On-Chip Nanoscale Light Source Based on Quantum Tunneling: Enabling Ultrafast Quantum Device and Sensing Applications

Received 16 November, 2020; accepted 21 December, 2020

Jihye Lee^{a,b} and Jong-Souk Yeo^{a,b,*}

^aSchool of Integrated Technology, Yonsei University, Incheon 21983, Republic of Korea

^bYonsei Institute of Convergence Technology, Yonsei University, Incheon 21983, Republic of Korea

*Corresponding author E-mail: jongsoukyeo@yonsei.ac.kr

ABSTRACT

Light sources are ubiquitous in our lives. Common light sources include sunlight and incandescent lamps, which emit visible spectrum to help us to see surrounding objects via blackbody radiation. Further understanding and control on the quanta of light, called photons, have led to numerous advances in nanophotonics and related research fields, but recently their impact has been most notable in quantum computing, communications, biosensing, and imaging technology. Most of these applications have been made possible through the development of ultrasmall and ultrafast light sources based on advanced nanotechnology. In this review, we aim to give a clear picture of the historical achievements regarding light sources and how recent studies have developed the field of ultrasmall light sources, especially in relation to quantum electron tunneling mechanisms. Finally, we discuss the potential applications for emerging quantum devices and sensing technologies.

Keywords: Light source, Quantum electron tunneling, On-chip quantum devices, Biosensors, Chemical sensors

1. Introduction

The story of the development of light sources begins before Edison's invention in 1879 [1, 2]. The Italian physicist, chemist, and pioneer of electricity and power Alessandro Volta invented the voltaic pile in 1799 by using zinc and copper disks with layers of cardboard soaked in salt water and connected to the glowing copper [Fig. 1(a)] [3]. It is considered the earliest battery, as well as an early incandescent lighting concept. Subsequently, Humphry Davy in 1815 and Warren de la Rue in 1840 demonstrated a very bright arc lamp and expensive platinum-filament-based bulb, respectively [Fig. 1(b)], but Joseph Swan and Thomas Edison made efforts to change these concepts to commercialize the electric light in practical use over the candle, gas light, and oil lamp [Fig. 1(c)] [4–7]. Swan first used a carbonized paper filament in a vacuum glass bulb, but it had a short lifetime (15). Edison realized that the major problems with Swan's bulb were the filament and the lack of a good vacuum state; therefore, he changed the filament material to electrically high-resistance tungsten and changed the vacuum state to a gas-filled state [8]. This made a significant difference. Because there was no oxidation, the bulb had a long lifetime, leading to great commercial success. His numerous trials and endeavors have had a profound impact on the modern industrialized world that illuminates our lives. Light-emitting diodes (LEDs), which were developed by Henry Joseph Round (first observation of electroluminescence from solid-state material, 1907), Oleg Losev (first invention of an LED, 1927), James R. Biard (first infrared LED from a tunnel diode, 1961), Nick Holonyak (first visible red LED, 1962), and Shuji Nakamura (high-brightness blue LED, 1993), now provide energy-efficient and long-lasting lighting with small size and fast switching speed [Fig. 1(d)] [8–10]. Thus, they can be used in diverse applications including advanced communication technology and medical devices

in addition to general lighting [9, 11]. An LED is a semiconductor light source that emits energy in the form of photons by recombining electron and holes in the semiconducting layer. This means it is categorized in electroluminescence. The wavelength of electroluminescent light is determined by the bandgap of the solid-state material, and the emitted light is spectrally and spatially incoherent compared to a coherent laser light source. These types of LED have become commercialized not only in energy-saving lighting but also in high-contrast-ratio pixels in displays [12]. Cavity structure and optoelectronic considerations are added to the design of LED to enable light amplification by the stimulated emission of radiation (LASER) with a narrow temporal (monochromatic) and spatial (collimated) frequency range. More coherent white light source can be made by combining laser sources of red, green, and blue [Fig. 1(e)] [13–15]. This type of coherent light source can also be used for the applications related to quantum physics.

With the recent development of nanotechnology, light sources have gradually become smaller, and now nanoscale light sources that can provide the potential to integrate subwavelength optics are being developed. In the late 1990s, metallic-tip-based fiber-optic scattering probes were developed that can be applied to near-field imaging [Fig. 1(f)] [16, 17]. In 2007, Nakayama *et al.* demonstrated electrode-free nanowire-based coherent light sources in the visible wavelength range [Fig. 1(g)] [18]. These were formed by low-toxicity and chemically stable materials at room temperature, thus enabling subwavelength imaging in a liquid-based environment for lab-on-a-chip technologies. Recently, metal-semiconductor hybrid structure was proposed as a tunable, nanoscale, and incandescent light source made by selective materials that can absorb heat and emit light [Fig. 1(h)] [19, 20]. This means that thermal radiation is engineered to generate the light, thus providing potential in applications related to infrared detection and sensing

