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Excitation of Surface Plasmons by Inelastic Electron Tunneling

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Surface plasmons are usually excited by diffraction-limited optical methods with the use of bulky optical components, which greatly limits the miniaturization and chip-scale high-density integration of plasmonic devices. By integrating a plasmonic nanostructure with a tunnel junction, plasmonic modes in the nanostructure can be directly excited by low-energy tunneling electrons with the advantages including an ultra-small footprint and an ultra-fast speed. In this mini-review, recent progress in the electric excitation of localized and propagating surface plasmons by inelastic electron tunneling is overviewed.

Keywords: surface plasmons, tunnel junctions, inelastic tunneling, optical antennas, light emission, waveguiding

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INTRODUCTION

Surface plasmons are highly confined electromagnetic modes coherently coupled to collective oscillations of free carriers at metallic (or doped semiconductor) interfaces. They exist in the form of surface plasmon polaritons (SPPs) propagating at an interface between a conductor and a dielectric or as localized surface plasmons (LSPs) supported by confined conductive nanostructures [1, 2]. Their ability to localize electromagnetic fields at a subwavelength scale and produce greatly enhanced local fields for strong light–matter interaction offers the opportunity to combine the advantages of nanoelectronics (small size) and dielectric nanophotonics (high speed), opening an avenue for merging electronics and photonics at the nanoscale [3]. In the past 20 years, a great progress has been made in the area of plasmonics, which have stimulated a variety of applications, such as nano waveguides [4–6], plasmonic lasers [7–9], ultrafast electro-optical [10–12] and all-optical [13, 14] modulation, photodetection [15, 16], bio-chemical sensing [17, 18], and enhancement of non linear optics [19, 20].

Usually, surface plasmons are excited by diffraction-limited optical methods with the use of bulky optical components (e.g., prisms, grating, objectives, etc.) [1], which greatly limits the miniaturization and chip-scale high-density integration of plasmonic devices. At the same time, there are some alternatives. In his seminal work, Ritchie proposed that fast electrons can be used for the excitation of surface plasmons in metal [21]. Later, both the excitation of SPPs [22, 23] and LSPs [24] have been experimentally demonstrated with high-energy (∼30 keV) electron beams with an advantage of highly precise and localized excitation (with a spatial resolution down to several nanometers). However, the requirements of a high electric voltage and a vacuum environment make it impossible for practical applications. Low-energy electrical excitation of SPPs has been demonstrated, e.g., by coupling plasmonic waveguides with electrically driven nano light sources [25, 26], but a highly compact and faster approach not related to the carrier lifetime would be highly desirable. In this mini-review, we focus on the recent breakthroughs in the low-energy direct excitation of surface plasmons based on an inelastic electron tunneling (IET) effect in tunnel junctions.

ORIGIN AND INITIAL STUDIES OF OPTICAL EMISSION BY IET

In 1976, Lambe and McCarthy [27] observed a broadband light emission from macroscopic planar metal-insulator-metal (MIM) tunnel junctions with an external quantum efficiency (EQE, i.e., electron-to-photon conversion efficiency) around 10^{-5}. This phenomenon can be explained in terms of IET-based excitation of surface plasmon modes subsequently coupled to photons on rough planar MIM tunnel junctions. When an electric bias is applied across an MIM structure with a nanometer-scale insulator thickness, electrons can quantum-mechanically tunnel through the insulating barrier. During the tunneling process (**Figure 1A**), most electrons tunnel elastically without energy loss, appearing as high-energy (in respect to the Fermi level) electrons on the other side of the junctions, so called "hot electrons." Some small fraction of electrons, however, tunnel inelastically, giving part of their energy to the excitation of plasmonic modes in the junction, which can then couple to extended propagating SPP modes or to free-space photons. The resulting emission spectral profile $I(\omega) \propto I_{tc}(V,\omega)\,\rho_{LDOS}\eta_{rad}$ is defined by the electromagnetic intensity spectrum of the tunneling current $I_{tc}(V,\omega) \propto \left(1 - \frac{\hbar\omega}{eV}\right)$ (which can be found from the calculation of the quantum transition matrix elements [30, 31] or from a Fourier transform of the tunneling current shot noise [32]), local density of optical states (LDOS) ρ_{LDOS} in the junction region and the radiative efficiency of the tunneling system in terms of generation of output photonic and/or plasmonic modes η_{rad}. In other words, the intrinsic electromagnetic spectrum from the tunneling current, $I_{tc}(V,\omega)$, is highly dependent on the applied bias V with a high-frequency cutoff ω_{co} (defined by the quantum relation for the maximal energy conversion $\hbar\omega_{co} = eV$) and a monotonic increase toward lower frequencies, and it is shaped into the final emission spectrum by the optical (frequently resonant) properties of the tunneling structure, defined by ρ_{LDOS} and η_{rad}. Later in the 1980's, IET-induced light emission was also reported from plasmonic tunnel junctions formed between a scanning tunneling microscope (STM) tip and a metallic substrate [33–35]. By analyzing the leakage radiation of a tunnel junction formed between an STM tip and a thin gold film in both image and Fourier planes, Wang et al. found that up to 99.5% of the detected photons come from leakage radiation of SPPs propagating on the gold film with the remaining photon emission attributed to the radiative decay of a localized plasmonic mode excited between the STM tip and the gold film [36], explicitly demonstrating the possibility of highly efficient coupling of inelastic tunneling to propagating plasmonic modes. Furthermore, despite its low EQE, this technique provides a high-spatial-resolution method for the study of LSPRs in metallic nanostructures [37–39]. At the same time, in combination with the atomic-scale spatial resolution of an STM, this approach has been developed into a useful optical spectroscopic method for single-molecule characterizations [40].

