Efficient Surface Plasmon Polariton Excitation and Control over Outcoupling Mechanisms in Metal–Insulator–Metal Tunneling Junctions

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1. Introduction

Sub-diffraction limit optoelectronic circuitry has been demonstrated by integrating surface plasmon polaritons (SPPs)\(^1\)\(^–\)\(^3\)—coherent, collective oscillations of free electrons propagating at metal–dielectric interfaces—into a myriad of applications in fields such as nano-optics,\(^4\)\(^–\)\(^8\) sensing,\(^9\)\(^–\)\(^11\) sub-wavelength imaging,\(^2\)\(^,\)\(^3\) or nano-optoelectronics.\(^12\)\(^,\)\(^13\) SPPs are typically excited with bulky light sources that are too large for nano-circuitry integration; hence, for many applications it would be desirable to have access to footprint-compatible electrically driven SPP sources. Metal–insulator–metal tunneling junctions (MIM-TJs) are known to electrically excite SPPs and photons via inelastic tunneling under an applied bias\(^14\)\(^–\)\(^17\) (Figure 1a) and are therefore interesting candidates.\(^18\)\(^–\)\(^20\) Until recently, prohibitively low electron-to-photon conversion efficiency (10\(^4\)–10\(^7\) electrons result in a single photon\(^14\)\(^–\)\(^16\),\(^21\)\(^–\)\(^23\)) due to the large momentum mismatch between...
the initially excited MIM-TJ cavity mode (MIM-SPP) and the radiating mode, coupled with an inelastic tunneling event probability of 10%[24] has prevented practical applications of MIM-TJs. Recently, we have shown that the electron-to-SPP conversion efficiency can be experimentally as high as 1% in a planar Al-AlO$_x$-Cr-Au MIM-TJ[17], and Qian et al.[25] demonstrated 2% electron-to-photon emission efficiency in a resonant Ag–polymer–Ag nanocrystal tunnel junction, both within one order of magnitude agreement with theoretical inelastic tunneling models[24,26], demonstrating the potential of MIM-TJs in nano-scale optoelectronics.

Despite this recent experimental evidence of efficient SPP excitation in MIM-TJs, theoretical treatises of these systems continue to argue that these efficiencies cannot be due to outcoupling of the MIM-SPP mode, and an alternative interpretation is needed.[27,28] In this work we demonstrate that, by modeling MIM–TJ interface surface roughness and electrode thickness with a simple ad hoc roughness model, electron-to-SPP efficiencies can be within an order of magnitude of experimental measurement. Furthermore, we demonstrate experimental control over outcoupling of the MIM-SPP mode and the outcoupling pathways to radiative (photons) and non-radiative (SPPs) modes. Our work shows experimental efficiencies can be further increased by exploiting surface roughness at the MIM–TJ interfaces[22,29,30] which, in principle, can be extended by coupling other types of structures to junctions, such as antennas or gratings.

The exact contributions from different SPP and photon excitation pathways from the MIM-SPP mode remain experimentally unclear in MIM-TJs.[26] MIM-TJ excitation of radiating and non-radiating modes can be split into two steps. Initially, the energy quanta donated from inelastically tunneling electron couples to the local density of optical states (LDOS—Figure 1a). The decay pathway of this energy is defined by the LDOS of the system, which preferentially couples to the cavity mode (MIM-SPP) in Al-AlO$_x$-Cr-Au MIM-TJs.[31–33] Once the MIM-SPP is excited, it then outcouples to both the radiative and non-radiative mode, as well as dissipating into the electrodes.[34] The MIM-SPP mode has a wavevector of an order of magnitude larger than the single-interface SPP modes at the metal/dielectric interfaces (bound-SPP modes) at visible/near-IR frequencies. Parzefall et al.[27] recently theoretically investigated the outcoupling efficiency of the MIM-SPP mode of an ideally smooth MIM-TJ without surface roughness or topography, considering only outcoupling via pathway 2 (the only possible pathway in smooth systems; see the following sections), and reported very low outcoupling efficiencies on the order of 10$^{-8}$. Therefore, in order to increase outcoupling efficiencies, momentum-matching conditions at the metal–dielectric interfaces are required to outcouple the mode to both free space photons and to bound-SPPs.[26,35] Randomly roughened electrodes,[17,22,29,30,36–42] gratings,[21,43,44] and prisms[45] have been integrated into MIM-TJs to preferentially outcouple the MIM-SPP mode or the bound-SPP modes to photons by reducing this momentum mismatch.[17,26,43–46]
Root-mean-squared (RMS) surface roughness $\sigma_m$ enhances both mode overlap and momentum matching between the MIM-SPP mode and the daughter modes. The mode overlap of the MIM-SPP depends on the decay length $\delta_{\text{MIM}}$ - the distance that the MIM-SPP extends into either electrode – which is typically tens of nanometers. Mode overlap is only efficient when the thickness of the electrode $t_m$ is on the order of the $\delta_{\text{MIM}}$. and even then requires a roughened topography to momentum match. Surface roughness enables momentum-matching of the MIM-SPP mode and the daughter mode by modifying the local permittivity $\varepsilon_m$ (as a function of frequency $\omega$) by $\varepsilon_m(\omega, \sigma_m) = \varepsilon_m(\omega) + \delta\varepsilon_m(\omega, \sigma_m)$ where $\delta\varepsilon_m$ is the local perturbation to $\varepsilon_m$ which modifies the local electromagnetic environment.