Figure 1. (a) Voltaic pile with zinc, copper, and cardboard soaked in salt water. ©1996 IEEE. Reprinted with permission [3]. (b) First electric lamp by Davy and Faraday [6]. Reproduced with permission of IOP Publishing. (c) Swan (left) and Edison (right) filament lamps [7]. (d) Development of the LED lamp from 1907. Photograph of a white LED lamp. Adapted with permission [10]. ©2017, Wiley (e) Mixing of color with R, G, and B lasers to make white light. Image reproduced with permission [36] under the terms of the Creative Commons Attribution 4.0 International license. (f) Sub-micrometer-sized metallic tips for near-field scanning optical microscope imaging; photograph of scattering of an evanescent field at a probe tip. Adapted with permission from [16] ©1994, The Optical Society. (g) Demonstration of a nanowire light source, which can be tunable as well as coherent [18]. ©2017, Nature Publishing Group. (h) Metal-insulator-semiconductor (MIS) structure for an incandescent light source. Adapted with permission [20]. ©2019, Wiley. (i) Selectively generated photon emitters in 2D materials. Image reproduced with permission [21] under the terms of the Creative Commons Attribution 4.0 International license. Other images are also reprinted by following individual copyright rules.

with high brightness and directionality, and optical switches potentially for the next generation of switches beyond silicon-based transistors. Klein *et al.*, in 2019, demonstrated nanoscale light sources for quantum computers [Fig. 1(i)] [21]. This demonstration was possible because of the defects of atomic layers of two-dimensional materials, which trap the excitons that can emit light [22]. This development can be used to replace electron-based devices with much faster photon-based devices by integrating the quantum light sources (single-photon emitters) with optical waveguides and circuits. Single-photon emission can also be exhibited by fluorescent atomic defects [23] such as nitrogen vacancy centers [24, 25], semiconductor quantum dots [26], and atomically thin two-dimensional layers with strain-induced wrinkles, ion-beam-induced defects, or etching [22, 27]. These types of material can be coupled to extremely small optical cavities, thereby maximizing their emission efficiency and controlling their directionality [28–30]. Other approaches to producing light at nanoscale include quantum-tunneling-based devices [31–34]. These approaches are currently being actively researched owing to their fast response time in the range of ~ 10 fs and easy controllability of wavelength and emission pattern based on the charge injection and coupled nanostructure. Through a metal-insulator-metal (MIM) or metal-insulator-semiconductor (MIS) structure, inelastic electron tunneling generates a photon within a junction and it can couple to another structure to exhibit efficient radiation to free space [31]. Furthermore, the quantum emitters can be coupled to an extremely small cavity structure to enhance the emission of light at the nano- and sub-nanometer scale [35].

In the following section, we review the recent advances related to quantum-tunneling-based light source devices based on their structure, materials, and functions. Finally, Section 3 briefly introduces the potential applications based on nanoscale light sources.

2. Quantum-electron-tunneling-based light-emitting devices

Recently, as the development of “Internet of Things” technology has rapidly increased demand for the amount of data to be processed for a hyper-connected world, there has been a corresponding growing demand for advanced materials and devices with information-processing speeds orders of magnitude faster than those of existing technology [36–38]. This demand has reignited interest in inelastic-electron-tunneling-based internal light sources that enable ultrafast transduc-

tion [36, 37]. However, for practical applications in data processing, telecommunication, optical interconnects, and optical sensing, the transduction efficiency of electrical-to-optical signal conversion at nanoscale is very low, with the current world record set at approximately 1 % in 2018 [39]. Furthermore, flexible modulation of bandwidth and multi-frequency generation for multi-channel on-chip platforms remains challenging. These challenges also apply to optical biosensing applications, for which efforts have been made to use the compact size of nanoscale sensors for practical point-of-care (POC) devices [40, 41]. In an effort to achieve progressively smaller reagent concentrations and sensing volumes, these approaches have experienced a paradigm shift from simple bulk measurements toward engineered nanoscale devices [37, 42]. In this size regime, plasmonic particles and nanostructures provide an ideal toolkit for the realization of novel sensing concepts due to their unique ability to simultaneously focus incident light into subwavelength hotspots and transmit minute changes in the local environment back into the far field as a modulation of their optical response. However, these optical sensors currently still rely on bulky light sources, such as LEDs or lasers, and detectors, limiting their usability in biochemical research and medical diagnostics in which miniaturized and/or integrated sensor devices are crucial for POC applications [43, 44]. In particular, one of the key components missing from current optical sensing approaches is a reliable and nanoscale light source, which can be directly integrated with nanophotonic sensing elements to detect target analytes in a miniaturized package.

In classical mechanics, electrons with insufficient energy to overcome a given potential barrier have zero probability of reaching the other side. However, when taking into account the uncertainty principle of quantum mechanics, there is a small probability for the electron to pass through the barrier with a reduced amplitude of the electron wave function [45, 46]. Regarding this quantum tunneling effect, there are two pathways for the electron to take through the tunneling junction [47, 48] [Fig. 2(a)]: elastic or inelastic electron tunneling. When electrons pass through the quantum-tunnel junction elastically, they connect electronic states of the same energy between the two electrodes and therefore do not lose any energy. Most quantum electron tunneling processes involve this elastic behavior. In inelastic electron tunneling, a rarer form of quantum electron tunneling, the transmitting electron loses some of its energy to excite a surface plasmon between the thin insulator tunnel junctions. The excited surface plasmon can either decay into the far field radiatively or decay non-radiatively by excitation of a hot electron. This optical mode can be tuned by 1)