IET-GENERATED LIGHT EMISSION FROM OPTICAL ANTENNAS

Together with the success of IET-induced plasmon excitation and light emission in the STM research community comes its main challenge for the application in practical devices related to its low efficiencies, including internal quantum efficiency (IQE, inelastic tunneling efficiency, which is defined by the ratio of the generated plasmonic quanta and the number of overall tunneling events) and EQE (for photon-related applications). Overcoming this has attracted continuous research interest in the past four decades because of the ultra-small footprint of tunnel junctions, which allows for high-density integration, and the ultra-fast speed of the IET process (at a scale of few femtoseconds [41]), which offers the potential for ultra-fast direct modulation of the excitation. These efforts are further motivated by a theoretical prediction that the IQE can be of the order of 10%, known from the early days of the research [42]. From the theoretical point of view, the IQE of a plasmonic tunnel junction is defined by the electronic densities of states in both electrodes (as well as any other electronic states inside the junction area) and the LDOS in the tunnel junction, while the EQE is defined by a product of the IQE and the radiation efficiency of the tunnel junction [43–45]. By engineering the LDOS and radiation efficiency, a significant increase in both IQE and EQE has been recently demonstrated [28, 46–48]. For example, in 2015, Kern et al. demonstrated the first electrically driven optical antenna by integrating a tunnel junction into it [28]. In this experiment, the tunnel junction was fabricated by placing a gold nanoparticle into a gap formed between two arms of a linear dipole antenna as shown in the left panel of **Figure 1B**. The emission spectrum from the electrically driven optical antenna is then defined by the applied bias and the nanoantenna plasmonic resonance, which can be tuned by changing the geometry of the nanoantenna (right panel of **Figure 1B**). Taking advantage of the high LDOS and radiation efficiency of the resonant antenna design, the EQE was increased to $\sim 10^{-4}$, which is about two orders of magnitude higher than that for a non-resonant design. Later in the same year, Parzefall et al. achieved resonantly enhanced light emission by structuring an array of slot antennas on the bottom electrode of a vertical MIM tunnel junction formed by two gold electrodes and an insulating h-BN crystal [46]. Compared with an unstructured MIM tunnel junction, the EQE of the nanostructured junctions is increased by two orders of magnitude from $\sim 4 \times 10^{-7}$ to $\sim 2.5 \times 10^{-5}$ at a bias of 2.5 V due to the enhanced radiation efficiency $\sim 4 \times 10^{-3}$ provided by the slot antennas. The authors further demonstrated direct temporal modulation of light emission from the MIM tunnel junctions at frequencies up to 1 GHz. In 2018, implementing a tunnel junction produced by two chemically synthesized silver nanocubes assembled into an edge-to-edge configuration with the stabilizing polymer simultaneously working as the insulating barrier, Qian et al. obtained a record-high EQE of up to 2% at near-infrared frequencies [47]. Such excellent efficiency values are underlined by a very high LDOS in the tunneling junction (a factor of 3.1 × 10^5 higher than in vacuum) provided by an atomic-level quality of the gap between the silver single crystals and prominent 24.6% radiative efficiency of the implemented edge-to-edge nanoantenna design. In 2019, by cross-placing an Ag nanowire and an Au nanostripe, He et al. demonstrated the excitation of cavity plasmons with highly tuneable multiple emission peaks and narrow (tens of nanometers) line widths [49]. By using a dielectrophoresis trapping method, they further demonstrated efficient fabrication of nanoparticle-based electrically driven optical antennas with a measured EQE of ∼2.5 × 10^{-4} [50]. Looking into the tunneling system from a conceptual point of view, Uskov et al. theoretically showed that the close-to-unity IQE can be achieved by introducing a quantum well structure in the tunneling gap with the energy level in the well aligned in a way that the inelastic tunneling happens in a resonant manner while the elastic counterpart does not [44]. However, as the authors noticed, this is done on the expense of the overall value of the tunneling probability, which dramatically decreases.