Dawson et al. calculated that, for Al-AlO$_x$-Au MIM-TJs, the ideal $\sigma_m$ for the most efficient out-coupling of the MIM-SPP mode to photons is $\sigma_m = 5$ nm. Moreover, recent studies into atomic-scale topographical defects in metallic nanoparticle clusters that can act as plasmonic “picocavities” that can cause highly localized perturbations to the electromagnetic topography.

Here we show through experiments supported by theory that the MIM-SPP mode readily outcouples via three possible outcoupling pathways of the MIM-SPP mode: i) scattering of the MIM-SPP to photons at the different dielectric–metal interfaces (Figure 1c,d, pathway 1), ii) coupling of the MIM-SPP to bound-SPP modes by spatial mode-overlap and roughness-induced momentum matching (Figure 1c,d, pathway 2), and iii) direct coupling of the MIM-SPP mode to photons as well as the bound-SPP modes at the edge of the MIM-TJ and the adjacent waveguides (Figure 1c,d, pathway 3). By studying light emission from Al-AlO$_x$-Cr-Au MIM-TJs integrated into Al and Au waveguides (Figure 1b) and varying the Al and Au thickness (Figure 1c,d) we demonstrate that the MIM-SPP mode couples to the bound-SPP modes (Figure 1c,d), where the coupling efficiency is a function of the effective electrode thickness. Our simulations show that the majority of the bound-SPP excitation originates from surface roughness-induced momentum matching of the MIM-SPP. The homogeneous light emission from the MIM-TJ area originates from the scattering of the MIM-SPP at the interfaces between the electrodes and the insulator layer. Our results uncover the origin of light emission from MIM-TJs and give new insights into the contributions and control of the different MIM-SPP mode outcoupling pathways, which serves to guide the rational design of future plasmonic-electronic devices based on tunneling junctions.

2. Results and Discussion

2.1. Electrical Characterization

The devices were fabricated on borosilicate coverslips following previously reported methods (see Section S1, Supporting Information). Figure 2a shows the AFM image from which we determined a value of $\sigma_m = 5 \pm 2$ nm for Al and $\sigma_m = 1.0 \pm 0.2$ nm for Au (both measured over an area of $1 \times 1 \mu m^2$). Figure 2b shows a line scan recorded from the Al surface revealing peak-to-valley roughness $\sigma_{\text{pv}}$ of $25 \pm 2$ nm. We fabricated four different MIM-TJs with varying electrode thickness $t_{\text{m}}$. A rough surface topography reduces the effective thickness of the electrodes by $t_{\text{eff}} = t_{\text{m}} - \sigma_m$ as indicated in Figure 2b, which further increases the coupling efficiency of the MIM-SPP mode via pathway 2. Table 1 gives the measured electrode thickness and effective thickness for each electrode from the AFM line scans of the four samples. All deposition methods and rates were kept the same in order to keep surface roughness similar for all devices. The rms surface roughness of the glass coverslip ($\sigma$ of $0.17 \pm 0.01$ nm measured over an area of $1 \times 1 \mu m^2$, see Section S1, Supporting Information) is two orders of magnitude smaller than the roughness of the electrodes and thus negligible.

