering

Single-molecule Raman scattering stands as a distinctive optical phenomenon, distinguished by shifts in spectral lines under optical excitation. These shifts, which can either be a decrease or an increase in frequency concerning the original spectral line, are rooted in molecular vibrations.^{54–58} Raman spectroscopy and imaging, an offshoot of Raman scattering, has risen to prominence as a crucial analytical tool for dissecting the architecture and characteristics of single molecules.^{59–63} This method is particularly powerful due to the direct correlation between unique vibrational states and specific molecular configurations, which are clearly mirrored in the changes in frequency. However, performing single-molecule analysis with Raman spectroscopy presents a considerable challenge, stemming from the inherently weak and inelastic nature of the Raman effect, where only a minuscule fraction, one in 10⁶ to 10⁸, of the incident photons undergo Raman scattering. To overcome this limitation, it is typically essential to amplify Raman signals by bolstering the local field of interaction with molecules. Surface-enhanced Raman scattering (SERS)^{59,64} and tip-enhanced Raman scattering (TERS)^{59,62} are two principal techniques utilized for enhancing Raman signals. The former occurs on the rough surface of nanostructures composed of metals. When the nanostructure is illuminated by light, collective oscillations of electrons emerge on their surface, a phenomenon referred to as localized surface plasmon resonance. This resonance can convert the excitation light into localized electromagnetic fields, which are confined to the surface of the metal material [Fig. 2(a)].⁶⁵ The localized electromagnetic fields are strong enough to amplify Raman spectral signals from adjacent molecules to a level that can achieve single-molecule resolution. In addition, sample properties and testing conditions also need to be considered. For instance, the orientation of single molecules can significantly affect the strength of signals, which is attributed to the particular orientation that governs the Raman selection rules of the fingerprints. Ultrahigh vacuum and low temperature environment can reduce additional scattering and interference, improving the signal-to-noise ratios. Single-molecule SERS⁶² exhibits a narrow half-width and a high quality factor, and the Raman signals can be significantly intensified with an enhancement factor $\sim 10^6$.

Single-molecule SERS can effectively reflect the anisotropy of molecules, enabling the acquisition of crucial information pertaining to the kinetic laws, adsorption state, and the evolution of chemical reactions. For instance, single-molecule SERS has garnered widespread application in exploring the intricate interactions among reactant molecules and elucidating the fundamental principles of chemical reactions pertinent to catalytic reaction kinetics. Utilizing gold (Au) nanoparticle dimers, the

photodegradation of 4-nitrothiophenol has been observed, thereby elucidating the pathway leading to the formation of thiophenol molecules [Fig. 2(b)], laying a valuable foundation for future investigations into the catalytic reaction kinetics of the single 4-nitrothiophenol molecule.⁶⁶ The other typical example is the study of the catalytic reaction process of the single rhodamine B isothiocyanate molecule within a nanocavity [Fig. 2(c)], which unveils crucial differences in the chemical behavior of a single molecule at the nanoscale compared to macroscopic reactions.⁶⁷

In addition, molecular orbital gating is an important approach in single-molecule SERS research. It can further enhance the Raman scattering signal, building upon the original electromagnetic field enhancement, by adjusting the gate voltage that is associated with the molecular orbital energy within single-molecule devices.⁶⁸ For instance, the mechanically controllable break junction technique is used to build field-effect transistors based on single 1,4-benzenedithiol molecules [Fig. 2(d)]. The enhanced electromagnetic fields are confined within the nanogap of the source-drain electrodes, allowing the acquisition of distinct Raman signals from the molecular junction. It can be observed that the Raman signals are further enhanced by an additional approximate 40% through the gating effect [Fig. 2(e)]. This enhancement is attributed to the increase in the polarizability derivatives of molecules, which is related to the reduced energy gap between the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) modulated by the gate voltage. The energy gap decreases from ~ 0.47 eV at 0 V to ~ 0.34 eV at -20 V, where the energies of HOMO and LUMO are ~ -5.12 eV and ~ -4.65 eV, respectively, at 0 V. When the applied gate voltage is -20 V, the energies of HOMO and LUMO are ~ -4.96 and ~ -4.62 eV, respectively.

Due to the insufficient precision in the size of nanostructures and the limitation imposed by the optical diffraction limit, the spatial resolution of single-molecule SERS characterization is constrained. To resolve the local chemical and electronic structures of molecules at the level of chemical bond identification, Raman characterization technique with a higher spatial resolution is required. The advent of TERS has the potential to further enhance this capability [Fig. 2(f)].⁶⁹ By employing specially designed tips, typically coated with precious metals such as Au or silver (Ag), TERS achieves a highly localized plasmonic resonance enhancement confined within the nanocavity defined by the metallic nanotip and the substrate, which is the primary reason for the enhancement of spatial resolution. Coating the tip with different metallic materials can further enhance the inelastic scattering signal by spectrally matching the downward transitions associated with Raman photon emission. At the same time, the control of the extremely small curvature radius of the tip enables a further improvement of spatial resolution. TERS exhibits exceptional signal enhancement of up to 10⁷, enabling the detection sensitivity to be improved to the subnanometer level.⁶⁷

TERS can demonstrate superior performance in single-molecule spectroscopic analysis while serving a pivotal role in single-molecule structural imaging. In 2012, initial reports on TERS-based investigations of catalytic processes emerged.⁷⁰ These studies involve the tip of an atomic force microscope coated with Ag and the precise focusing of a 532 nm laser onto the nanoscale gap between the Ag-coated tip of the atomic force microscope and Au nanoplates.⁷⁰ Through this arrangement, the

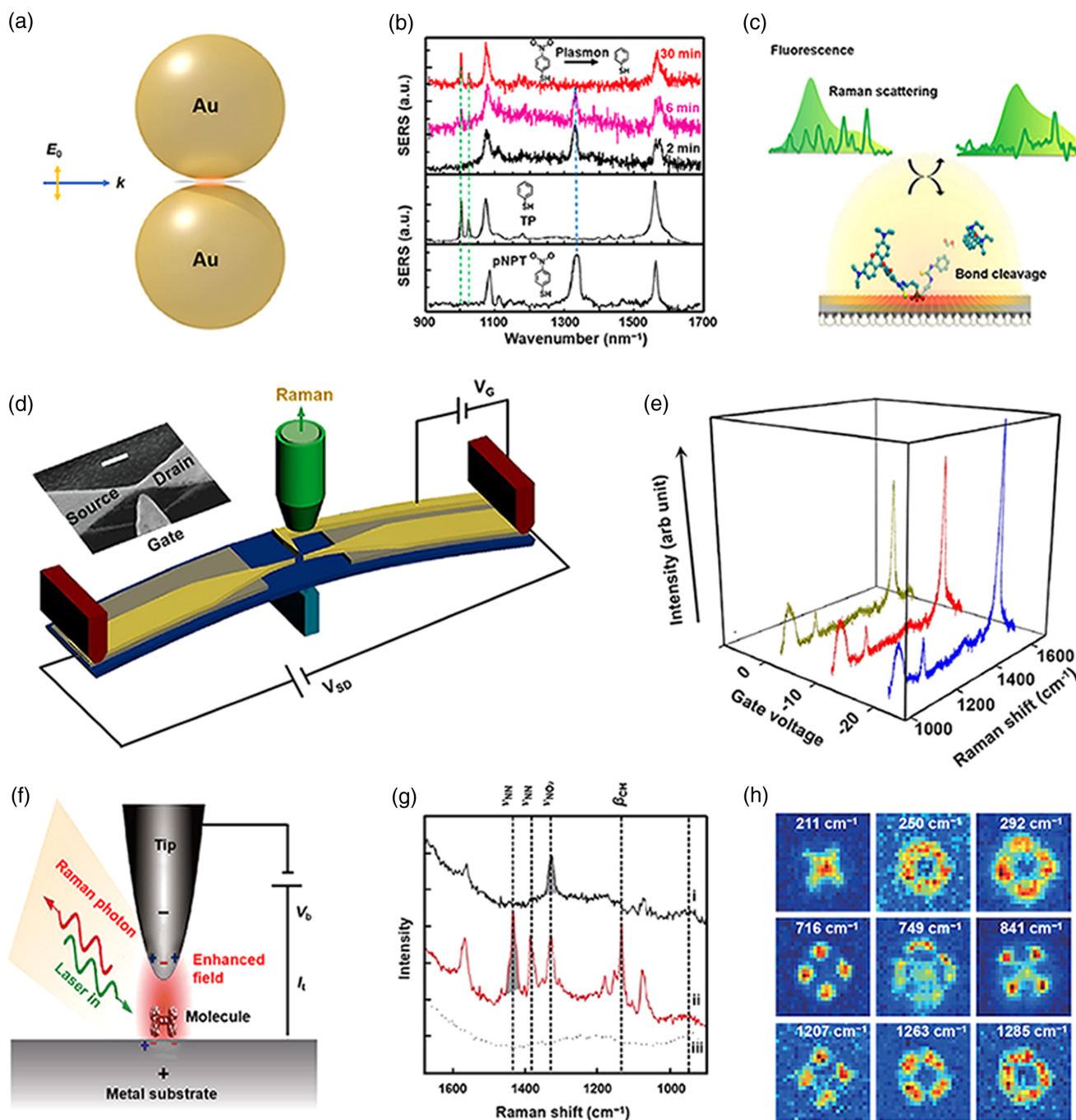


Fig. 2 Single-molecule Raman imaging. (a) Schematic illustration of the SERS mechanism within metal nanoparticles. (b) Photodegradation of 4-nitrothiophenol based on SERS. (c) Catalytic reaction process of single rhodamine B isothiocyanate molecule. (d) Schematic illustration of the SERS for side-gating modulation. (e) SERS spectra of 1,4-benzenedithiol molecular junction at different gate voltages. (f) Schematic illustration of the experimental setup of TERS. (g) Characteristic peaks corresponding to the reaction product *p,p'*-dimercaptoazobisbenzene in the TERS spectra. (h) Raman images obtained at different Raman peak positions of single magnesium porphyrin molecule. [(a) Reproduced with permission from Ref. 65. Copyright 2019, American Chemical Society. (b) Reproduced with permission from Ref. 66. Copyright 2015, Royal Society of Chemistry. (c) Reproduced with permission from Ref. 67. Copyright 2020, American Association for the Advancement of Science. (d), (e) Reproduced with permission from Ref. 68. Copyright 2018, American Chemical Society. (f) Reproduced with permission from Ref. 69. Copyright 2013, Springer Nature Limited. (g) Reproduced with permission from Ref. 70. Copyright 2012, Springer Nature Limited. (h) Reproduced with permission from Ref. 71. Copyright 2019, Oxford University Press on behalf of China Science Publishing & Media Ltd.]