Figure 2. (a) Basic principles of elastic and inelastic electron tunneling. (b) First experiment of quantum-tunneling-based light emission from a metal-insulator-metal (MIM) tunnel junction. Adapted with permission [32]. ©1977, American Institute of Physics. (c) Roughened tunnel junction for photon emission. Reprinted with permission [49]. ©1979, American Physical Society. (d) Thin-film-based transducer that converts electrical signal into plasmonic signal for on-chip generation of plasmons, and vice versa for on-chip detection for plasmons [39]. ©2017, Nature Publishing Group. (e) Scanning tunneling microscope (STM) light emission from atomic silver chains. Image reproduced with permission [54]. ©2009, The American Association for the Advancement of Science.

the bias voltage that determines the cutoff frequency of the device, 2) the structure design of the MIM geometry, and 3) the antenna geometry and its configuration. In this section, we summarize the state of the art on inelastic-electron-tunneling-based devices.

2.1. MIM structures for on-chip generation, manipulation, and detection of plasmons

It has been shown that high-frequency broadband light can be generated based on inelastic electron tunneling. The cutoff frequency of this broadband light is determined by the applied voltage, as described by the quantum relation $h\nu_{\text{cutoff}} = eV_{\text{bias}}$ [31]. Following this discovery, from 1976 to 1979, there was extensive exploration by quantum-tunneling-based studies [31–34, 49]. In 1976, Lambe and McCarthy discovered a new concept of generation of light by using a metal (Al)-insulator (Al_2O_3)-metal (Au) thin film structure [31]. This first investigation demonstrated the possibility of direct transduction between electrons and photons, in which the excess energy of the tunneling electrons can generate light via the radiative decay of plasmon excitations. Subsequently, they added a plasmon structure onto the top electrode, which demonstrated scattering of non-radiative optical fields from the quantum-tunnel junction, thus proving the occurrence of enhanced plasmon-photon coupling at 77 [Fig. 2(b)] [32]. Another group, Larks *et al.*, calculated the effect of surface roughness on the mean free path of surface polaritons for photon emission [Fig. 2(c)] [49]. They cited Lambe and McCarthy's findings that increased roughness results in strong damping of surface polaritons. This indicates that a roughened tunnel junction increases photon emission. Following these previous studies, in 2017 the same structure and material selection evolved into a highly efficient on-chip transducer exhibiting both generation and detection of plasmons [Fig. 2(d)] [39]. Owing to the direct conversion of electrical signal to surface-plasmon polaritons (SPPs), they achieved ~14 % efficiency with high operational frequencies in the region of 300–350 THz and a broadband spectrum that enables fast operational on-chip plasmonic circuits. However, the input sources of these structures are restricted by the electron injection through the source meter. This electron source can be changed when a scanning tunneling microscope (STM) is exploited in the STM community [50–53]. The probe tip enables atomic-scale

spatial imaging, through which Chen *et al.*, in 2009, resolved the emission pattern of a silver atom chain on a nickel-aluminum alloy surface with sub-nanometer resolution [54]. They measured the dI/dV spectra along the chain to understand the spatial distribution of the local density of states (LDOS), thereby imaging the radiative electronic transition state correlated to the emitted light probability, as shown in Fig. 2(e). Further, studies on the STM-based photon emission pattern were expanded to the morphology of the surface of the structure, such as its height and width [50]. According to Shkoldin *et al.*, a decrease in height and increase in width of the gold grains enables photon emission from the tunnel junction. In other words, the surface quality is crucial to overcoming the low emission efficiency of quantum-tunneling-based devices.

2.2. Optical antenna structure coupled to the quantum tunnel junctions

Optical antennas can manipulate and control light at subwavelength scales [55,56]. Generally, they can be used for energy transfer between a source (or receiver) and the free-radiation field, that is, transduction into the far field [57,58]. Recently, optical antennas have been shown to couple to quantum-tunnel junctions and to convert electrons into free-space photons efficiently. In this configuration, the optical antenna plays a crucial role in bridging the size mismatch between far-field radiation and nanoscale volumes, and in strongly enhancing the transduction between electrons and photons [59,60].

In this section, we first discuss the vertical MIM junction as shown in [Fig. 3(a)–(c)]. In 2015, Parzefall *et al.* studied antenna-mediated photon emission from hexagonal boron nitride (h-BN) vertical tunnel junctions [61,62] [Fig. 3(a)]. This research group first combined 3D optical nanoantennas with a 2D material and modulated the frequency up to 1 GHz using this hybridized structure. Although the efficiency of the emitted light from the junction was low, the emitted light was strongly polarized in the direction of the short axis of the antenna slot. This was the first demonstration that dipolar radiation generated by inelastic electron tunneling can be modulated by the optical characteristics of the optical antenna. They further demonstrated a 2D-material-based vertical MIM using h-BN and graphene

Figure 3. Vertical and lateral antenna structure coupled to the quantum tunnel junction. Vertical MIM. (a) Rectangular slot of optical antenna with single-crystalline hexagonal boron nitride (h-BN) multilayer [61], ©2015, Nature Publishing Group. (b) h-BN and graphene stacked with nanocube antenna. Image reproduced with permission [62] under the terms of the Creative Commons Attribution 4.0 International license. (c) Template dielectrophoretic trapped nanoparticle optical antenna. Reprinted with permission [58]. ©2019, American Chemical Society. Lateral MIM. (d) Light emission by Yagi-Uda antenna, Image reproduced with permission [67] under the terms of the Creative Commons Attribution 4.0 International license. (e) Directional and unidirectional emission of light by V-shape antenna, Reprinted with permission [69]. ©2017, American Chemical Society. (f) Atomically smoothed tunnel junction between highly crystalline nanocubes [73]. ©2018, Nature Publishing Group.