Although the IQE and EQE in plasmonic tunnel junctions have been significantly improved by engineering the LDOS and radiation efficiency, the overall generated plasmonic or photonic power is still quite low (pW level or smaller), which causes a difficulty in the signal detection and greatly limits their applications. This, however, is mainly due to the intrinsically low tunneling current in single nanoscale tunnel junctions. A promising way to solve this problem is increasing the number and density of the optical antenna-coupled tunnel junctions. For example, by constructing a macroscopic and high-density plasmonic tunnel junction array at the top of a plasmonic metamaterial produced by vertically oriented gold nanorods (**Figure 1C**, nanorod areal density is around 1 × 10^{10} cm^{-2}), Wang et al. realized IET-driven light emission visible by the naked eye (**Figure 1D**) [29, 51]. The spectrum of the emission in this case is shaped by the metamaterial plasmonic modes, which can be tuned throughout the visible and near-infrared

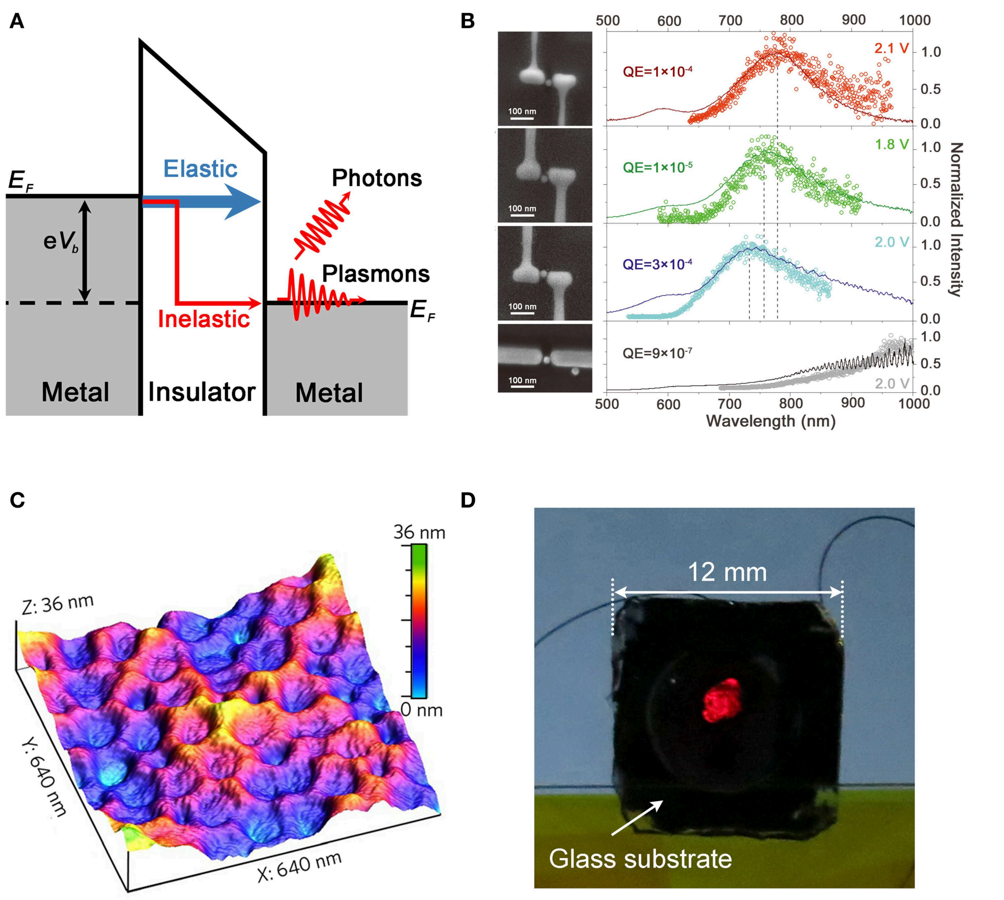

FIGURE 1 | (A) Schematic diagram of generation of surface plasmons and photons through IET. **(B)** Electroluminescence (data points) and scattering spectra (solid lines) for antennas with various geometries and a micrometer-long non-resonant wire, together with the corresponding SEM images. **(C)** Atomic force microscopy image of a gold nanorod array with the dips marking the positions of Au few nanometers below the surface of an Al_2O_3 matrix. **(D)** Photograph of an Au nanorod array electrically driven at a bias of 2.5 V. **(B)** is reprinted with permission from Ref. [28]. Copyright © 2015, Springer Nature. **(C,D)** are reprinted with permission from Ref. [29]. Copyright © 2018, Springer Nature.

ranges by tuning the metamaterial modes via the nanostructure geometric parameters [51]. The measured emission power was around 100 nW, which makes the signal detection trivial for applications such as optical sensing. Based on this, Wang et al. further demonstrated an ultra-compact electrically driven optical sensor by exploiting hot electrons generated via elastic tunneling (usually ignored, as it decays by the generation of heat) for the activation of chemical reactions in the junctions and IET-generated photons for the monitoring of this process [29].

IET-BASED EXCITATION OF WAVEGUIDED MODES