localized surface plasmon resonance of the system is harnessed to trigger the reaction of *p*-nitrothiophenol with *p,p'*-dimercaptoazobisbenzene on the surface of Au nanoplate. The characteristic peak corresponding to the *p,p'*-dimercaptoazobisbenzene (ν_{NN} , β_{CH}) is exclusively observed following irradiation with the laser, signifying the occurrence of photocatalytic reactions of single *p*-nitrothiophenol molecules [Fig. 2(g)]. This study underscores the capability of TERS to monitor nanoscale catalytic reactions through the utilization of molecular fingerprints. By meticulously managing the distance between the tip and the target molecule, the local electric field enhancement can be finely tuned to achieve a better spatial resolution. For instance, STM has been used to capture significant Raman scattering signals with minimal photon flux, enabling successful imaging of a single meso-tetrakis(3,5-di-tertiarybutylphenyl)-porphyrin molecule.⁷¹ An in-depth analysis of the Raman image profiles of the molecules discloses an extraordinary spatial resolution beneath 1 nm. Subsequently, employing a comprehensive vibrational imaging strategy, the chemical structure of magnesium porphyrin molecules is successfully reconstructed at the single-molecule level. Figure 2(h) presents the Raman images captured at distinct Raman peak positions of the magnesium porphyrin molecule. These images provide a direct visual representation of the unique structural attributes linked to each vibrational mode of the molecules in real space, underscoring the vast potential of single-molecule Raman imaging for unraveling the complexities of molecular structures.

3 Photonic Properties of Single Molecules

The core of single-molecule photonics is to use photonic technologies to detect and analyze the behavior of single molecules. Due to the high spatial resolution and single-photon manipulation ability of single-molecule electroluminescence (SMEL), it is considered one of the ideal technologies for studying single-molecule photonics. The in-depth study of SMEL, in contrast to the electroluminescence of macroscopic molecules, offers insights into the intrinsic quantum-level properties and holds significant scientific and practical application value for the advancement of single-molecule devices and quantum information processing. Two main platforms have emerged in investigating the photonic properties based on SMEL: STM and single-molecule devices. These platforms have facilitated the elucidation of electron transition and energy conversion mechanisms within single molecules, leading to advancements in the applications of photonics, such as quantum information and photonic chips. In the following, we provide a detailed overview of the research progress in SMEL through these two platforms.

3.1 Electroluminescence in STM Nanocavity

The STM technique not only leverages the exceptional spatial resolution of the tip to directly acquire geometric structural features and electronic state information of molecules in real space but also enables the utilization of highly localized tunneling electrons to excite the molecules and capture their local optical signals. These capabilities render STM a powerful tool for in-depth studies of single-molecule electroluminescence characteristics. As the primary means for investigating SMEL, the fundamental mechanism of STM-based SMEL lies in the tunneling electron injection from the STM tip into the LUMO of a single molecule located on the substrate surface, driven by an externally applied electric field. Subsequently, the electrons

undergo radiative transitions back to the HOMO. During this process, the electrons recombine with the holes in the LUMO, which subsequently release energy through the emission of photons. The SMEL within STM is crucial for exploring molecular properties at the single-photon scale and provides vital scientific insights and a basis for the development of functional devices and quantum light sources based on molecular systems.^{15,38}

Consistent with photoluminescence, the STM-based electroluminescence signals necessitate effective amplification under the combined influence of NCP resonance and decoupling layer. Leveraging this principle, the detector-captured signals can be subsequently utilized for imaging and spectral analysis. For instance, the electroluminescence spectra of single ZnPc molecules have been investigated by placing the molecule on a three-layer NaCl substrate [Fig. 3(a)], achieving visualization of the coherent dipole–dipole coupling between molecules.⁷² Initially, two separate ZnPc molecules are combined to form a dimer, and the state of the dimer can be distinguished through spatial structural imaging and spectroscopy [Fig. 3(b)]. When the STM tip is positioned at different locations on the dimer, the number and intensity of spectral peaks vary significantly, exhibiting blueshifts or redshifts compared to a single ZnPc monomer. These spectral variations are attributed to the interaction between the exciton states of the two molecules, namely, intermolecular exciton coupling. The strength of the coupling varies at different positions, leading to changes in the molecular energy levels corresponding to the emission peaks. In addition to visualizing intermolecular exciton coupling at the single-molecule scale, STM-based electroluminescence spectroscopy also plays a crucial role in studying the charge states of molecules.⁴⁶ The spectral differences between the neutral and cationic configurations of single ZnPc molecules are particularly significant. Specifically, the neutral state of the ZnPc molecule is generated under a positive bias, exhibiting a peak at 1.89 eV in the electroluminescence spectrum. On the contrary, the cationic state of the ZnPc molecule forms under a negative bias, displaying a peak at 1.52 eV in the spectrum. The primary reason for this observation lies in the fact that under a negative bias, electrons tunnel from the molecule to the tip and evolve into a stable free radical cation state. Conversely, when a positive bias is applied, the molecule reverts to its neutral state. In the spectrum under negative bias, the peaks of emitted photons of both cationic and neutral molecules can be detected, indicating rapid flickering between the neutral and cationic states of the ZnPc molecule. Therefore, the photonic properties of STM-based electroluminescence can be used to investigate both neutral and charged systems.

Besides, STM-based electroluminescence can be utilized for the construction of single-photon source arrays. Hou et al. confirmed that STM-based electroluminescence can enable single ZnPc molecules to emit photons with high single-photon purity. Furthermore, through precise manipulation, a 3×3 array of single-photon emitters has been constructed [Fig. 3(c)].⁷³ Each molecule within the array exhibits nearly identical emission characteristics, demonstrating its potential for integrating quantum devices at the nanoscale. In addition, STM-based electroluminescence can selectively study triplet excitons under a specific bias voltage. For example, under high bias voltages, both phosphorescence and fluorescence signals can be observed from the 3,4,9,10-perylenetetracarboxylicdianhydride molecule.⁷⁴ However, at low bias voltages, only phosphorescence signals are detectable [Fig. 3(d)]. This observation suggests that under low-bias conditions, the molecules exclusively form triplet excitons

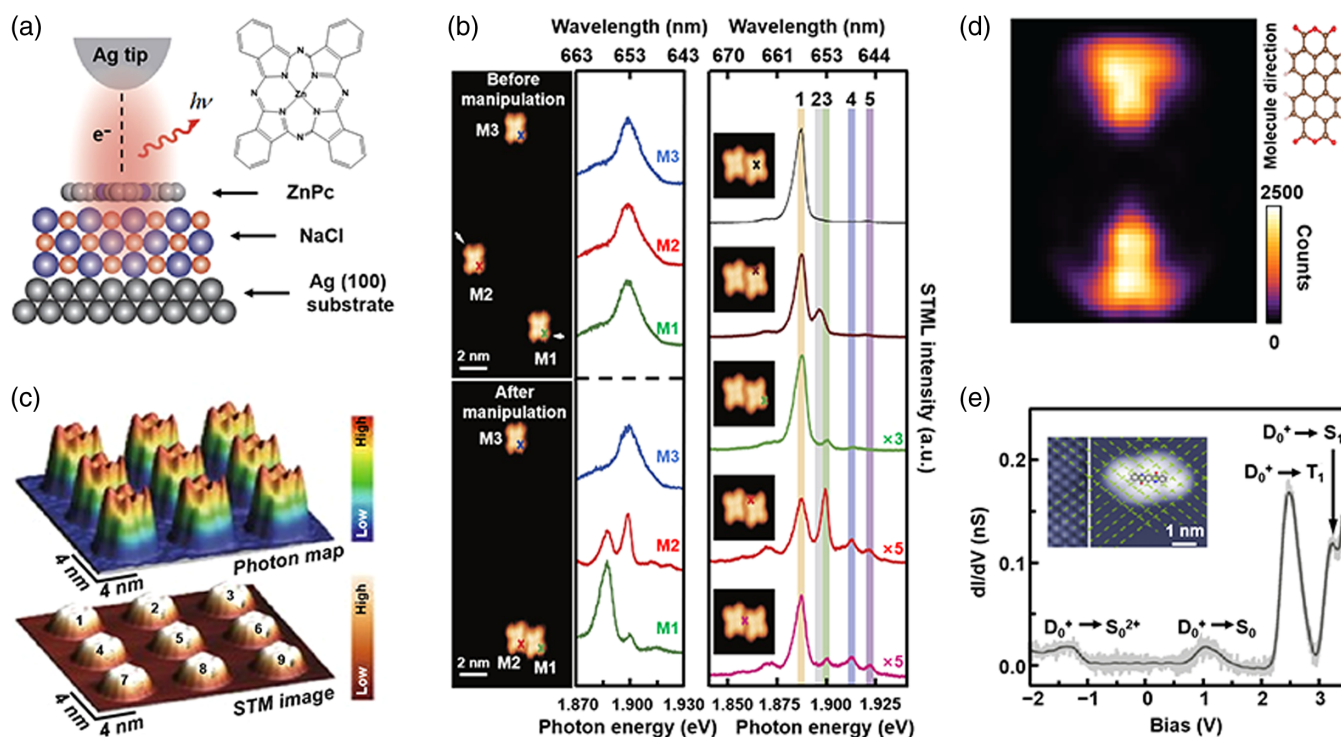


Fig. 3 Single-molecule electroluminescence based on STM. (a) Schematic diagram of STM-electroluminescence from a single ZnPc molecule on NaCl/Ag(100) substrate. (b) Spectral evolution from isolated ZnPc monomers to an artificially constructed molecular dimer. (c) Simultaneously acquired photon map (top) and STM image (bottom) of a 3x3 molecular array composed of ZnPc molecules. (d) Phosphorescence map of the 3,4,9,10-perylenetetracarboxylicdianhydride system. (e) dI/dV spectrum recorded at the center of a single quinacridone molecule. [(a), (b) Reproduced with permission from Ref. 72. Copyright 2016, Springer Nature Limited. (c) Reproduced with permission from Ref. 73. Copyright 2017, Springer Nature Limited. (d) Reproduced with permission from Ref. 74. Copyright 2019, Springer Nature Limited. (e) Reproduced with permission from Ref. 75. Copyright 2023, American Physical Society.]

without the formation of singlet excitons. Furthermore, a universal many-body model has been established for the electroluminescence of the quinacridone through the examination of voltage, current, and spatial dependencies [Fig. 3(e)].⁷⁵ This universal many-body model elucidates the mechanisms of charge transfer and luminescence in STM, providing potential applications for studying complex molecular systems with multiple charged states in other experimental environments.