with a nanocube antenna [Fig. 3(b)] [62]. This nanocube antenna provided resonant enhancement of photon emission with narrow frequency based on a Purcell effect that can increase the mode density. The photon emission rate was thus enhanced. In 2018, Namgung *et al.* also combined the 2D material graphene with a quantum-tunnel junction and coupled metal nanoparticle antenna to induce gap plasmons [63]. This configuration allowed wide-frequency tunability ranging from the near-infrared to the visible range by using an additional dielectric layer combined with the nanoparticle structure [64, 65]. To form this versatile nanoparticle antenna in the MIM structure, He *et al.* used the dielectrophoretic trapping method to accurately position the thiol-covered particles [66]. This method not only provides controllable positioning of the antenna on the devices but also maintains the stability of the insulating molecule layers on the particle. The field-driven dielectrophoresis method was also used in Yagi-Uda antenna configuration to create a gap of a few nanometers between the metals [67]. This is illustrated in Fig. 3(d).

The configuration of the MIM junction can also form a lateral structure, as shown in [Fig. 3(d)–(f)]. The size of a lateral-type quantum-tunnel junction is controlled and fabricated by several smart methods and engineering of interface or crystallinity control. In 2015, Kern *et al.* demonstrated electrically driven optical antennas via the broadband quantum-shot noise of electron tunneling [68]. This configuration comprises a lateral tunnel junction with an air energy barrier made by the drop-casting method, by pushing a cetyltrimethylammonium-bromide-shelled nanoparticle between the focused-ion-beam-milled antenna structure. The spectrum of a quantum-tunnel junction with an antenna structure is mainly defined by the antenna geometry and the bias voltage through the two electrodes. In 2020, this group further explored the Yagi-Uda antenna for large directionalities as reported by Kullock *et al.* [Fig. 3(d)] [67]. The lateral junction was formed by field-controlled dielectrophoresis, which exhibits superior reproducibility and stability to the drop-casting method. Depending on the voltage and frequency of the applied electric source, particles are attracted to the specified position of the highest field gradient. Using this optimized fabrication method, highly unidirectional emission was achieved by adding the reflector and director between the active quantum-tunnel junction. This indicates that constructive interference in the forward direction and destructive interference in the backward direction are both maximized for unidirectional emission.

In 2017, Gurunaryanan *et al.* demonstrated an in-plane nanoan-

tenna structure with a V-shape to provide broadband emission of light [Fig. 3(e)] [69]. Depending on the shape and size of the antenna and its angle between them, the directivity of the emitted light pattern is tuned passively. The antenna cutoff frequency is limited by the applied bias voltage. This type of antenna is made via the electromigration method [70–72]. Electromigration is the transport of materials according to the momentum transfer between electrons and diffusive metal atoms. It rearranges the atoms in a weak junction, thus resulting in a nanogap in the antenna structure.

Most research efforts have been focused on the nanoantenna structure. In 2018, Qian *et al.* demonstrated efficient light emission through a silver nanocube antenna structure by engineering of the material property [Fig. 3(f)] [73]. They reported that single-crystalline material has lower plasmonic loss compared to amorphous and polycrystalline structure, and can thus be a key parameter in enhancing light emission performance. Using this single-crystalline silver nanoantenna structure, the researchers aligned it by edge-to-edge assembly to maximize the LDOS, thereby increasing the far-field light emission efficiency to ~2 %.

2.3. Alternative materials ranging from molecules to 2D materials

Most studies have attempted to create photon emitters using a standard metal and insulating materials. In this section, we introduce the alternative materials that can replace them in MIM and new MIS structures for light-emitting devices.

In 2016, Du *et al.* demonstrated highly efficient on-chip direct electronic-plasmonic transducers and molecular plasmon sources based on a self-assembled monolayer (SAM) tunnel junction [Fig. 4(a)] [74, 75]. This group used a similar thin film structure but increased the efficiency of the quantum tunneling by direct conversion of electrical signals into SPPs with an efficiency of approximately 14 %. When a molecule is used for this type of on-chip integrated plasmonic device, bias-sensitive excitation of a plasmon is exhibited depending on the symmetry of the chemical structure. Through the electrical measurement of different symmetry molecules, it was found that molecular through-bond tunneling is crucial to excite and control the plasmon, thus enabling single-molecule-level plasmonic devices and circuits at the on-chip scale. According to their further studies on SAM-based devices, the typical angle of an even number of carbon in SAM is 45°, whereas that of an odd number of carbon is 30° [Fig. 4(b)] [75]. This angle difference has a significant effect on the radia-

Figure 4. Various types of materials in a quantum-tunnel junction and its electrode. (a) Symmetric and asymmetric molecule chains between metal layers for an MIM device [74]. ©2016, Nature Publishing Group. (b) Chemical-structure-dependent radiation pattern from MIM. Reprinted with permission [75]. ©2019, American Chemical Society. (c) Metal-polymer-metal (MPM) tunnel junction using a monolayer of poly-L-histidine (PLH). Reprinted with permission [76]. ©2020, American Chemical Society. (d) MIS device for solid-state light sources. Reprinted with permission [79]. ©2018, American Chemical Society.

