Electrical Driven Light Emitting From a Tunneling Junction With Negative Resistance Effect

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This work was supported in part by the National Basic Research Program of China under Grant 2016YFA0202000 and Grant 2015CB932400, and in part by the National Natural Science Foundation of China under Grant 11427808 and Grant 51372045.

ABSTRACT We investigate the electrical and optical performance of surface-plasmon-mediated light emission in electrical-driven metal-insulator-metal (MIM) tunnel junctions fabricated from a gold (Au) film on the top and a heavily doped silicon chip on the bottom. A silicon dioxide layer is used as the tunneling barrier. The experimental results show that the device's performance strongly depends on the morphology of the Au film. A negative resistance effect was observed with high Au film roughness, from which higher efficient light emission was observed compared to that of our device exhibiting less roughness. Such MIM tunneling junctions are compatible with common metal-oxide semiconductor technology and thus open up a route toward the development of novel integrated optoelectronic and plasmonic devices.

INDEX TERMS Metal-insulator-metal, surface plasmon, inelastic tunneling, light emitting, negative resistance effect field.

I. INTRODUCTION

Polarization mode (plasmons) at the interface of a metal and a dielectric is a good mediator of energy transfer between electrons and photons [1]. Unlike photons, energetic electrons can easily couple to plasmons due to their higher momentum [2]. Energetic electrons can be generated by tunnelling one electrode to its counterpart in a biased metal-insulator-metal junction. Most electrons tunnel elastically, maintaining their energy during the tunneling process, while others tunneling inelastically [3], [4]. The latter is a process in which a tunneling electron excites a surface plasmon in the junction and its energy loss is equal to a quantum of plasmon. This phenomenon was first observed by Lambe and McCarthy in 1976 [5].

Accompanying the inelastic tunneling processes is light emission, which occurs as a result of the scatter or decay of plasmons [5]. Compared to other light emitting processes, one major appeal of inelastic electron tunneling light emission is its extremely high speed [4]. Because it does not rely on intermediate excitations such as electron-hole-pairs, the operating speed of the devices is fundamentally only limited by the tunneling time of electrons through the junction. This speed is on the order of tens of femtoseconds [6], therefore, this mode of light emission has many applications in ultrafast optoelectronics and communication.

The prevailing mechanism for the inelastic electron tunneling excitation of light involves a three-step process [2], [7]: (I) electrons tunnel inelastically from one electrode to another in the junction; (II) surface plasmon polaritons (propagating or localized) are excited by coupling from the electron energy loss $\hbar\omega$, accompanied by the generation of "hot electrons" [8]; and (III) the coupled surface plasmon may subsequently decay into free-space radiation when it encounters structural asymmetry, such as edges or bulges [9]–[11]. Consequently, the light emitting performance is strongly dependent on the morphology of the top electrode [4].

In this manuscript, we investigate the electrical and optical performance of a metal-insulator-insulator tunneling junction with several different top electrode structures (i.e., roughness). At a certain optimized roughness, higher efficient light emitting is obtained compared with other devices with lower roughness. Accompanying the light emission process is a negative resistance (NR) [12] phenomenon observed in devices with a high roughness top electrode. This phenomenon is not observed in devices featuring electrodes with low roughness. The electrical driven light emitting diode

with this NR effect is of great interest for future optoelectronic devices. The device structure is shown in Fig. 1a, where highly doped Si acts as the bottom electron emitting cathode, followed by a SiO_2 tunneling barrier, and Au plasmonic nanostructures are situated on the top as the light emitting mediator. The three physical steps involved in the light emitting process are shown in Fig. 1b.

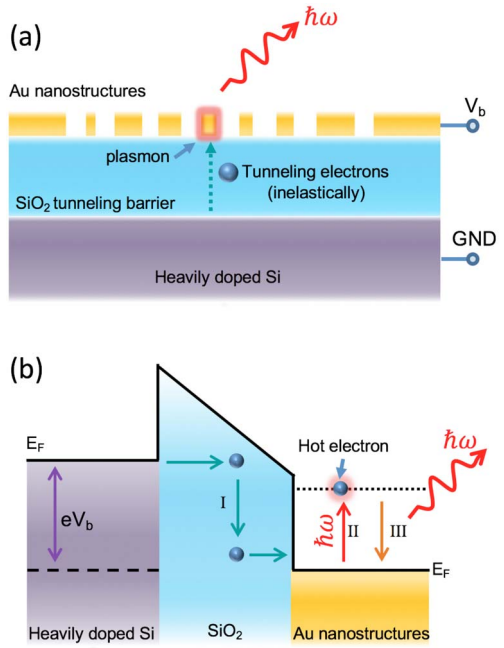

FIGURE 1. (a) Schematic diagram of the electrical driven light emitting tunneling junction and (b) illustration of light emitting mechanism.