To ensure that the Al-AlO$_x$-Cr-Au MIM-TJs were dominated by direct tunneling, the IV-characteristics prior to the optical measurements (Figure 2c) were recorded. All IV-curves show typical tunneling behavior with a positive, parabolic differential conductance (dI/dV — Figure 2d). Furthermore, we have previously shown that these MIM-TJs have currents that scale with $I = dV/dt$ conductance (d$V$/d$t$— Figure 2d). We fabricated four different MIM-TJs with varying electrode thickness $t_{\text{m}}$, $t_{\text{Al}}$, and $t_{\text{Au}}$ so that no modes at the Al-Au interface could be excited (see the following sections). Each image intensity was scaled such to show any light emission from both the MIM-TJ area and scattered bound-SPPs at the end of the waveguides. From these three images, we make the following four observations: 1) light emission is visible from both the MIM-TJ area and the end of the Al and Au plasmonic waveguides, 2) light emission from the MIM-TJ area decreases with increasing Al thickness, 3) light emission from the end of the waveguides decreases with increasing Al thickness, and 4) the light emission intensity is not homogeneous from the MIM-TJ.

These observations can be explained as follows. The light emitted from the MIM-TJ area is the result of scattering of the MIM-SPP mode (pathway 1). The light emitted from the end of the waveguides is caused by scattering of the bound-SPP
modes, which were excited by the MIM-SPP mode (pathway 2). $t_{\text{eff}}$ for each MIM-TJ is reduced by $\sigma_b$, making all electrodes effectively thinner, enhancing mode overlap and increasing coupling efficiency. For the MIM-TJ shown in Figure 3a, $t_{\text{Al},\text{eff}} = 15 \text{ nm}$ and therefore the MIM-SPP mode can readily outcouple via pathways 1 and 2 because $t_{\text{Al},\text{eff}} < \delta_{\text{MIM}}$ (see Section 2.4) resulting in light emission from both the MIM-TJ area and the ends of the waveguides. In contrast, as $t_{\text{Al},\text{eff}}$ becomes increasingly thicker than $\delta_{\text{MIM}}$, the mode overlap between the MIM-SPP and SPP$_{\text{Al-glass}}$ decreases rapidly, and the MIM-SPP mode cannot directly outcouple to photons, diminishing the outcoupling via pathways 1 and 2. Figure 3c shows that the MIM-TJ area light emission when $t_{\text{Al},\text{eff}} = 85 \text{ nm}$ is reduced by a factor of $\approx 20$ compared to $t_{\text{Al},\text{eff}} = 15 \text{ nm}$, indicating that outcoupling via channel 1 is effectively quenched. This observation is in line with that reported by Kroó et al.$^{[48]}$ who reported complete quenching of the MIM-SPP scattered emission at $t_{\text{Ag}} > 80 \text{ nm}$ in Al-ALO$_x$-Ag MIM-TJs. Light emission from the ends of both waveguides is also very weak, indicating that pathway 2 is also effectively quenched. Light emission from this MIM-TJ is now dominated by scattering of the MIM-SPP from the edges of the MIM-TJ. Here the MIM-SPP mode mainly outcouples to the bound-SPPs by pathway 3.

Further indicating the role of surface roughness, the light intensity in Figure 3a–c varies spatially across the MIM-TJ area and edges. This is because $\sigma_p \approx \delta_{\text{MIM}}$ in each MIM-TJ, which implies that in areas where the Al is thickest (Figure 2b), the photons need to travel an extra factor of $\delta_{\text{MIM}}$ to reach the glass interface, leading to strong attenuation (see Figure S1, Supporting Information, for Al skin depth) relative to the photons close to the surface (valleys; Figure 2b). Hence, surface roughness can also explain the inhomogeneous light emission from the MIM-TJ area.