tion pattern. The direction of a tunneling electron in an even number of carbon is larger than in an odd number of carbon chain, thus generating a large directional SPP at an even number of carbon chained molecule structure. This achievement of unidirectionality control is possible without any directional nanoantenna structure in MIM devices. The molecule insulating layer can be changed to a monolayer for a polymer. In 2020, Wang *et al.* demonstrated a metal-polymer-metal (MPM) structure using poly-L-histidine (PLH) as a tunnel barrier [Fig. 4(c)] [76]. This plays a role in the tunneling layer allowing electron flow and in the storage layer storing and erasing the information via chemical reaction, thus providing scope for future neuromorphic devices. The chemical reaction in this layer is a hot-electron-mediated reaction; therefore, the PLH layer near the nanorod tip begins oxidative dehydrogenation and finally changes the tunnel barrier, which enables changes to both plasmon excitation and light emission. Using this smart polymer, this MPM device can further tune the light intensity and wavelength according to the bias voltage.

There have not only been changes to the insulating layer via molecules and polymers, but also several attempts to change the configuration from MIM to MIS [77–79]. Göktas *et al.* changed the configuration by using silicon [Fig. 4(d)] [79]. They fabricated a silicon-based MIS device to use its indirect bandgap structure, which prevents direct recombination of the carrier. Furthermore, the MIS structure can couple the internal field enhancement in the junction (large LDOS) with an external k -vector matching condition (matching between nanoscale volume field enhancement and far-field radiation to the air) to enhance the quantum efficiency, thus providing complementary metal-oxide-semiconductor-compatible and silicon-based on-chip light sources at room temperature. The semiconductor layer can be changed to the chalcogenide-based 2D material. Bharadwaj *et al.* coupled a Au dimer antenna to excited excitons in a MoS₂ layer to record the luminescence [80]. When the two particles are aligned with a gap, the maximum luminescence is exhibited because the field enhancement by the particles aligned along the vertical is not absorbed by the horizontally flat 2D material but coupled to the MoS₂ exciton.

3. Applications and summary

The new concept of an internal light source can be applied in various fields, such as on-chip photonics, optical communications, bio- and chemical sensing, and optically and electrically operating memory and switching devices [81–83]. In this section, representative applica-

tions based on quantum tunneling devices are explained and summarized.

When high-density metamaterials are used for inelastic-quantum-tunneling devices, they can be used to monitor the chemical reaction in real time with high sensitivity. Wang *et al.* fabricated large-area and high-density metamaterials to induce a large flux of hot electrons through an MIM junction [Fig. 5(a)] [84]. The induced hot electrons make the tunnel junction reactive. The changed property of the tunnel junction enables monitoring of the oxidation and reduction of the exposed gas via the change in intensity of the light emission from the tunnel junction. This process can be further explored in relation to neuromorphic computing by emulating the function of a synapse [Fig. 5(b)] [76]. The artificial synapse is modeled as an MPM structure using the reactively changed polymer monolayer between the metal metamaterial and metal electrode. The characteristic of the polymer layer can be changed depending on the hot-electron flux via quantum electron tunneling, and is measured by the changes in resistance electrically as well as light emission intensity optically. The researchers have further demonstrated multilevel nonvolatile memory characteristics similar to a memristor by storing the information of the environment in the reactive polymer layers. Consequently, this can potentially be used for memory, switches, and optoelectronic devices.

Inelastic quantum tunneling can be used as a form of spectroscopy for fingerprinting the chemical vibrational mode from the molecules in the tunnel junction [85,86]. Fereiro *et al.* fingerprinted metallo-proteins based on current-voltage (I - V) electrical data, conductance, and second-derivative data [Fig. 5(c)] [87]. The peak from the second derivative of the I - V curve (d^2I/dV^2), which is induced from the inelastic electron tunneling, is well matched to the energy of each vibrational mode in the molecules.

The quantum-tunnel junction can provide a solution to the interface of photons and electrons for an on-chip wireless optical interconnect, as shown in Fig. 5(d) [88]. It can convert the optical signal directly into an electrical signal by exhibiting a change in conductance depending on the power of the optical signal, and thus enabling a rectenna to rectify the current at the gap antenna structure. It can be further applied to ultrafast information transfer by the antenna and sub-nanometer gap rectenna structure. This ultrafast modulation of tunneling and its light emission pattern has been experimentally monitored by Parzefall *et al.* [Fig. 5(e)] [61]. Using time-correlated single-photon counting, they modulated and recorded the device operation ranging from 10 MHz to 1 GHz. This indicates that these quantum-electron-tunneling devices can potentially provide on-chip ultrafast, ultrasmall, and ultracompact light sources.