II. EXPERIMENT AND SIMULATION DETAILS

In the fabrication process (sample 1), the 10 nm SiO_2 layer is formed by annealing a Si chip (doped with Sb to a concentration of 2.5×10^{19} cm^{-3}) in air for two hours at 400°C. The challenge is the formation of an optimized nanoscale plasmonic nanostructure on the top electrode for efficient light emission. One key reason is that, while preserving high surface roughness, the electrical conductivity should also be maintained, which requires well-connected nanostructured islands, rather than isolated islands. It is found that the Au plasmonic structure can be formed by annealing the thin Au film in air [13]. The morphology can be tuned by changing the annealing temperature. This is possible due to the higher surface energy of the metallic layers than that of the oxide layers. The diffusion rate of metal atoms increases at high temperature, causing the thin film to assume an island structure. To allow for the best conditions, the sample was annealed in air at an optimized temperature of 180°C for two hours, resulting in an Au nanomesh structure with nanoscale features (Fig. 2a, b). We fabricated another device possessing a nanohole plasmonic structure (sample 2), employing an electrical annealing method. In this device, one applies a bias voltage between the bottom and top electrodes to generate a high tunneling current. The non-uniform tunneling current then generates randomly-distributed "hot spots" on the Au film. Nanoscale hole structures form as a result of the local heating effect at the tunneling sites (Fig. 2c, d). Compared with the before-mentioned Au nanomesh film formed via the annealing method, fewer edges and bulges are observed in the latter process requiring a bias voltage.

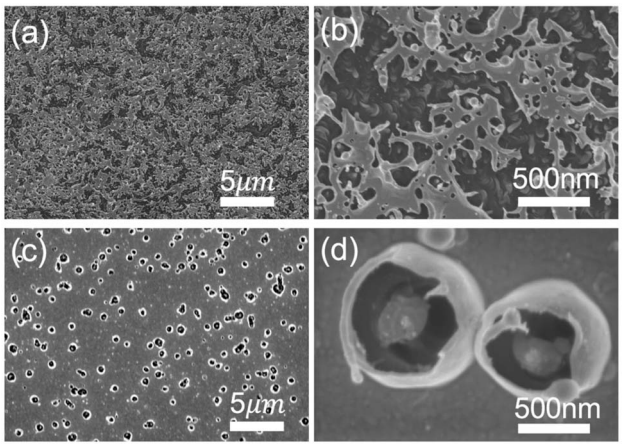

FIGURE 2. Scanning electron microscopy images for the top electrode of two MIM devices: sample 1 in (a) low magnification, (b) high magnification, (c) sample 2 in low magnification, and (d) high magnification.

The active area of both devices were 3 mm × 3 mm. The measurements were carried out in air. Electron tunneling was controlled by applying a bias voltage using a source meter. The light emission spectrum was recorded with a spectrometer.

III. RESULTS AND DISCUSSION

The tunneling current dependence on the bias voltage (I-V curves) of sample 1 is shown in Fig. 3a. As shown in the inset, at lower bias voltages (<2.9 V), the tunneling current increases linearly, corresponding with direct tunneling (DT). With increasing voltage (>2.9 V), the current reveals a nonlinear increase, corresponding with a triangular barrier field emission (FE) process. The corresponding Fowler–Nordheim (FN) plot (Fig. 3b, low voltage region) is fully compatible with a MIM tunnel junction that shows direct tunneling for field emissions at higher voltages with a transition voltage of ~2.9 V. As the bias voltage increases (>3.3 V), we observe a deviation of the tunneling current from the FE regime (Fig. 3b), followed by a rapid decrease from ~40 mA to ~9 mA when the bias voltage increases from 13.3 V to 23.4 V. This effect is known as negative resistance (NR) [14]. After that, the current increases again as the bias voltage rises (> 23.4 V). In the bias region of >3.3 V, the tunneling electrons gain enough energy to effectively excite the plasmons. The excited plasmons elevate the occupied energy level at the top Au nanostructure due to their "hot electrons" [15] (Fig. 1b), thereby reducing the electron tunneling probability from the bottom electrode.

This limits the tunneling current flow, causing a deviation in the emission current from the FE regime (here defined as plasmon limited FE, PLFE) and NR effect. With an increase in the bias voltage (> 23.4 V), the highest occupied energy level on top of the Au nanostructure can be lowered enough to enable electron tunneling again. Thus, the current flow increases with an increase in the bias voltage (> 23.4 V).