3.2 Electroluminescence of Single-Molecule Devices

Through the study of STM-based SMEL, the luminescence mechanism of molecules under electrical excitation can be deeply understood, thereby laying a crucial theoretical foundation for fields, such as functional devices and quantum information. Promoting the applications of SMEL further stands as a common aspiration, and one promising approach is to develop stable and long-lasting luminescent single-molecule devices that are also easily integrable. Certainly, this necessitates the overcoming of significant challenges in device fabrication and luminescent characterization. Drawing inspiration from molecular engineering design and micro/nanoprocessing techniques, a range of functional single-molecule devices have been realized and iteratively enhanced by optimizing the functional molecular

cores, electrode materials, and interfacial coupling.⁷⁶ As such, a bottom-up approach toward the preparation of single-molecule electroluminescence devices emerges as a promising strategy after considering the robustness of the device, atomic scale configuration of the electrode, molecule-electrode interface, and molecular isolation from the external environment. Chemical modification of the electrode edge ensures robust device stability by securely anchoring the organic molecules with luminescent properties through chemical covalent bonds. The triangular configuration of terminal electrodes provides the ability for point contact, enabling decoupling between electrodes and luminescent centers. Molecular designs can also adjust the coupling strength between molecular functional centers and electrodes, thereby mitigating fluorescence quenching. The device can be encapsulated by deoxygenated polydimethylsiloxane to prevent electrical burning at high bias voltages. In addition, high luminescent efficiency requires alignment of the energy levels between the electroluminescent center and the electrode to avoid nonradiative losses, which can be achieved by adjusting the gate electric field. Based on this, Guo et al. successfully demonstrated the fabrication of single-molecule electroluminescent diodes exhibiting remarkable quantum yields [Figs. 4(a) and 4(b)].²⁴ The photonic characteristics of the single-molecule electroluminescence can be effectively revealed using stochastic

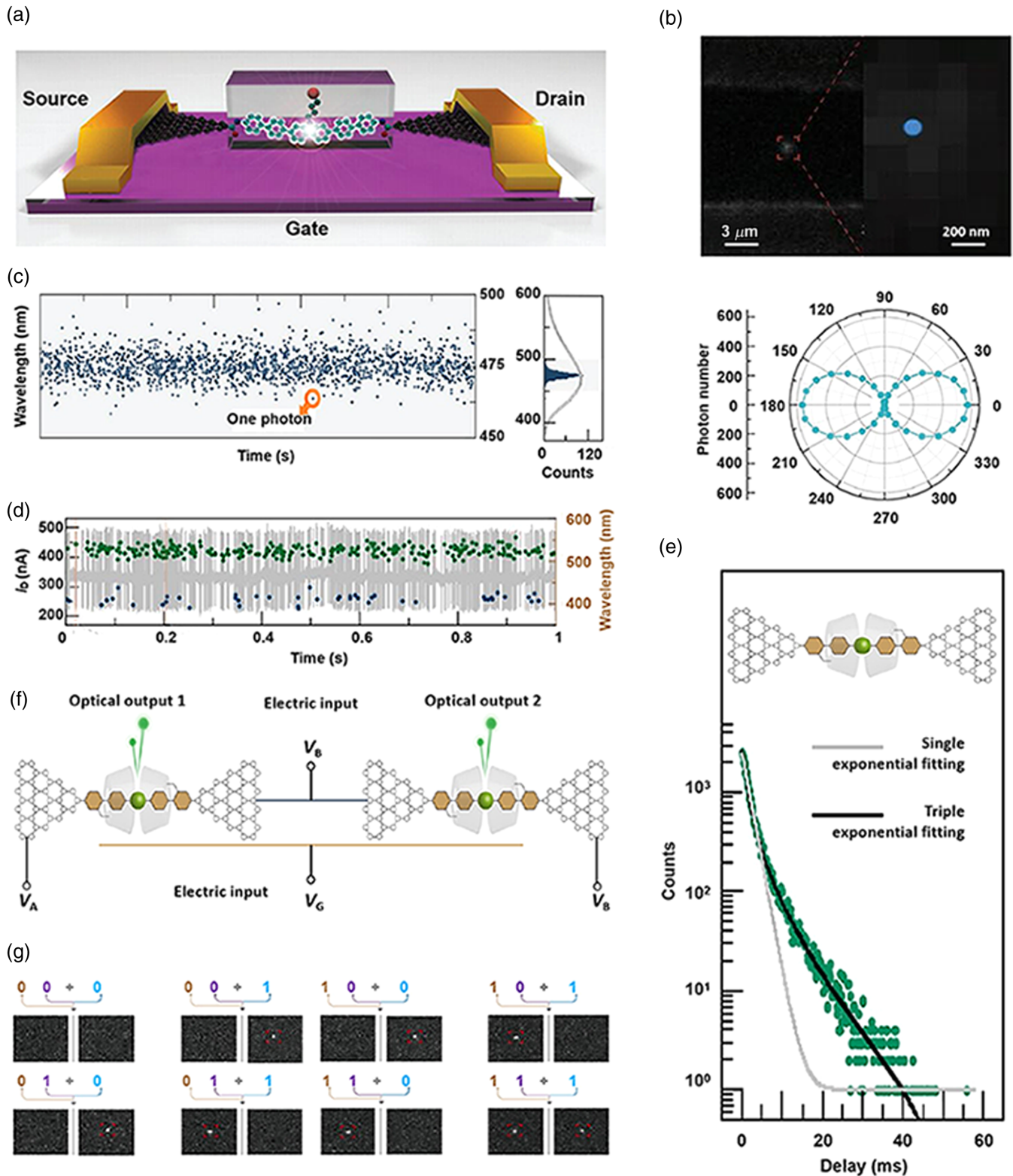


Fig. 4 Single-molecule electroluminescence of graphene-based single-molecule devices. (a) Schematic illustration of single-molecule luminescent diode. (b) Superhigh-resolution image (top) and the polar diagram (bottom) of the single-molecule electroluminescence. (c) Counting of emitted photons for different wavelengths. (d) Optoelectronic joint detection of the graphene-based single-molecule devices. (e) Statistical histogram of the decay time of phosphorescent signals. (f) Schematic of the single-molecule devices used for the logic operation. (g) Construction of the full-adder. [(a)–(c) Reproduced with permission from Ref. 24. Copyright 2023, Wiley-VCH GmbH. (d)–(g) Reproduced with permission from Ref. 25. Copyright 2024, Elsevier Inc.]

optical reconstruction microscopy.²¹ For example, the emitted photons display remarkable polarization homogeneity [Fig. 4(b)] and energy uniformity [Fig. 4(c)]. These phenomena arise from the fact that there is only one molecule participating in luminescence, eliminating variations among different molecules, which leads to a remarkably narrow emission bandwidth associated with energy. Furthermore, the confinement of the molecule, which is covalently connected to graphene electrodes, reduces vibrational and rotational modes, resulting in photon emission with high polarized purity. In addition, by altering the donor and acceptor groups, the modulation of multicolor photon emission can be realized. This is attributed to the substituent versatility of both donor and acceptor moieties, affording extensive flexibility in fine-tuning the magnitude of molecular energy levels.

The robust stability of graphene-based single-molecule light-emitting devices confers them with significant practical promise. For instance, single-molecule logic-operating device leveraging phosphorescence/fluorescence electroluminescence has been successfully fabricated through covalent connection of a single Pt-centered molecule encapsulated within cyclodextrin (Pt-MB@cyclodextrin) between graphene electrodes.²⁵ Utilizing photoelectric joint detection approaches, it is possible to observe transitions among the three distinct electrical states of molecules, alongside the emission of two different types of photons: fluorescence at 410 nm and phosphorescence at 524 nm [Fig. 4(d)]. By extracting the time intervals between applied electrical pulses and photons and analyzing the statistical distribution of corresponding photon decay times, millisecond-scale phosphorescent lifetimes are revealed, as shown in Fig. 4(e). Further, both single exponential and triple exponential fittings were performed on the lifetime of electroluminescence, where the single/triple exponential fitting represents the definition of the luminescence intensity by one/three-time constants, respectively, implying the presence of one/three potential decay pathways. The triple exponential fitting was found to be more consistent with the experimental results, indicating that the luminescence lifetime curve originates from three emission pathways from the triple excited state. This further proves that optical detection methods can clearly distinguish phosphorescent signals and their origins. In addition, from the perspective of single-molecule electrical characterization, information related to energy level can be clarified. The switching of the main current signals in Fig. 4(d) can be attributed to the triplet state (labeled as T_1) and the ground state (labeled as Pt_0). The variations in current associated with fluorescent photon emission arise from the transition from the positively charged state (Pt^+) to the Pt_0 . The current modifications corresponding to phosphorescent photon emission stem from the transition of T_1 to Pt_0 . At bias voltages below 3 V, preferential formation of the lower triplet state is observed, whereas at elevated bias voltages, both the singlet and triplet states become populated. According to the electrical tunability inherent in this photophysical process, two single-molecule devices have been integrated to realize a binary logic operation device operating at the single-molecule level [Fig. 4(f)]. By concurrently adjusting the electroluminescence state of the device via precise control of gate and bias voltages, the dark and bright states that correspond to logical operation symbols (“0” and “1”) are achieved within single-molecule devices. This approach enables the innovative implementation of Boolean logic gates, encompassing AND, OR, NOR, and other operations. Furthermore, the successful demonstration of a full adder subtractor is achieved by

introducing additional voltage settings on distinct terminals of the two single-molecule devices [Fig. 4(g)]. These robust single-molecule photonic devices, leveraging luminescent mechanisms, demonstrate the powerful potential of single-photon emission, control, and detection.