Figure 5. Applications based on inelastic-quantum-tunneling-based devices. (a) Nanoscale chemical reaction sensor through quantum-tunneling devices [84]. ©2018, Nature Publishing Group. (b) Multilevel nonvolatile memory through optical and electrical readout for neuromorphic computing. Reprinted with permission [76]. ©2020, American Chemical Society. (c) Molecule fingerprinting through inelastic electron spectroscopy. Image reproduced with permission [87] under the terms of the Creative Commons Attribution 4.0 International license. (d) Optical transceiver for on-chip wireless broadcasting of information. Image reproduced with permission [88] under the terms of the Creative Commons Attribution 4.0 International license. (e) Ultrafast information processing and computing through quantum electron devices [61]. ©2015, Nature Publishing Group.

The development of quantum-tunneling-based light sources must overcome some hurdles, such as enhancing the efficiency, providing wide bandwidth tunability from visible to infrared range, ensuring stability of light emission without blinking, and extending the emission time [89–91]. When these problems are solved, it will be possible to achieve highly improved speed communications, data processing, and ultrasensitive sensing technology. By exploring the wide range of material selection and coupling of active materials such as quantum dots, fluorescent materials, diamond, or 2D materials in quantum devices, we can expand the potential of ultrasmall quantum devices in large areas for application in emerging quantum information technology [91–97].

Acknowledgements

This research was supported by the National Research Foundation (NRF) of Korea under the “Korean-Swiss Science and Technology Program” (2019K1A3A1A1406720011), the Ministry of Trade, Industry and Energy (MOTIE, project number 10080625), the Korea Semiconductor Research Consortium (KSRC) program for the development of future semiconductor devices, Samsung Electronics, and the framework of Warm Heart Center program managed by the Institute of Convergence Science, Yonsei University.

References

- [1] T. Stefano, *Nature* 459, 312 (2009).
- [2] S. Kitsinelis, *Light Sources: Technologies and Applications* (CRC Press, Boca Raton, FL, 2016).
- [3] C. E. Moore, A. Von Smolinski, and B. Jaselskis, *IEEE Spectrum* 33, 38 (1996).
- [4] S. S. McPherson, *War of the Currents: Thomas Edison vs Nikola Tesla* (Twenty-First Century Books, Minneapolis, MN, 2012).
- [5] G. Wise, *IEEE Spectrum* 19, 66 (1982).
- [6] G. Zissis and S. Kitsinelis, *J. Phys. D: Appl. Phys.* 42, 173001 (2009).
- [7] B. Roberts, *Joseph Wilson Swan Electric Lamp Pioneer* <http://www.hevac-heritage.org> (accessed: Dec. 22, 2020).
- [8] C. J. Humphreys, *MRS Bull.* 33, 459 (2008).
- [9] Y. Wang, J. M. Alonso, and X. Ruan, *IEEE Trans. Ind. Electron.* 64, 5754 (2017).
- [10] J. Cho, J. H. Park, J. K. Kim, and E. F. Schubert, *Laser Photonics Rev.* 11, 1600147 (2017).
- [11] S. Pimpitkar, J. S. Speck, S. P. DenBaars, and S. Nakamura, *Nat. Photonics* 3, 180 (2009).
- [12] Y. Huang, E. L. Hsiang, M. Y. Deng, and S. T. Wu, *Light: Sci. Appl.* 9, 1 (2020).
- [13] M. Dantus and V. V. Lozovoy, *Chem. Rev.* 104, 1813 (2004).
- [14] G. Popescu, *Principles of Biophotonics*, Volume 2: Light emission, detection, and statistics (Institute of Physics Publishing, Bristol, UK, 2019).
- [15] F. Schütt *et al.*, *Nat. Commun.* 11, 1 (2020).
- [16] Y. Inouye and S. Kawata, *Opt. Lett.* 19, 159 (1994).
- [17] E. J. Sánchez, L. Novotny, and X. S. Xie, *Phys. Rev. Lett.* 82, 4014 (1999).
- [18] Y. Nakayama, P. J. Pauzauskie, A. Radenovic, R. M. Onorato, R. J. Saykally, J. Liphardt, and P. Yang, *Nature* 447, 1098 (2007).
- [19] W. Gao, C. F. Doiron, X. Li, J. Kono, and G. V. Naik, *ACS Photonics* 6, 1602 (2019).
- [20] C. F. Doiron and G. V. Naik, *Adv. Mater.* 31, 1904154 (2019).
- [21] J. Klein *et al.*, *Nat. Commun.* 10, 1 (2019).
- [22] K. Barthelmi *et al.*, *Appl. Phys. Lett.* 117, 070501 (2020).
- [23] S. G. Lukishova, A. W. Schmid, A. J. McNamara, R. W. Boyd, and C. R. Stroud, *IEEE J. Sel. Top. Quantum Electron.* 9, 1512 (2003).
- [24] D. Englund, B. Shields, K. Rivoire, F. Hatami, J. Vučković, H. Park, and M. D. Lukin, *Nano Lett.* 10, 3922 (2010).
- [25] P. P. Schrinner, J. Olthaus, D. E. Reiter, and C. Schuck, *Nano Lett.* 20, 8170 (2020).
- [26] X. Brokmann, G. Messin, P. Desbiolles, E. Giacobino, M. Dahan,