Our results are not in agreement with the theoretical work of Parzefall et al.$^{[14,27]}$ who did not take the role of surface roughness into consideration and argued that light emission should mainly come from the MIM-TJ edges (pathway 3) in smooth MIM-TJs. They claim that due to the small MIM-SPP propagation length $\Lambda_{\text{MIM-SPP}} \approx 20 \text{ nm}$ for optical frequencies, and small value of $\delta_{\text{MIM}} \approx 20 \text{ nm}$, the MIM-SPP mode can only outcouple to other modes at the MIM-TJ edge (pathway 3), as MIM-SPP modes excited further than $\Lambda_{\text{MIM-SPP}}$ would be attenuated. This leads them to conclude that MIM-SPP out-coupling efficiency should be very low (on the order of $10^{-8}$)$^{[14,27]}$. These works are accurate for smooth MIM-TJs with no roughness but neglect to consider the role of surface roughness and coupling via pathway 2. Their argument cannot explain considerable literature that reports light emission from the MIM-TJ area.$^{[16,21,22,26,30,37,38,43-46,48,49,60-62]}$ and cannot explain the observations shown in Figure 3.

### Table 1. Metal thicknesses and effective thicknesses for each sample.

| Sample | $t_{\text{Al}}$ [nm] | $t_{\text{Au}}$ [nm] | $t_{\text{Al},\text{eff}}$ [nm] | $t_{\text{Au},\text{eff}}$ [nm] |
|--------|----------------------|----------------------|--------------------------|---------------------------|
| 1      | 20                   | 155                  | 14 ± 2                   | 149 ± 2                   |
| 2      | 65                   | 155                  | 59 ± 2                   | 149 ± 2                   |
| 3      | 90                   | 155                  | 84 ± 2                   | 149 ± 2                   |
| 4      | 40                   | 55                   | 34 ± 2                   | 49 ± 2                    |

Figure 2. a) AFM image of a typical MIM-TJ with $\sigma_m = 5\pm 2 \text{ nm}$. b) Measured line scan of the MIM-TJ as indicated by the dashed line indicated in (a) showing the $t_{\text{eff}}$ and $t_m$. c) IV-characteristics of the MIM-TJs of different thicknesses. d) Differential conductance $dI/dV$ showing parabolic bias dependence.
Figure 3. Wide-field inverted optical microscopy images of the Al-AlOₓ-Cr-Au MIM-TJs with different electrodes thickness. EMCCD images of the devices measured at $V_{\text{bias}} = -1.45$ V with 155 nm thick Au top electrode and a) 20, b) 65, and c) 90 nm thick Al bottom electrode; d) device with 55 nm thick Au top and 40 nm thick bottom Al electrodes. e–h) Back focal plane (BFP) images of the corresponding MIM-TJs; $V_{\text{bias}} = -1.5$ V for (e)–(f), -1.55 V for (g), and -1.6 V for (h), and corresponding normalized simulated BFP images in i–l). All EMCCD images have had their intensity individually scaled to best show MIM-TJ light emission and scattering at the end of the waveguides. All BFP images have had their respective intensities scaled to give the best contrast. The inner white dashed line in the BFP images refers to the critical angle of the total internal reflection at the air–glass interface ($k_{\text{SPP}}/k_0 = 1.0$); the outer white dashed line indicates the maximum angle of the collected light limited by the NA of the objective ($k_{\text{SPP}}/k_0 = 1.49$).
2.3. Optical Characterization: BFP Imaging

To explore the outcoupling pathways described above in more detail, we recorded back focal plane (BFP) images to characterize the light emission from the entire device in k-space as a ratio of SPP momenta $k_{\text{SPP}}$ to the momentum of light in a vacuum $k_0$, which splits up the collected intensity from light scattered directly from the MIM-TJ ($k/k_0 < 1$) and light from SPP scattering ($k/k_0 > 1$). Figure 3e–g shows corresponding BFP images recorded from the MIM-TJs as a function of $t_{\text{Al,eff}}$. Figure 3e shows that light is not scattered discretely in all angles from the MIM-TJ area, while the opposite is true for Figure 3g where only light emitted under discrete angles is visible. This observation supports the hypothesis that the MIM-SPP mode outcoupling is dominated by pathways 1 and 2 for MIM-TJs with $t_{\text{Al,eff}} = \delta_{\text{MIM}}$ but not when $t_{\text{Al,eff}} > \delta_{\text{MIM}}$, where only outcoupling by pathway 3 is possible. Furthermore, the intensity background within $k/k_0 < 1$ implies that only roughness-scattered photons can be detected at these angles; no background would be expected for an MIM-TJ with no roughness, and this background is only visible in Figure 3e,h (see Figure S4, Supporting Information).