4 Optoelectronic Properties of Single Molecules

One of the key directions in the field of single-molecule science^{77–82} is single-molecule optoelectronics, which integrates theoretical and experimental techniques from different disciplines, including molecular optics and electronics, covering multiple aspects, such as molecular optoelectronic effects, molecular energy levels and spectroscopy, and electronic behavior and structure. Single-molecule optoelectronics focuses on harnessing optical approaches to manipulate the electronic properties of molecular junctions. This means that it exploits the effect of photons on electrons to uncover their electron transport properties, to elucidate the photoelectric conversion mechanism at the single-molecule scale and explore its potential applications in optoelectronic devices, thereby promoting the construction of functional single-molecule devices.^{80,81}

Two key contents will be discussed as follows: one is photo-induced molecular isomerization, and the other is photo-conductance. Photo-induced isomerization refers to the change in the geometric structure of a single molecule under light irradiation.^{83–85} When the conjugation states of the two isomeric forms differ significantly, it often leads to a dramatic change in conductivity, thereby enabling the realization of molecular switch devices. Photo-conductance of single molecules refers to the sharp molecular conductance change with light-induced electronic state alteration. In recent years, investigations into photo-conductance have facilitated the production of various types of stable and controllable single-molecule optoelectronic devices.^{30–33,86,87} These advancements have enabled us to elaborate on the microscopic mechanisms underlying molecular photoelectric effects, delve deeper into the principles governing the intricate interactions between single molecules and light, and finally realize the practical applications of single-molecule optoelectronic devices.

4.1 Photo-Induced Isomerization Switching

Photo-induced isomerization refers to structural changes of the molecule under illumination, such as cyclization and decyclization processes, and *cis-trans* isomerization. These geometric transformations lead to modifications in the molecular backbone, resulting in variations in the charge transport capabilities. The conductivity can vary by several orders of magnitude between different isomers, enabling the realization of single-molecule optoelectronic switches through transitions among these isomers. Currently, the primary application of photo-induced isomerization lies in the realm of single-molecule optoelectronic switches. For instance, diarylethene can reversibly transition between its cyclized and decyclized states upon alternating ultraviolet and visible light irradiation, resulting in the molecular current fluctuations between high and low conductance states [Fig. 5(a)].⁸³ Initial studies of diarylethene have encountered reversibility of its switching functions. Subsequently, the limitation is broken by refining the coupling between the electrode and the functional molecular center. Jia et al. employed three methylene groups at the end of the diarylethene molecule

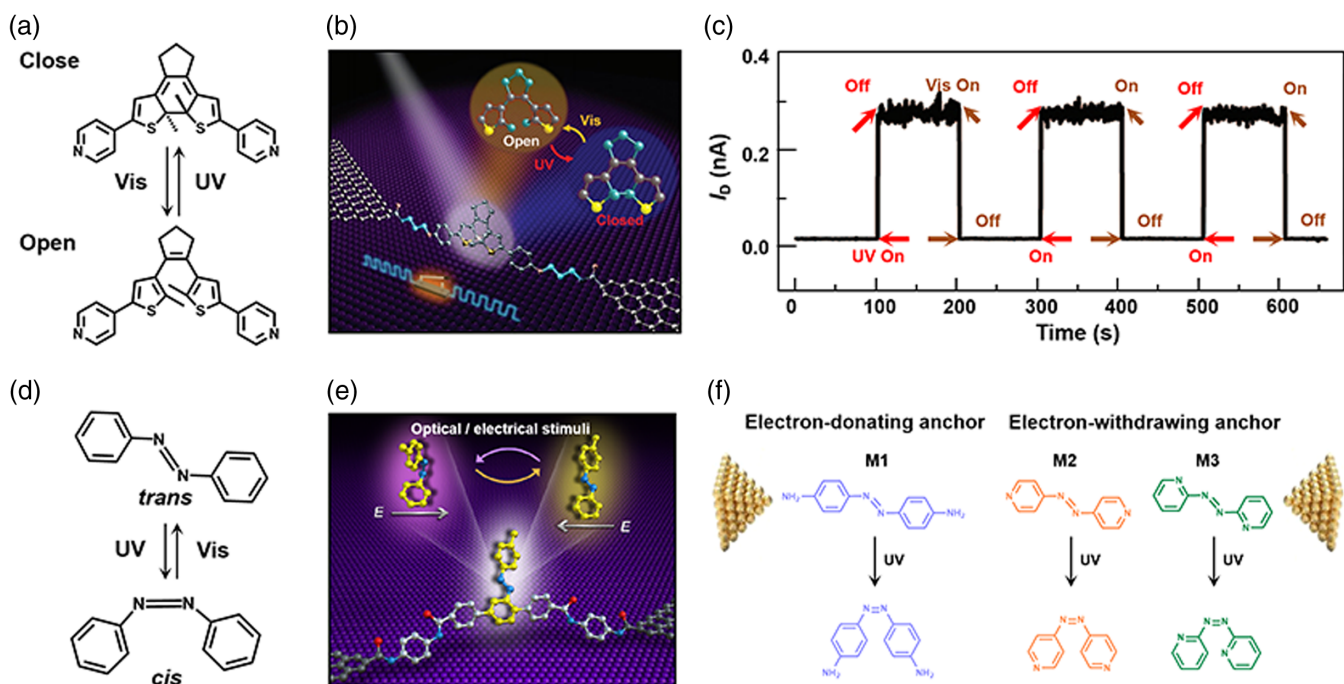


Fig. 5 Photo-induced isomerization switching. (a) Photochemically induced isomerization process of diarylethene molecule. (b) Schematic representation of the reversible switching of a single diarylethene molecule device. (c) $I-t$ curves of the molecule in its photo-induced open and closed ring states. (d) Schematic representation of the reversible *cis*-to-*trans* isomerization of azobenzene triggered by light irradiation. (e) Schematic representation of the reversible *cis*-to-*trans* isomerization of azobenzene-based single-molecule device. (f) Azobenzene single-molecule junctions featuring distinct side groups. [(a) Reproduced with permission from Ref. 83. Copyright 2011, American Chemical Society. (b), (c) Reproduced with permission from Ref. 84. Copyright 2016, American Association for the Advancement of Science. (d) Reproduced with permission from Ref. 90. Copyright 2013, Wiley-VCH GmbH. (e) Reproduced with permission from Ref. 91. Copyright 2019, Springer Nature Limited. (f) Reproduced with permission from Ref. 92. Copyright 2024, American Chemical Society.]

through molecular engineering to solve the electrode quenching problem, solving the reversibility problem of single-molecule switches. Diarylethene molecules are covalently attached to the electrode nanogap through amide bonds, resulting in a fully reversible photoisomerization-based single-molecule switch device [Figs. 5(b) and 5(c)]. The superior switching performance of this device firmly establishes a robust foundation for the utilization of diarylethene-based devices^{27,84,85} in the field of single-molecule optoelectronics. The reliability and efficiency of single-molecule isomerization can be significantly improved by utilizing diarylethene derivatives with diverse bridging groups, along with the application of gate voltage modulation to address challenges, such as electrode quenching. By integrating the field-effect function into the photoisomerization-based single-molecule switch device,^{88,89} the on/off ratio can exceed 3 orders of magnitude.

In addition, except for cyclization-decyclization photo-induced isomerization, *cis*-*trans* isomerism is another typical type of photoisomerization. *Cis*-*trans* isomers refer to isomers having the same chemical bond connection, but the functional groups at either end of the chemical bond are directional. Among them, when the functional groups are on the same side, it denotes as *cis*, otherwise, it is a *trans* isomer. Azobenzene molecule is a typical example. The reversible light-induced

cis-*trans* isomerization of azobenzene is facilitated by conjugated double bonds connecting two benzene rings, allowing for such a transformation under ultraviolet or visible light irradiation [Fig. 5(d)].⁹⁰ The *trans*-configuration of azobenzene possesses an approximately planar structure, whereas the *cis*-configuration has a bend of ~ 55 deg between the benzene rings, which enables azobenzene to rapidly and reversibly change the structure under illumination, thereby modulating its electric properties. Guo et al. attached azobenzene as a functional side chain covalently to graphene electrodes based on the photo-induced isomerization properties of azobenzene molecules, successfully creating a dual-mode single-molecule optoelectronic device responding to light. This single-molecule optoelectronic device realizes the switching function while ensuring its reversibility, expanding the research ideas for molecular switches [Fig. 5(e)].⁹¹

Based on the azobenzene molecular switch, the effect of anchor groups on the photo-induced *cis*-*trans* isomerization of azobenzene molecules has also been investigated by conducting real-time *in situ* conductance monitoring of the azobenzene molecule anchored with amino or pyridine groups [Fig. 5(f)].⁹² The experimental results show that with the continuous increase of ultraviolet irradiation time, the conductance change of molecular junctions with amino or pyridine anchor groups is opposite,

indicating the *cis*-configuration cannot turn into *trans*-configuration simply under the mechanical force. After further analysis, it is confirmed that the pyridine group can be excited by ultraviolet light ($n \rightarrow \pi^*$), while the amino group does not have this transition, revealing that the photoexcitation of anchor groups plays a crucial role in the *cis-trans* isomerization. These analyses break the traditional belief that the conductivity of azobenzene molecules is only determined by conformational changes during photoisomerization. The findings also provide a unique perspective for charge transport in single-molecule junctions with photo-induced isomerization and offer new avenues for the logical design of optomechanical synergistic molecular switches.

4.2 Photo-Conductance

The formation of photo-conductance encompasses the absorption of incident photon energy by the molecule, followed by processes, such as internal electronic transition, charge separation, and transportation, ultimately leading to conversion into electrical signals.^{93–95} The photo-conductance of single molecules exhibits significant potential in various applications within the fields of molecular optoelectronic devices.^{31,96} Furthermore, it enables us to delve deeper into the underlying microscopic mechanisms governing molecular optoelectronic effects, offering a clearer understanding of the intricate interactions between single molecules and photons. Up until now, several well-established mechanisms have been recognized for single-molecule photo-conductance. The first mechanism arises from inherent asymmetries within the molecular junction, leading to charge separation under illumination, which exhibits some similarities to donor–acceptor systems of semiconductors.³² The second is the formation of bound excitons in the junction.³³ The third is photo-assisted tunneling,^{35,97} while the fourth mechanism involves light-induced hot electrons in single-molecule devices.⁹⁸

The photo-conductance of charge separation arises from the large degree of molecular asymmetry.³² By designing asymmetric molecules with donor–acceptor structures, the transfer of electrons toward the acceptor moiety is facilitated under illumination. This process leads to the spatial separation of charges. The resulting charges migrate away from the site of initial electron transfer, either by hopping to nearby molecules or by migrating into the substrate, ultimately leading to enhanced conductance. For instance, a porphyrin- C_{60} dynamic molecule has been designed and subsequently connected to both ends of STM electrodes [Fig. 6(a)]. A laser beam, fiber-coupled and irradiated onto the molecule from beneath the STM, is employed to investigate its photo-conductance behavior. The electrons within the porphyrin moiety absorbed photonic energy, becoming excited and transferred to the fullerene segment, resulting in a remarkable charge-separated state. Notably, the recombination process of the separated charges is sluggish, enabling the molecular junction to maintain an enhanced conductance under illumination. Consequently, the ultimate conductance change after light excitation is reflected in the electrical measurements.