- and J. Hermier, *New J. Phys.* 6, 99 (2004).
- [27] Z. Shotan *et al.*, *ACS Photonics* 3, 2490 (2016).
- [28] J. T. Hugall, A. Singh, and N. F. van Hulst, *ACS Photonics* 5, 43 (2018).
- [29] F. Peyskens, C. Chakraborty, M. Muneeb, D. Van Thourhout, and D. Englund, *Nat. Commun.* 10, 1 (2019).
- [30] A. F. Koenderink, *ACS Photonics* 4, 710 (2017).
- [31] J. Lambe and S. McCarthy, *Phys. Rev. Lett.* 37, 923 (1976).
- [32] S. McCarthy and J. Lambe, *Appl. Phys. Lett.* 30, 427 (1977).
- [33] S. McCarthy and J. Lambe, *Appl. Phys. Lett.* 33, 858 (1978).
- [34] S. McCarthy and J. Lambe, *Appl. Phys. Lett.* 37, 554 (1980).
- [35] P. Törmä and W. L. Barnes, *Rep. Prog. Phys.* 78, 013901 (2014).
- [36] A. Karabchevsky, A. Katiyi, A. S. Ang, and A. Hazan, *Nanophotonics* 9, 3733 (2020).
- [37] R. Kirchain and L. Kimerling, *Nat. Photonics* 1, 303 (2007).
- [38] T. Zhong *et al.*, *Science* 357, 1392 (2017).
- [39] W. Du, T. Wang, H. S. Chu, and C. A. Nijhuis, *Nat. Photonics* 11, 623 (2017).
- [40] S. Zhang, C. L. Wong, S. Zeng, R. Bi, K. Tai, K. Dholakia, and M. Olivo, *Nanophotonics* 1 (2020).
- [41] J. Lee, J. Park, J. Y. Lee, and J. S. Yeo, *Adv. Sci.* 2, 1500121 (2015).
- [42] P. N. Prasad, *Nanophotonics* (John Wiley & Sons, Hoboken, 2004).
- [43] N. Kinsey, M. Ferrera, V. Shalaev, and A. Boltasseva, *J. Opt. Soc. Am. B* 32, 121 (2015).
- [44] J. Conde, J. Rosa, J. C. Lima, and P. V. Baptista, *Int. J. Photoenergy* 2012, 619530 (2012).
- [45] M. Grifoni and P. Hänggi, *Phys. Rep.* 304, 229 (1998).
- [46] R. Banerjee and B. R. Majhi, *J. High Energy Phys.* 6, 95 (2008).
- [47] J. Valentine, *Nat. Nanotechnol.* 13, 96 (2018).
- [48] T. Wang and C. A. Nijhuis, *Appl. Mater. Today* 3, 73 (2016).
- [49] B. Laks and D. Mills, *Phys. Rev. B* 20, 4962 (1979).
- [50] V. A. Shkoldin *et al.*, *J. Phys. Chem. C* 123, 8813 (2019).
- [51] B. Schuler *et al.*, *Sci. Adv.* 6, eabb5988 (2020).
- [52] N. Krane, C. Lotze, J. M. Läger, G. I. Reecht, and K. J. Franke, *Nano Lett.* 16, 5163 (2016).
- [53] L. Zhang *et al.*, *Nat. Commun.* 8, 1 (2017).
- [54] C. Chen, C. Bobisch, and W. Ho, *Science* 325, 981 (2009).
- [55] W. Dickson, G. A. Wurtz, P. Evans, D. O'Connor, R. Atkinson, R. Pollard, and A. V. Zayats, *Phys. Rev. B* 76, 115411 (2007).
- [56] F. Monticone and A. Alu, *Chin. Phys. B* 23(4), 047809 (2014).
- [57] Y. Yang, Q. Li, and M. Qiu, *Sci. Rep.* 6, 1 (2016).
- [58] J. Cambiasso, G. Grinblat, Y. Li, A. Rakovich, E. Cortés, and S. A. Maier, *Nano Lett.* 17, 1219 (2017).
- [59] A. F. Koenderink, A. Alu, and A. Polman, *Science* 348, 516 (2015).
- [60] J. Alda, J. M. Rico-García, J. M. López-Alonso, and G. Boreman, *Nanotechnology* 16, S230 (2005).
- [61] M. Parzefall, P. Bharadwaj, A. Jain, T. Taniguchi, K. Watanabe, and L. Novotny, *Nat. Nanotechnol.* 10, 1058 (2015).
- [62] M. Parzefall, Á. Szabó, T. Taniguchi, K. Watanabe, M. Luisier, and L. Novotny, *Nat. Commun.* 10, 292 (2019).
- [63] S. Namgung, D. A. Mohr, D. Yoo, P. Bharadwaj, S. J. Koester, and S. H. Oh, *ACS Nano* 12, 2780 (2018).
- [64] N. Cazier, M. Buret, A. V. Uskov, L. Markey, J. Arocas, G. Colas Des Francs, and A. Bouhelier, *Opt. Express* 24, 3873 (2016).
- [65] C. Zhang, J. P. Hugonin, A. L. Coutrot, C. Sauvan, F. Marquier, and J. J. Greffet, *Nat. Commun.* 10, 1 (2019).
- [66] X. He, J. Tang, H. Hu, J. Shi, Z. Guan, S. Zhang, and H. Xu, *ACS Nano* 13, 14041 (2019).
- [67] R. Kullock, M. Ochs, P. Grimm, M. Emmerling, and B. Hecht, *Nat. Commun.* 11, 1 (2020).