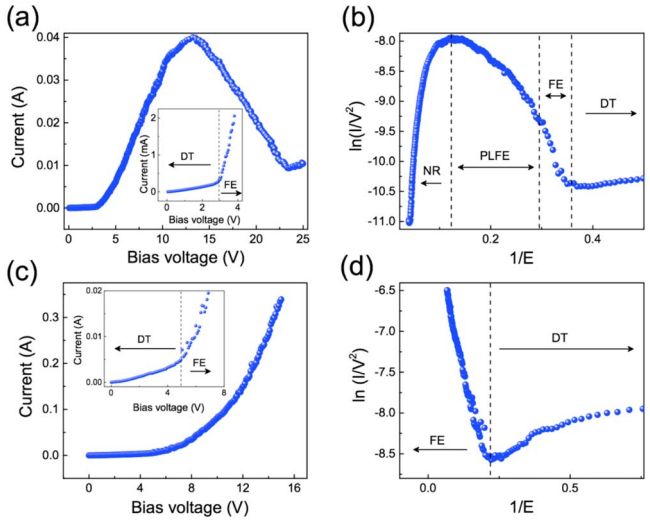

FIGURE 3. **(a) I-V curve of sample 1 and (b) its FN plot, (c) I-V curve of sample 2 and its FN plot.**

The I-V curve for sample 2 is shown in Fig. 2c. No NR effect is observed in the measurement bias range. Unlike sample 1, a large portion of the excited plasmons in sample 2 propagated in the gap region, followed by a rapid decay in distance (<30 nm) [4]. Conversely, only a small portion near the hole structure was able to couple to free space radiation [4]. Due to Ohmic loss, the majority of the system's energy transfers to thermal rather than photon energies. The "hot electron" will also be generated on the Au film; however, due to the limited area (i.e., "hot spot"), it has little effect on the total tunneling electron population. Consequently, the tunneling current of sample 2 is more than one order higher than that of sample 1 at the same bias voltage (Fig. 3a, c). The comparsion supports our explaination of the NR effect.

The light emission images are shown in the inset of Fig. 4, which displays high uniformity for the whole device. The high magnification image reveals many bright dots, which suggests that the light emission is mainly located at the edges of the nanostructures. The measured light emission spectra of sample 1 under various bias voltages are shown in Fig. 4. According to both theory and experimental observations [3], inelastic electron tunneling results in broadband light emission with a high-energy photon cutoff given by

$$\hbar\omega_{\max} = |eV_{bias}| \qquad (1)$$

where $\hbar\omega_{\max}$ is the photon energy, e is the electron charge and V_{bias} is the applied bias voltage in the junction. According to equation (1), higher resonances should only be excitable if the applied bias exceeds the energy of the resonances mode. As shown in Fig. 4, this behavior is indeed observed. The peak of the spectrum shifts to blue with an increase in the bias voltage. The experimental data show good agreement with equation (1). In addition, in the NR region, the light intensity scales linearly with the bias voltage, and inversely with the tunneling current. This is generally good for high efficiency light emission.

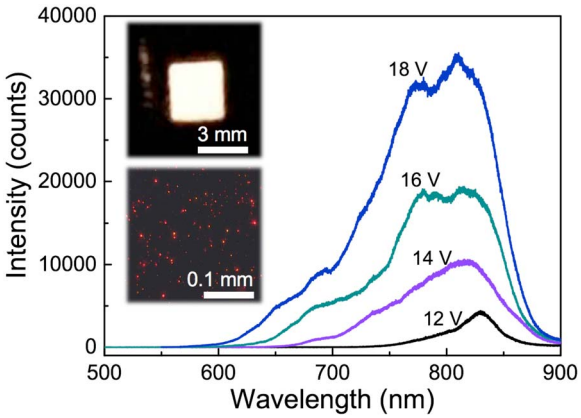

FIGURE 4. **Light emitting spectrum of sample 1 under different bias voltages. The insets show the light emitting images in low (top) and high (bottom) magnifications.**

IV. CONCLUSION

In conclusion, large uniform MIM tunneling junctions based on heavily-doped Si, SiO_2, and Au nanostructures were fabricated and characterized electrically and optically. The electrical driven light emitting performance is shown to be strongly dependent on the roughness of the top electrode. Along with efficient light emitting in the optimized devices, the NR effect is observed, which has potential applications in next generation opto-electronic devices, including communications and computing. Additionally, the MIM structure here is also capable of converting far-field radiation into localized energy, which is of particular interest in photovoltaics, photodetection and optical sensors.

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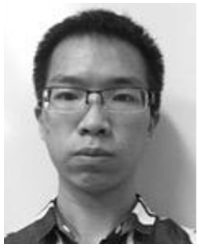

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