The bound excitons formed in symmetric molecules have also been shown to exhibit remarkable photo-conductance characteristics, which can be explained by the simplest model of molecular junction transport.³³ In this model, the HOMO or LUMO of the molecule is coupled to the Fermi level of electrodes. When the Fermi level of the electrodes lies closer to the HOMO than to the LUMO, conductivity is primarily governed by hole tunneling to the HOMO (analogous arguments

hold for LUMO-dominated transport) [Fig. 6(b)]. In the absence of illumination, the conductivity exponentially depends on the energy difference between the HOMO and the Fermi level of the electrodes. Under illumination conditions where the frequency resonates with or is proximal to the HOMO-LUMO gap, electrons within the HOMO are excited and transferred to the partially filled LUMO. Concurrently, a hole tunneling from the electrode to the HOMO becomes attracted to the charge present in the LUMO. Through Coulomb interaction, electrons and holes form tightly bound excitons. Concurrently, the Coulomb interaction induces a shift of the HOMO level toward the Fermi level, reducing the energetic disparity between the HOMO and the electrode's Fermi level [Fig. 6(c)]. This shift amplifies the possibility of electron tunneling from the electrode to the HOMO, ultimately leading to an enhancement in the conductivity of the molecular junction. Based on this, a perylene-3,4,9,10-tetracarboxylic acid diamide molecular junction has been constructed, and the results of photo-conductance reveal that, upon illumination, the molecular conductance increases from 1.25×10^{-5} S in the dark to 1.75×10^{-5} S, demonstrating a photo-induced conductivity enhancement of $\sim 40\%$, providing innovative avenues for the development and fabrication of highly efficient molecular optoelectronic devices.

Photo-assisted tunneling is an important way to enhance single-molecule conductance and has garnered extensive attention.^{34,97} Photo-assisted tunneling refers to the process whereby the transmitted electrons traverse the molecule under irradiation, emitting or absorbing photons of energy $h\nu$. This means that new conduction sidebands open in the molecular transmission function, which enhances the conductivity of molecular junctions.^{97–102} Xiang et al. investigated the photo-induced electronic transport properties of a single-molecule device based on the mechanically controllable break junction technique.⁹⁷ Photonic absorption significantly modifies the energy levels of the tunneling electron, leading to a notable enhancement in the conductance of the junction. Based on the experimental results, it is found that the photo-assisted tunneling has a strong dependence on the transmission function characteristics of the single-molecule device [Fig. 6(d)]. In recent years, with the constant diversification of research methodologies, a photoelectric phenomenon analogous to the photo-assisted tunneling mechanism has been uncovered, namely, the resonant transport mediated by molecular orbitals upon photonic absorption. This phenomenon is achieved when the energy difference between the transported electrons near the Fermi level of the electrodes and the molecular orbital energy levels is matched with the energy of photons.⁹⁹ Under such conditions, the transported electrons absorb the photonic energy and access the resonant transmission window of the molecular orbitals, leading to a significant enhancement in conductance [Fig. 6(e)]. For instance, the STM-break junction technique has been used to investigate the photocurrent mediated by a diketopyrrolopyrrole molecule, and notable photocurrent signals from the junctions stimulated by distinct wavelengths can be observed. By utilizing the interfacial energy matching model, the proposition is made that the observed photocurrent peaks originate from the resonance transmission enhancement. This enhancement is facilitated by the interaction of two resonant orbitals, specifically LUMO+1 and LUMO+2 [Fig. 6(f)]. Leveraging this mechanism provides a novel approach for characterizing single-molecule photoelectron tunneling spectroscopy with precise energy resolution.

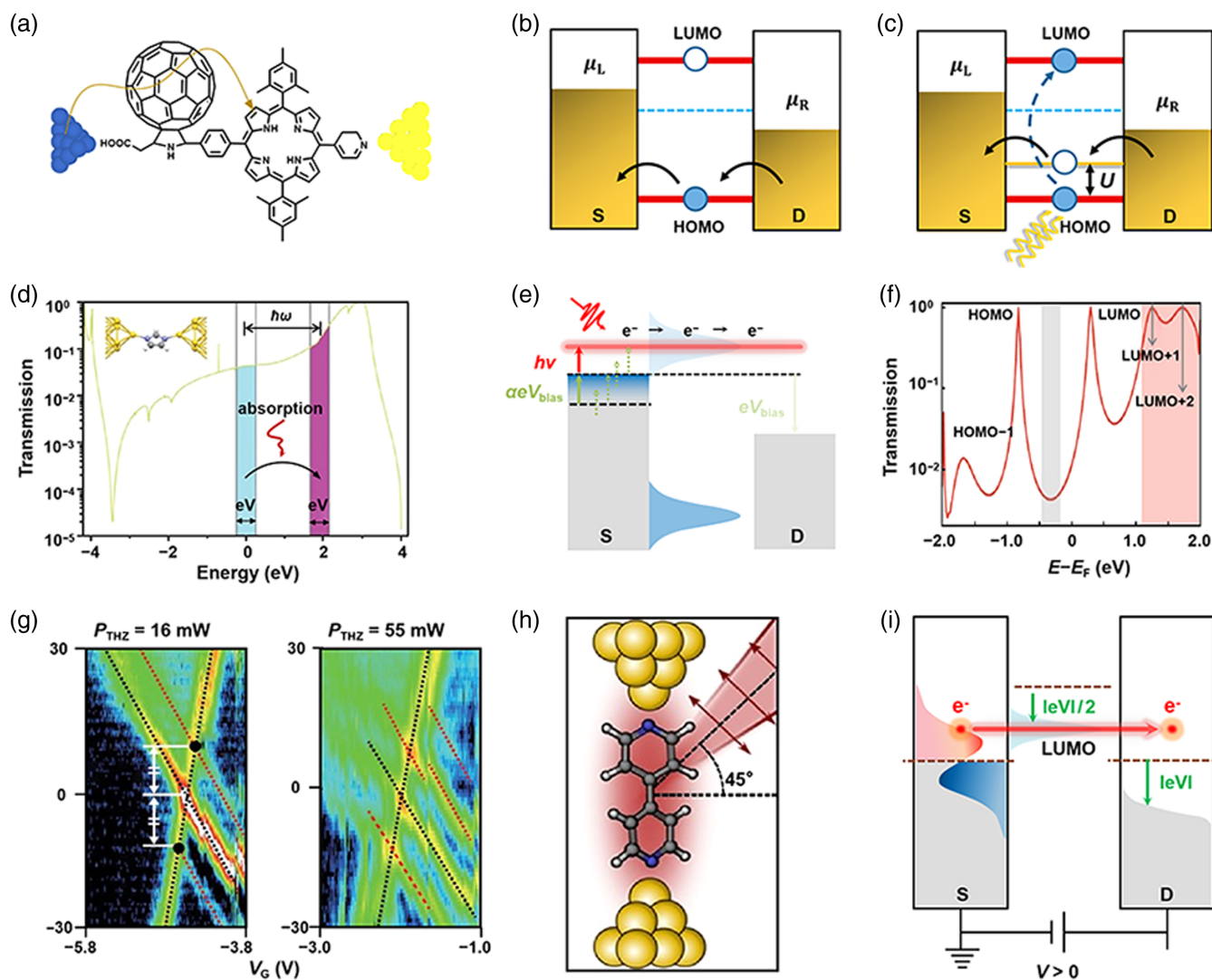


Fig. 6 Photo-conductance effects in single-molecule devices. (a) Schematic diagram of single porphyrin- C_{60} molecular junction. (b) Schematic representation of charge transport mediated by the HOMO. (c) Schematic representation of the charge transfer when an exciton is formed through photoexcitation. (d) A substantial shift in the electrode energy level in single imidazole molecular junction upon photon absorption. (e) Schematic representation of light-induced electron tunneling through frontier molecular orbitals. (f) Transmission spectra of the DPP molecular junction. (g) Coulomb stability diagram of a single C_{60} molecule transistor under THz irradiation. (h) Schematic demonstration of generating hot electrons in single 4,4'-bipyridine molecule junction. (i) Nonequilibrium distribution of hot electrons in a single-molecule junction. [(a) Reproduced with permission from Ref. 32. Copyright 2011, American Chemical Society. (b), (c) Reproduced with permission from Ref. 33. Copyright 2018, American Chemical Society. (d) Reproduced with permission from Ref. 97. Copyright 2021, Royal Society of Chemistry. (e), (f) Reproduced with permission from Ref. 99. Copyright 2023, Springer Nature Limited. (g) Reproduced with permission from Ref. 34. Copyright 2015, American Physical Society. (h) Reproduced with permission from Ref. 98. Copyright 2017, American Chemical Society. (i) Reproduced with permission from Ref. 103. Copyright 2020, American Association for the Advancement of Science.]

Moreover, irradiation with terahertz (THz) waves also involves the photo-assisted tunneling mechanism. For instance, in single C_{60} molecule transistors where the molecule is physically adsorbed between the gaps of electrodes, several parallel satellite lines emerge near the ground-state line in the Coulomb stability diagram when irradiated with THz waves of varying intensities [Fig. 6(g)].³⁴ The emergence of satellite lines

indicates the formation of photon sidebands. Furthermore, the increasing satellite lines with escalating light intensity imply that the additional tunneling pathways for transport electrons are established via multiphoton absorption.