- [68] J. Kern, R. Kullock, J. Prangasma, M. Emmerling, M. Kamp, and B. Hecht, *Nat. Photonics* 9, 582 (2015).
- [69] S. P. Gurunaranan, N. Verellen, Vy. S. Zharinov, F. J. Shirley, V. V. Moshchalkov, M. Heyns, J. Van de Vondel, I. P. Radu, and P. Van Dorpe, *Nano Lett.* 17, 7433 (2017).
- [70] M. Buret *et al.*, *Nano Lett.* 15, 5811 (2015).
- [71] A. Stolz, J. Berthelot, M. M. Mennemanteuil, G. Colas des Francs, L. Markey, V. Meunier, and Al. Bouhelier, *Nano Lett.* 14, 2330 (2014).
- [72] A. Dasgupta *et al.*, *Beilstein J. Nanotechnol.* 9, 1964 (2018).
- [73] H. Qian, S. W. Hsu, K. Gurunatha, C. T. Riley, J. Zhao, D. Lu, A. R. Tao, and Z. Liu, *Nat. Photonics* 12, 485 (2018).
- [74] W. Du *et al.*, *Nat. Photonics* 10, 274 (2016).
- [75] W. Du, Y. Han, H. Hu, H. S. Chu, H. V. Annadata, T. Wang, N. Tomczak, and C. A. Nijhuis, *Nano Lett.* 19, 4634 (2019).
- [76] P. Wang, M. E. Nasir, A. V. Krasavin, W. Dickson, and A. V. Zayats, *Nano Lett.* 20, 1536 (2020).
- [77] S. Zhou, K. Chen, X. Guo, M. T. Cole, Y. Wu, Z. Li, S. Zhang, C. Li, and Q. Dai, *Nanoscale* 12, 1495 (2020).
- [78] B. Huang, Si. Gao, Y. Liu, J. Wang, Z. Liu, Y. Guo, and W. Lu, *Opt. Lett.* 44, 2330 (2019).
- [79] H. Göktas, F. S. Gökhan, and V. J. Sorger, *ACS Photonics* 5, 4928 (2018).
- [80] P. Bharadwaj, M. Parzefall, A. Jain, and L. Novotny, *Micro-and Nanotechnology Sensors, Systems, and Applications IX*, Proceedings of SPIE, edited by T. George, A. K. Dutta, and M. S. Islam (Bellingham, WA, June 19, 2017), Vol. 10194, p. 101940H.
- [81] J. J. Baumberg, J. Aizpurua, M. H. Mikkelsen, and D. R. Smith, *Nat. Mater.* 18, 668 (2019).
- [82] W. Zhu, R. Esteban, A. G. Borisov, J. J. Baumberg, P. Nordlander, H. J. Lezec, J. Aizpurua, and K. B. Crozier, *Nat. Commun.* 7, 1 (2016).
- [83] N. Kongsuwan, A. Demetriadou, M. Horton, R. Chikkaraddy, J. J. Baumberg, and O. Hess, *ACS Photonics* 7, 463 (2020).
- [84] P. Wang, A. V. Krasavin, M. E. Nasir, W. Dickson, and A. V. Zayats, *Nat. Nanotechnol.* 13, 159 (2018).
- [85] M. A. Reed, *Mater. Today* 11, 46 (2008).
- [86] R. G. Keil, T. P. Graham, and K. P. Roenker, *Appl. Spectrosc.* 30, 1 (1976).
- [87] J. A. Fereiro, X. Yu, I. Pecht, M. Sheves, J. C. Cuevas, and D. Cahen, *Proceedings of the National Academy of Sciences*, edited by N. V. Raikhel (Washington, DC, May 15, 2018), Vol. 115(20), pp. E4577-E4583.
- [88] A. Dasgupta, M. M. Mennemanteuil, M. Buret, N. Cazier, G. Colas-des-Francs, and A. Bouhelier, *Nat. Commun.* 9, 1 (2018).
- [89] M. Parzefall and L. Novotny, *Rep. Prog. Physics* 82, 112401 (2019).
- [90] M. Parzefall and L. Novotny, *ACS Photonics* 5, 4195 (2018).
- [91] M. Parzefall, P. Bharadwaj, and L. Novotny, in *Quantum Plasmonics*, edited by S. Bozhevolnyi, L. Martin-Moreno, and F. Garcia-Vidal (Springer, New York, 2017), pp. 211-236.
- [92] S. I. Bozhevolnyi and J. B. Khurgin, *Nat. Photonics* 11, 398 (2017).
- [93] S. I. Bozhevolnyi and N. A. Mortensen, *Nanophotonics* 6, 1185 (2017).
- [94] J. Lee, J. Y. Lee, J. S. Yeo, *ACS Appl. Mater. Interfaces* 11, 36177, (2019).
- [95] Z. Jacob and V. M. Shalaev, *Science* 334, 463 (2011).
- [96] M. S. Tame, K. McEnery, Ş. Özdemir, J. Lee, S. A. Maier, and M. Kim, *Nat. Phys.* 9, 329 (2013).
- [97] J. M. Fitzgerald, P. Narang, R. V. Craster, S. A. Maier, and V. Giannini, *Proceedings of the IEEE*, edited by G. Setti (New York, December 2016), Vol. 104(12), pp. 2307-2322.