Apart from coupling to free-space light emission and 2D plasmonic modes, IET can also be coupled to waveguided plasmonic or photonic modes, which is highly desired for on-chip applications, as they have a crucial advantage as information carriers in comparison with traditional electronic signals in terms of a higher bandwidth and lower loss. In 2011, Bharadwaj et al. reported an electrical excitation of propagating SPPs in a Au nanowire (**Figure 2A**) [52]. A plasmonic mode excited with an STM tip at the left end of the nanowire by IET, was subsequently coupled to SPPs propagating along the nanowire and then converted to free-space photons at the right end. However, the excitation of propagating SPPs with the use of STM is difficult for practical applications where on-chip integration is highly desired. In this respect, a promising design was realized by integrating an electromigrated tunnel junction on the top of a dielectric-loaded surface plasmon waveguide (DLSPPW) (**Figure 2B**) [53] or by crossing a gold plasmonic waveguide and an thin aluminum strip covered with a nanoscale oxide layer [56, 57]. For the latter case, an SPP excitation efficiency exceeding 1% was reported [56], which was further explained by surface roughness–induced momentum matching between the MIM modes in the junction and the output SPP modes present in the system [57]. In 2019, Zhang et al. further demonstrated enhanced excitation of SPPs along an aluminum–air interface by fabricating an array of linear gold antennas on the top of an oxidized aluminum surface [58]. The emitted SPP power was increased to ∼10 pW, and the emission spectrum/polarization was controlled by the design of the antenna arrays. According to a recent calculation by Parzefall et al. [59], the IET-induced excitation efficiency of SPPs in extended conventional plasmonic waveguides is limited by a low coupling efficiency between the extremely confined MIM modes excited in the tunnel junction and the propagating waveguided SPPs due to the dramatic mismatch between their propagation constants. An additional problem might be caused by the low modal overlap. This shows that more attention is required in the future to improve the coupling efficiency, e.g., via structural design of the coupling area.

It is worth mentioning that in addition to plasmon excitation based on metallic tunnel junctions plasmon and light emission can also be generated with metal-insulator-semiconductor (MIS) tunnel junctions [60]. The advantage of the MIS tunnel junctions is that they can be directly integrated into, e.g., a silicon photonic waveguides for on-chip applications [61, 62]. Particularly, with the coupling efficiency of the hybrid junction optical mode to the silicon waveguide of ∼ 75%, Doderer et al. experimentally generated a waveguided optical power of 6.8 pW [61].

DIRECTIVITY CONTROL OF THE PLASMONIC EXCITATION AND LIGHT EMISSION

The ability to control the flow of optical energy is of great importance in nanophotonic applications. The directional control of SPPs and light emission excited by IET has been demonstrated in a variety of systems [54, 55, 63–65]. For