The last one is hot-electron-based photo-conductance, which refers to the conductivity facilitated by hot electrons. Under illumination and thermal equilibrium conditions, hot electrons

possess an average energy exceeding the lattice thermal energy. A common technique for generating hot electrons in nanostructures and devices involves exciting surface plasmon resonance in metals within the nanostructure through irradiation with light of a specific frequency. Subsequently, this resonance induces nonradiative decay, resulting in the generation of hot electrons with elevated energy levels. The exploration of photo-induced hot electrons in single-molecule devices remains scarce, primarily constrained by the limitations in current experimental methodologies for detecting hot electrons. In several seminal experimental studies, the observed enhancement of photo-induced currents of molecular junctions has been attributed to photo-assisted tunneling, with the underlying mechanisms remaining controversial for a significant duration. Recently, studies have elucidated and confirmed the pivotal role played by hot electrons in boosting the conductance of single-molecule devices. For example, Fereiro et al. intuitively demonstrated the process of photoexcitation, inducing the generation of hot electrons and their subsequent contribution to the current.⁹⁸ The photocurrent spectra for large-area aliphatic and aromatic molecular junctions with copper top contacts are observed, even in the absence of molecular absorption. The molecule serves as a modulator of the tunneling barrier, and the hot electrons are generated within the copper electrode through the barrier via quantum tunneling, leading to the generation of a photocurrent. The charge transport in 4,4'-bipyridine-based single-molecule devices has been revealed to be primarily mediated by hot electrons [Fig. 6(h)]. The nonradiative decay of plasmonic energy leads to a nonequilibrium distribution of the generated hot electrons, characterized by an energy distribution that surpasses the Fermi level of the electrodes.¹⁰³ The close alignment between the LUMO and the energy level of the hot electron enables electron transport across the STM, effectively creating the conductive pathway [Fig. 6(i)]. In addition, it is found that when the lifetime of hot electrons is sufficiently prolonged and significantly exceeds the electron transfer time, the conductivity response of the device can be primarily attributed to the contribution of hot electrons.

The above-mentioned photo-conductance offers a comprehensive understanding of light-enhanced charge transport, paving the way for elucidating energy relationships at the molecule-electrode interface and precisely controlling the tunneling processes of electrons by leveraging the electronic structure of molecules. Furthermore, it provides valuable guidance for effectively optimizing the performance of single-molecule photo-electronic devices and fostering innovative functional device applications.

5 Ultrafast Optoelectronic Characterization of Single Molecules

Ultrafast lasers, distinguished by pulse durations that are shorter than molecular relaxation times, cover temporal scales ranging from picoseconds (10^{-12} s) to femtoseconds (10^{-15} s) and even attoseconds (10^{-18} s).¹⁰⁴⁻¹⁰⁷ Contributing to the emergence of ultrafast laser technology, the investigation of ultrafast dynamics has garnered considerable attention. This technology possesses two frequently utilized metrics. First, its remarkable temporal resolution, stemming from ultrafast pulse durations, facilitates the observation of transient electronic, atomic, and molecular dynamics. This capability provides valuable insights into chemical reactions, photophysical processes, and other phenomena.^{108,109}

Second, it offers an exceptional power density intensity within individual pulses, leading to widespread applications in high-energy physics and accelerator physics.^{110,111} Based on these, ultrafast lasers facilitate precise manipulation and processing of materials within ultrashort timescales, thereby catalyzing novel scientific research and technological advancements across various disciplines, including physics, chemistry, and biology.¹¹²⁻¹¹⁵

A pump/probe scheme based on ultrafast laser pulses has emerged as the paramount method for real-time visualization on internal structure of the material and dynamics.^{116,117} These techniques essentially involve the utilization of two laser pulses separated by a precise time delay. The earlier applied pulse with higher energy is denoted as the pump beam, initiating various physical phenomena within the sample, such as electronic excitation. Subsequently, a lower-energy pulse with a certain delay time, typically in the femtosecond range, is denoted as the probe beam, interacting with the sample previously exposed to the pump light. By quantifying variations in the transmittance and reflectivity of the sample to the probe light, this technology enables the extraction of dynamic insights into the excited state of the sample. A salient advantage of this method is the temporal resolution, which is primarily determined by the pulse duration, rather than being constrained by the bandwidth of photodetectors or electrical devices. In recent years, numerous studies have reported the integration of pump-probe technology into molecular systems, unveiling intricate physical processes, such as energy transfer, charge transfer, and vibrational dynamics.¹¹⁸⁻¹²⁰

Notably, these studies have primarily relied on ensemble-averaged measurements, disregarding the crucial fact that light-molecule interactions and photochemical reactions typically occur at the single-molecule level. By incorporating optoelectronic characterization approaches based on the interaction between ultrafast optical pulses and single molecules, it becomes possible to acquire a more precise comprehension of the intrinsic dynamic mechanisms, offering pivotal insights into the disparities that exist between single molecules and ensembles. In addition, it could open new avenues for the development of ultrafast coherent manipulation techniques for single electrons.

5.1 Ultrafast Light-Induced Single-Molecule Dynamics

Nowadays, decoding molecular structures through vibrational and rotational information has become a well-established technique in molecular characterization.^{121,122} However, most pertinent studies have focused on macroscopic molecules. The investigation of single-molecule vibrational dynamics is paramount for advancing quantum coherent manipulation at the molecular level. Lower energy ultrafast optical pulses such as THz pulses can be employed to unveil the inherent dynamic behaviors of single molecules. Some molecular vibration frequencies reside within the THz band, rendering THz radiation effective in exciting molecular vibrations.^{123,124} Therefore, leveraging ultrafast THz light sources to study single-molecule vibrations holds significant value in gaining a profound understanding of authentic vibrational information and revealing intriguing physical phenomena associated with molecular vibrations. The integration of ultrafast THz pulses generated by femtosecond lasers with single molecules in STM or nanostructures has yielded several breakthroughs in the study of vibrational dynamics.^{35,125,126} The introduction of ultrafast THz pulses into static single-molecule devices is a powerful tool

for investigating vibrational or rotational dynamics. Detection of these ultrafast vibrations can be achieved by monitoring changes in the current signal through the device. For instance, a time-domain THz autocorrelation measurement technique is employed to characterize the oscillation of a single C_{60} molecule adsorbed within an Au nanogap.³⁵ Two time-resolved THz pulses are focused on the C_{60} single-molecule device [Fig. 7(a)]. Subsequently, the evolution of the current signal in the device is recorded as a function of the time interval (t) between the pulses, revealing a distinct periodic oscillation pattern in the current signal [Fig. 7(b)]. The frequency of this oscillation is consistent with the oscillation frequency of the C_{60} molecular centroid, demonstrating the discrimination of THz-induced oscillations of the single C_{60} molecule at its inherent frequency. Numerous phonons are generated from the molecular oscillation

and can assist the electron tunneling, thus leading to electrical signal change. Specifically, electrons located near the Fermi level of the Au electrodes absorb energy from these phonons, enabling them to transition from the electrodes to the LUMO, thus charging the molecule. Concurrently, electrons within the molecule absorb phonons, facilitating their excitation from the HOMO to the other electrode [Fig. 7(c)], thereby establishing a cycle for electron transfer. This capability signifies the integration of ultrafast THz pulses with single-molecule devices, enabling the exceptional sensitivity required for real-time monitoring of single-molecule vibrational dynamics.

Ultrafast THz pulses can manipulate the conformational dynamics of single molecules based on the optical force effect. For instance, the THz pulses are employed to control the conformational oscillation of single magnesium phthalocyanine (MgPc)

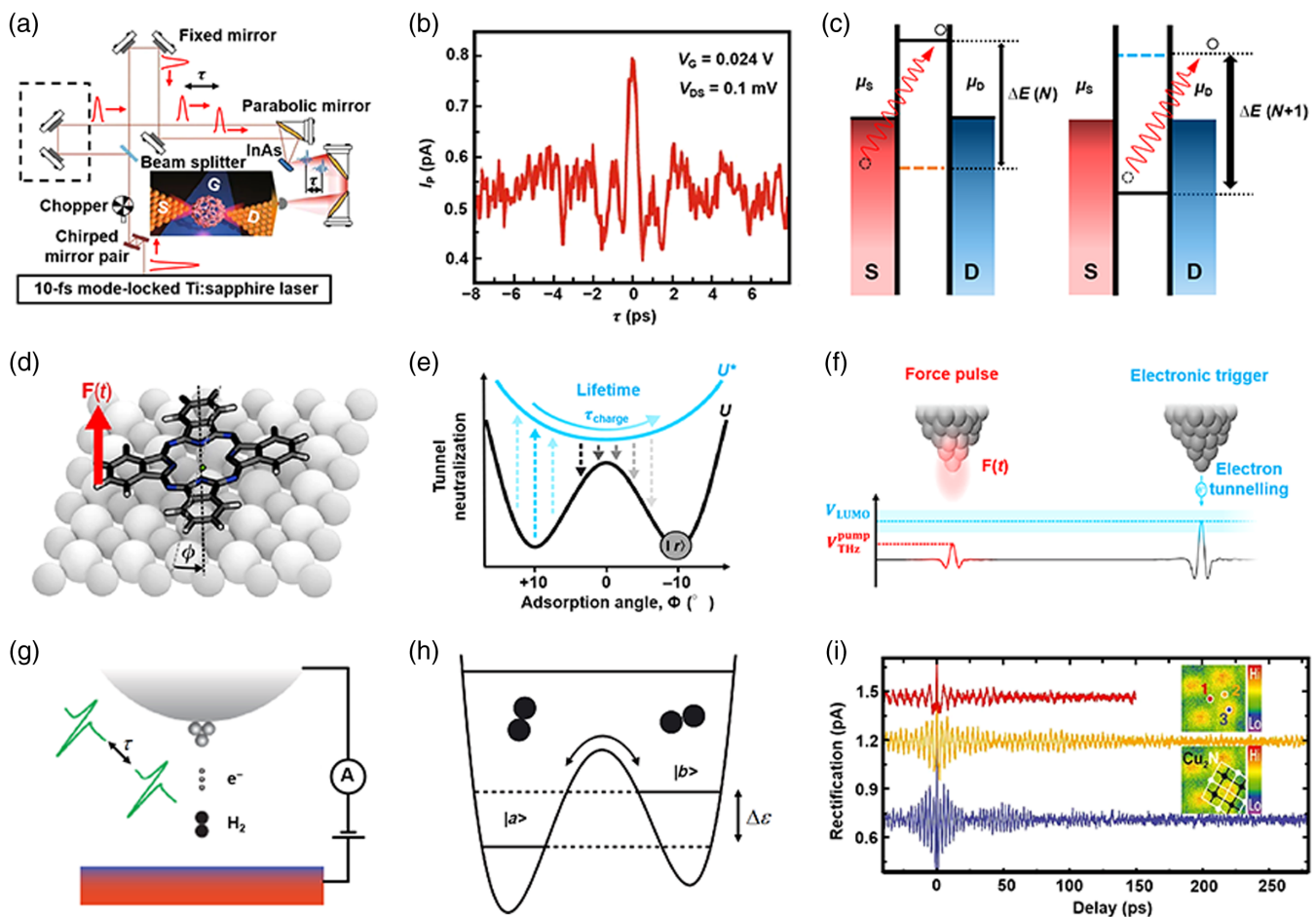


Fig. 7 Ultrafast light-induced single-molecule dynamics. (a) Schematic representation of the experimental setup of combining THz pulses with a single C_{60} molecule transistor. (b) Interferogram of photocurrent of the C_{60} transistor. (c) Schematic representation of the energy band of phonon-assisted tunneling. (d) Schematic illustration of a single MgPc molecule adsorbed on the substrate. (e) Potential energy diagram of the single MgPc molecule. (f) Schematic diagram of coherent control of switching probability of single molecules through THz pulses. (g) Schematic representation of THz–STM experimental setup. (h) Asymmetric double-well potential exhibited by a single H_2 molecule in STM. (i) THz rectification current of H_2 molecules with the tip at different lateral positions. [(a)–(c) Reproduced with permission from Ref. 35. Copyright 2018, Springer Nature Limited. (d)–(f) Reproduced with permission from Ref. 125. Copyright 2020, Springer Nature Limited. (g)–(i) Reproduced with permission from Ref. 126. Copyright 2022, American Association for the Advancement of Science.]

molecules adsorbed on the STM substrate [Fig. 7(d)].¹²⁵ Specifically, a pair of THz pulses is used. One with a high-intensity pulse designated as the probe pulse is focused at the STM junction. This probe pulse induces a transient bias effect, enabling resonant single-electron tunneling injection into the LUMO of the molecule. Consequently, the molecule is in a charged state where the molecular axis aligns along the direction of the NaCl substrate lattice ($\theta = 0$ deg). As the single electron rapidly tunnels into the substrate, the molecular axis relaxes into one of two equivalent ground-state configurations, designated as the “l” ($\theta = 10$ deg) or “r” ($\theta = -10$ deg) conformations [Fig. 7(e)]. These distinct conformations exhibit different tunneling current magnitudes. Through repeated measurements and statistical counting of the tunneling current, the probability of the molecule transitioning from the charged state to the “l” (or “r”) conformation can be determined.

The conformational transition probability exhibits robustness in the absence of external interference. Nevertheless, using a THz pulse of comparatively weaker intensity, which can be denoted as a pump pulse due to its generation of a subtle transient electric field, can interact with the molecule and serve as an ultrafast force to modulate this transition probability. The energy of the pump pulse must be controlled to remain below the threshold required for exciting electron injection into the LUMO, thereby ensuring that the conformational transition does not occur because of electron injection induced by the pump pulse [Fig. 7(f)]. By characterizing the relationship between the probability of conformational transitions and the delay time between the pump and probe pulses, the oscillations in the transition probability can be observed at a frequency of 0.3 THz. This oscillatory period is consistent with the planar vibrational frequency of the MgPc molecule, indicating that the pump pulse is the primary stimulus for conformational oscillation. Furthermore, the coherent inversion of the oscillating phase could be achieved by reversing the polarization of the pump pulse. The oscillation induced by the pump light comes from the ultrafast optical field acting on the molecule, effectively “kicking” it and causing it to oscillate within its ground-state potential well. This transient oscillation significantly alters the probability of the molecule relaxing from its charged state to its ground-state configuration. Ultimately, the ultrafast oscillations induced in the single MgPc molecule by the optical force effect are successfully realized.

In addition, the variations in the quantum state of a single molecule at the picosecond scale can be unveiled through real-time detection of THz-induced rectification currents. These variations are influenced by the immediate surroundings of the molecule, rendering the possibility of developing it as a quantum sensor to perceive the environmental conditions within nanoscale spaces. An STM, coupled with a pump–probe femtosecond THz laser, is utilized to demonstrate quantum sensing capabilities based on a single hydrogen (H_2) molecule [Fig. 7(g)].¹²⁶ The single H_2 molecule adsorbed on the substrate exhibits the double-well potential structure of the quantum ground state, corresponding to two distinct adsorption configurations [Fig. 7(h)]. When the THz pulses are focused within the STM, they facilitate ultrafast transitions between these configurations, thereby resulting in periodic oscillations of the molecule. The frequency of the oscillation is consistent with the energy separation between the two quantum ground states. The visualization of the ultrafast oscillation is achieved by recording the variations in the waveform of the rectified current as

a function of pump–probe time delays. Notably, the oscillation waveform exhibits significant variations, depending on the specific position of the tip on the substrate [Fig. 7(i)]. The oscillation frequency shifts 0.11 THz between position 2 and position 3 in the corresponding fast Fourier transform spectra. The inhomogeneous atomic distribution across different substrate locations results in varying dipolar moments exhibited by a single H_2 molecule adsorbed thereon. This alteration in dipolar moments modifies the energy separation between the two quantum ground states, subsequently leading to changes in the oscillation frequency of the rectification current. This finding demonstrates the potential of a single molecule as an ultrafast time-resolved probe for assessing surface chemical environments, offering a novel avenue to advance ultrasensitive sensing at the quantum level.

5.2 Ultrafast Modulation of Charge Transport

The investigation of the interaction between ultrafast light and single molecules facilitates a profound understanding of the inherent laws of charge transport within molecules, as well as exploring the influence of molecular energy level arrangement on their transport properties.¹²⁷ The ultrafast intense near-infrared laser pulses, focused on the tip of STM, have achieved ultrafast modulation and dynamic probing of electron tunneling processes. Due to the high intensity of the single pulse, the laser at the STM junction can elicit two distinct types of tunneling phenomena: photon-driven and field-driven tunneling.^{128,129} Below a specific intensity threshold, typically $\sim 10^{13}$ W/cm², rapid nonlinear enhancement can be observed in the laser-induced tunneling current. Nevertheless, upon exceeding this threshold, the nonlinear growth of the laser-induced current becomes increasingly less evident [Fig. 8(a)]. At lower laser intensities, electron tunneling is predominantly mediated by photonic interactions within the vacuum barrier. Electrons residing at the tip undergo the absorption process of two or more photons, transitioning into a spatially delocalized virtual state that lies above the Fermi level, thereby eliciting a photon-driven tunneling current [Fig. 8(b)]. Conversely, at higher laser intensities, the vacuum barrier is substantially suppressed by the intense laser electric fields. Electrons undergo direct tunneling between the tip and the substrate with the help of the intense laser electric fields, resulting in the generation of field-driven tunneling current [Fig. 8(c)].

Apart from the influence of relatively high-energy pulses on the modulation of electron tunneling in STM, lower-energy THz pulses can be employed to manipulate the electron tunneling within single molecules.³⁶ The electric field component of ultrafast THz waves is harnessed to induce transient bias effects. Specifically, electric fields of THz waveforms can modulate the static potential between the tip of STM and the substrate-adsorbed sample, effectively acting as transient bias voltage for opening the resonance transport window of molecular orbitals. The electron tunneling modulated by the bias voltage based on THz pulses is schematically illustrated in Fig. 8(d). At low THz intensities, where the transient bias voltage is relatively small, molecular orbitals fail to enter the resonant transport window. As the THz intensity increases, resulting in a higher transient bias voltage, the HOMO or LUMO molecular orbitals enter the resonant transport window, facilitating a charge transfer from the molecular orbitals. This method enables the temporary activation of resonant transport channels for molecular

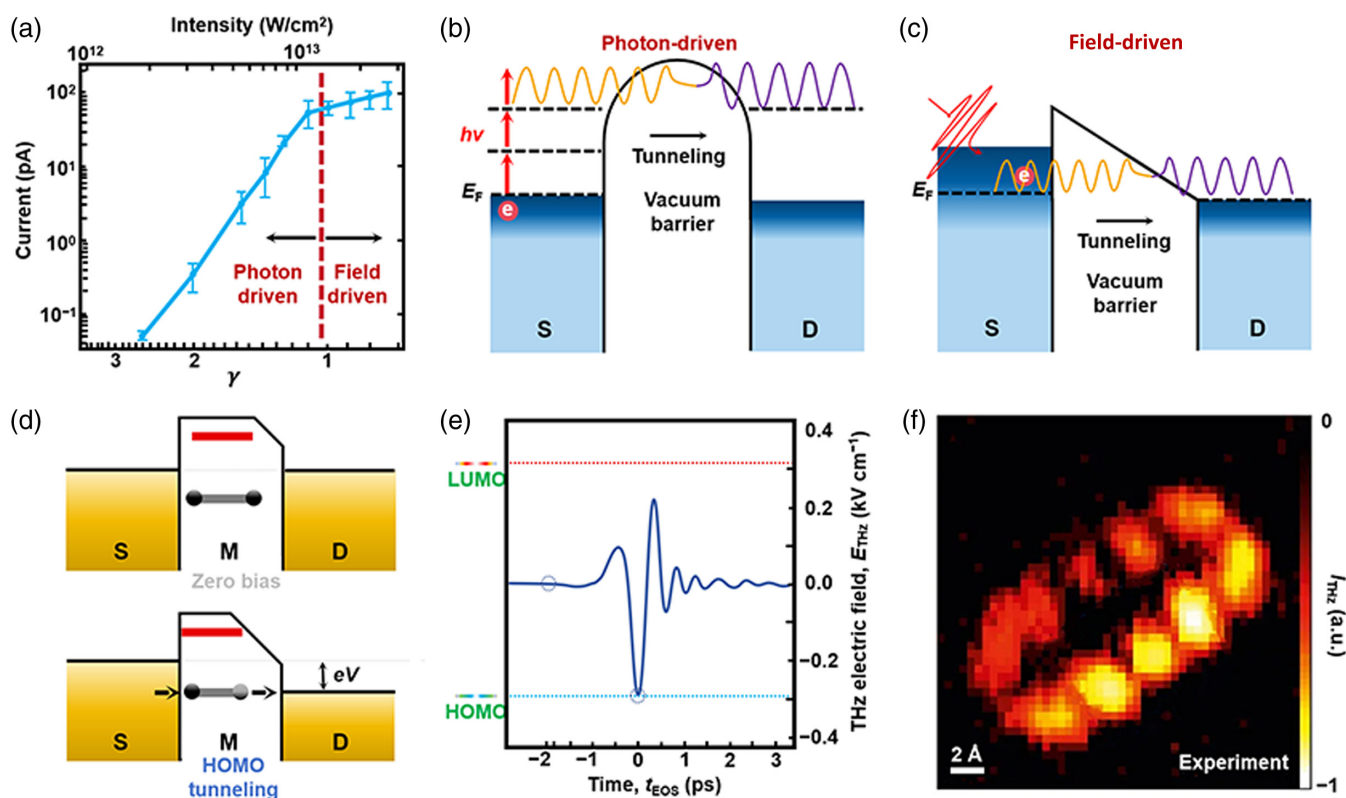


Fig. 8 Ultrafast modulation of charge transfer. (a) Schematic illustration of the laser-induced tunneling current as a function of increasing laser intensity. (b) Schematic representation of electron tunneling driven by photons. (c) Schematic representation of electron tunneling driven by fields. (d) Schematic illustration of ultrafast electron tunneling through the HOMO. (e) Bias effect modulated by the THz pulses. (f) THz-induced imaging of the HOMO density contours. [(a)–(c) Reproduced with permission from Ref. 127. Copyright 2020, American Association for the Advancement of Science. (d)–(f) Reproduced with permission from Ref. 36. Copyright 2016, Springer Nature Limited.]