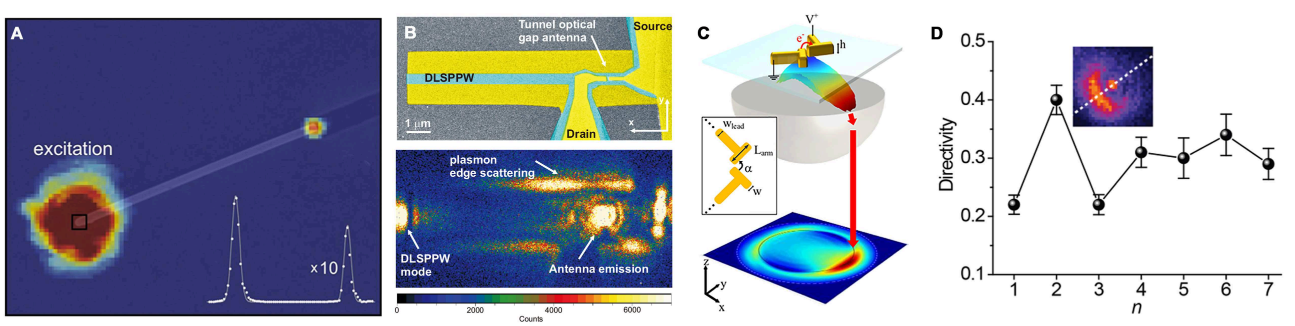

FIGURE 2 | (A) A map of optical emission intensity from an Au nanowire excited by an STM tip. **(B)** False color SEM image of a tunnel junction on the top of a DLSPPW waveguide, together with an optical intensity map, showing generation of the propagating plasmonic mode at the junction region and its outcoupling to the free-space radiation at the other end of the waveguide. **(C)** Tunneling-driven highly directional emission from a V-shaped nanoantenna. **(D)** The dependence of the directivity of the SPP excitation on the structural characteristics of self-assembled S(CH2)nBPh polymer molecules filling the tunneling gap defined by the chain parameter *n*. The inset shows an experimental measure defocused patterns corresponding to a tunnel junction with $n = 2$. **(A)** is reprinted with permission from Ref. [52]. © 2011 American Physical Society. **(B)** is reprinted with permission from Ref. [53]. Copyright © 2016 Optical Society of America. **(C)** is reprinted with permission from Ref. [54]. Copyright © 2017 American Chemical Society. **(D)** is reprinted with permission from Ref. [55]. Copyright © 2019 American Chemical Society.

example, Dong et al. demonstrated a directional control of SPP-assisted light emission from a gold stripe cavity with a directivity of extinction ratio around 2.6 : 1, which was realized by varying the distance between an STM probe and the edge of the cavity to attain a constructive or destructive interference with the generated and reflected SPP waves [63]. Taking advantage from an excellent directivity provided by optical antennas, Gurunarayanan et al. achieved a directivity of light emission of $\sim\ 5\ dB$ by aligning two nanorod antennas edge-to-edge at an angle of 90° (**Figure 2C**) [54]. Such a strong directivity is provided by an interplay between the dipolar radiation pattern of the tunnel junction emission and the quadrupole-like resonance of the rod antennas. Recently, Kullock et al. obtained a directivity of light emission as high as 9.1 dB in an optical Yagi-Uda antenna with a tunneling feed [64]. The directivity control can also be achieved by placing molecules in the junction region, particularly utilizing their chemical composition and/or orientation [55, 65]. For example, implementing tunneling through a self-assembled monolayer of polymer molecules (**Figure 2D**, inset), Du et al. experimentally achieved directional launching of SPPs by adjusting the tilt angle of a self-assembled monolayer of $S(CH_2)_nBPh$ (BPh = biphenyl) molecules in respect to the electrode surface, which was realized by controlling the length of the alkyl chain n [55]. The highest directivity (defined as $\frac{I_L-I_R}{I_L+I_R}$, where I_L and I_R are the maximum intensities of left and right lobes of the emission pattern, respectively) of 0.4 was obtained for n = 2 (**Figure 2D**, main graph), corresponding to a left/right intensity ratio of ∼2.3.

CONCLUSION AND OUTLOOK

In this review, we have overviewed the recent developments in the IET-assisted excitation of surface plasmons, including both LSPRs and SPPs, which open an opportunity for the miniaturization and chip-scale integration of plasmonic devices. However, for practical applications, there are still many things to be done and questions to be answered. For example, how to improve the overall output power from single tunnel junctions? How to optimize the coupling efficiency between an MIM mode excited by IET and SPPs in an extended waveguide for on-chip integration? How to achieve narrow-band excitation of surface plasmons? Finally, the question of long-term stability of tunnel junctions is a key concern for applications. Despite these challenges, as an ultra-fast and compact approach that can bridge electronics and plasmonics directly at the nanoscale, IET-based plasmonic excitation will continue to attract research interest and find applications in areas, such as optical interconnections and sensing.

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PW and LL organized and wrote the article. All the authors participated in discussion and revision.

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