orbitals within a duration as brief as 100 fs. By subsequently recording variations in the current magnitude of STM, the time of electron tunneling within single molecules can be determined [Fig. 8(e)]. Introducing the pump/probe scheme, where two successive, identical THz pulses are individually tuned in resonance with the HOMO, the single electron tunneling can be used to capture the ultrafast dynamics. In addition, the modulation of the THz waveform can also facilitate the selective tunneling of electrons through distinct molecular orbitals. Furthermore, leveraging the imaging capabilities of STM, the femtosecond-scale tunneling enables transient imaging of the orbital density distribution of single pentacene molecule with sub-angstrom resolution [Fig. 8(f)].

6 Conclusion and Outlook

In summary, this review provides a comprehensive overview on the optics, photonics, and optoelectronics of single molecules. The first part discusses the optical phenomena of single-molecule photoluminescence and Raman scattering separately. Then, the photonic properties based on the electroluminescence of a single molecule, as well as photonic applications of molecular devices, are enumerated. Subsequently, the optoelectronic properties of single molecules are introduced, encompassing the mechanisms of photo-induced isomerization and photo-

conductance of light-induced electronic state alteration. Finally, the significant progress achieved in the study of ultrafast dynamics and photoelectronic properties of single molecules based on ultrafast optical methods is described. These studies possess significant scientific importance and practical application value in revealing the mysteries of the microscopic world, especially those related to high-resolution imaging, molecular structures, and molecular dynamics. In addition, it provides new ideas for exploring the nonlinear optical properties and ultrafast photochemical processes of single molecules. Despite notable advancements in integrating single molecules and optical methods, the single-molecule world remains rich in novel properties and phenomena awaiting continuous exploration. In the following, we enumerate three new ideas in this field: the advancement of novel techniques, the investigation of novel physical and chemical properties, and the exploration of emerging applications. We eagerly anticipate the burgeoning progress of single-molecule research encompassing these cutting-edge ideas.

6.1 Develop Advanced Techniques

Vigorously developing more advanced research techniques in the single-molecule field that are suitable for different experiments and detection requirements can help to explore the

unknown field of single molecules in depth. To this end, we propose three advanced optical techniques: ultracold,^{130–134} ultrafast,^{135–143} and multimodal characterization.⁷⁶ These techniques not only enable the establishment of accurate multidimensional methods to analyze the properties of single molecules but also facilitate high-precision manipulation of single-molecule systems. The ultracold technique, based on the laser cooling approach, has long served as a pivotal starting point in the realm of quantum science. The origin of this technique can be traced back to the 1980s, during which scientists successfully cooled atoms and subsequently observed their inherent quantum properties. However, cooling molecules to a temperature level similar to that of atoms, in the face of their more complex energy landscape, is undoubtedly a highly challenging task. To overcome this challenge, the development of novel laser cooling techniques combined with single-molecule research is particularly crucial. This not only aids in the deep understanding of molecular quantum behavior but also provides new perspectives for exploring molecular dynamics. For instance, existing optical trap cooling techniques can be improved to be compatible with a wide range of single-molecule characterization platforms, enabling the detailed study of single-molecule dynamics, such as energy level transitions. At extremely low temperatures, the reduced molecular motion makes these processes more visible, potentially enabling us to uncover detailed structural information and interaction mechanisms within the molecular interior. Furthermore, as the temperature decreases, thermal excitation phenomena in the electronic system, such as electron–phonon scattering and electron–electron scattering, will also be suppressed. Simultaneously, various decoherence mechanisms, including spin-orbit coupling, will weaken, contributing to the prolonged maintenance of quantum decoherence. This creates more ideal conditions for us to study quantum coherence properties in single molecules.

Ultrafast technique, based on femtosecond laser pulses, has achieved remarkable success in elucidating physical and chemical kinetics of molecules on the femtosecond timescale.^{139,140} Nevertheless, electronic phenomena within molecules, including tunneling processes, typically manifest at timescales exceeding the femtosecond level, necessitating the development of faster measurement techniques for their observation. Attosecond laser technology has unlocked access to ultrafast electronic motion processes, establishing a formidable instrument background for further exploring the intricate nuances of the microscopic world. Currently, investigations on the interaction between attosecond lasers and molecules have facilitated precise measurements of fundamental processes, including photoelectron ionization and chemical bond formation within molecules.^{141,142} In addition, coherent manipulation of electrons at the nanoscale has also been achieved. Integrating the attosecond technique into single-molecule studies permits the real-time tracking of fundamental electronic processes, encompassing excited states, charge transport, and photoelectric effects.^{142,143} This will be of great significance for manipulating the material world at the level of single-electron motions.

The multimodal characterization technique for single molecules represents an integrated approach encompassing various advanced methods, with the aim of achieving highly sensitive and high-resolution detection and characterization of single molecules across multiple dimensions. By capturing and analyzing the multimodal signals emanating from single molecules, this technique elucidates molecular structures and functions

at the microscopic level. The combined single-molecule testing platform integrating optical and electrical excitation and detection is a powerful tool to characterize single molecules through a fusion of methods, such as stochastic optical reconstruction superresolution imaging, single-molecule spectroscopy, electric field-dependent experiments, and inelastic electron tunneling spectroscopy,^{24,133} facilitating research progress related to fundamental molecular properties, chemical reactions, and biophysics. However, given the plethora of physical and chemical properties and phenomena exhibited by single molecules, current multimodal testing remains insufficient to fulfill the demands of comprehensive characterization. Therefore, there is a pressing need for the integration of additional single-molecule approaches to propel significant breakthroughs in single-molecule research.

6.2 Explore Novel Properties

In recent years, with the rapid development of single-molecule technology, novel physical and chemical properties at the single-molecule scale need to be explored.¹⁴⁴ Among them, the aspect of single-molecule excited states, encompassing electronic, vibrational, and spin-excited states, is a promising area for investigation.^{145,146} Currently, research into these physical and chemical properties is primarily conducted at the macroscopic molecular level. Furthermore, chemical reactions involve changes at the single-molecule level, often requiring molecules to be excited from ground state to the excited state. Therefore, utilizing optical excitation of single molecules to study their excited-state properties is crucial for precisely understanding chemical mechanisms. This approach promises to distinguish diverse intermediate reaction species with single-bond precision, explore the regulation of chemical reaction rate-determining steps, and address common issues, such as bond forming/breaking selectivity, reaction pathway selection, and efficient energy transfer and conversion in elementary reactions. In addition, circularly polarized light possesses a unique sense of rotation, and upon interacting with the chiral and spin of the molecules,¹⁴⁷ the molecules generate different responses, which can be precisely detected by measuring the electrical signals of single-molecule junctions. The combination of optical methods and electrical characterization of single-molecule devices holds distinct advantages in characterizing the chiral and spin characteristics of single molecules, including high sensitivity, high resolution, noninvasiveness, and remote operability. This enables the chiral and spin features of molecules to be revealed without disturbing their natural state, thus gaining a more accurate understanding of their properties and functions.

6.3 Exploit Emerging Applications

Single molecules have been proven to have significant potential in the construction of nanoelectronic devices and the facilitation of high-computing applications.^{148–155} Nevertheless, their current capabilities are inadequate to meet the urgent demands of today's rapidly evolving information landscape. Therefore, it is of utmost importance to fabricate innovative single-molecule devices and attain unprecedented levels of computing capacity.^{153–156} It is firmly believed that quantum computing and photonic chips based on single-molecule devices are two viable avenues for attaining this objective.^{25,156} Single molecules, as the smallest stable quantum units, can exhibit rich quantum properties. A crucial aspect of advancing quantum

computing lies in the coherent manipulation of the inherent quantum properties of single molecules through microwave pulses. It is noteworthy that substantial advancements have been achieved in qubit manipulation utilizing single-molecule nuclear spin. These advancements include the initialization of spin states, the establishment and manipulation of quantum bits, as well as the read-out of spin quantum states.¹⁵⁶ However, the relatively short coherence time of quantum systems poses a challenge to achieving reliable long-term quantum manipulation. Developing suitable new molecular systems and further promoting quantum manipulation to practical computing applications are key issues that need to be addressed in the future. Single-molecule photonic chips, leveraging the electroluminescence of single molecules and employing light waves as the medium for data transmission, storage, and computation, exhibit significant advantages, including superior transmission speeds and expanded bandwidths.²⁵ It is feasible to design arrays of single-molecule devices with electroluminescent properties, as well as molecular constructs exhibiting diverse fluorescence and phosphorescence emissions. Through selective induction of emission from specific single-molecule devices, selective optical signal outputs can be achieved, enabling the realization of logical operations. Currently, the research on photonic chips is still in its experimental infancy, facing challenges in large-scale integration. Extensive research is required to promote their development towards practical applications.

Conflict of Interest

The authors declare that they have no competing interests.

Author Contributions

X.G., C.J., and D.X. proposed the concept and framework of this work. All the authors contributed to the writing, review, and editing of the paper.

Code and Data Availability

Data sharing is not applicable to this article as no datasets were generated or analyzed during the current study.

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