The BFP images make it possible to identify all bound-SPP modes as indicated in Figure 3e (supported by theory, see the following sections). The bound-SPP modes propagating along the waveguides at the metal–glass interface can be observed via scattering at the end of the Au or Al waveguides (as in Figure 3e–h), but cannot be detected efficiently by leakage radiation due to limitations of the NA of our detector (NA = 1.49 < $k_{\text{SPP}}/k_0$).[17] BFP imaging allows us to identify which bound-SPP modes are excited as a function of the electrode thickness. As mentioned above, with increasing Al thickness the coupling efficiency of the MIM-SPP mode to the SPP$_{\text{Al,glass}}$ mode is expected to decrease due to the $t_{\text{Al,eff}} > \delta_{\text{MIM}}$. Since we measure the optical properties of the MIM-TJs from the back side of the MIM-TJs, the optical contribution of the SPP$_{\text{Al-air}}$ mode excited by pathway 3 also diminishes with increasing Al thickness as the Al becomes increasingly opaque. The SPP$_{\text{Al-air}}$ mode is still visible in Figure 3g, but the MIM-SPP mode is not since $t_{\text{Al,eff}} > \delta_{\text{MIM}}$. The BFP images in Figure 3e–g show the presence of the SPP$_{\text{Al,glass}}$ mode, but the 155 nm thick electrode prevents coupling of the MIM-SPP mode to the SPP$_{\text{Al-air}}$ mode ($t_{\text{Al,eff}} > \delta_{\text{MIM}}$).

To see coupling of the MIM-SPP mode to the SPP$_{\text{Au-air}}$ mode, an MIM-TJ with $t_{\text{Al,eff}} = 35$ nm and $t_{\text{Au,eff}} = 50$ nm was fabricated (Figure 3d). The Al electrode thickness was chosen so that the MIM-SPP mode would be still visible but not too intense to saturate the detector, obscuring all bound-SPP modes. As anticipated, we observed lower light intensity from the MIM-TJ area due to scattering of the MIM-SPP mode relative $t_{\text{Al,eff}} = 15$ nm Al electrode (Figure 3a) but higher than for the MIM-TJ $t_{\text{Al,eff}} = 60$ nm (Figure 3b). However, now the SPP$_{\text{Au-air}}$ mode is clearly visible in the BFP image (Figure 3h), confirming that reducing the top electrode thickness allows excitation of the SPP$_{\text{Au-air}}$ mode by pathway 2.

2.4. Simulations

We performed numerical calculations (finite-different time-domain—FDTD—Lumerical software[65]) of the angular light distribution from an incident MIM-SPP excitation as a function of $t_{\text{Al,eff}}$ and $t_{\text{Au,eff}}$ (Figure 3i–l; for full simulation details see Section S4, Supporting Information), which show good agreement with the experimentally collected BFP (Figure 3e–h). This agreement in BFP features for different bound-SPP modes in both theory and experiment confirms that the MIM-SPP mode is initially excited and then couples to all available bound-SPP modes, and that this coupling efficiency depends on $t_{\text{Al,eff}}$ and $t_{\text{Au,eff}}$ as described in detail above and shown in Figure 1.

The above observations lead us to conclude that the bound-SPP modes outcouple predominantly at the ends of the Au and Al waveguides, while the majority of the light emission in the MIM-TJ area comes from the direct outcoupling to photons of the MIM-SPP mode, scattering off of roughness in the MIM-TJ. Furthermore, by ranging from $t_{\text{Al,eff}}$, $t_{\text{Au,eff}} = \delta_{\text{MIM}}$ to $t_{\text{Al,eff}}, t_{\text{Au,eff}} >> \delta_{\text{MIM}}$, we can control the outcoupling of the MIM-SPP mode to all possible bound-SPP modes.

2.5. MIM-SPP Outcoupling Pathways

To demonstrate how the outcoupling of the MIM-SPP mode can be affected by surface roughness and changes in $t_{\text{eff}}$, we simulated the mode profile for MIM-TJ structures with changes in the topography of the electrodes that represent various types of defects (see Section S4.4, Supporting Information for simulation details). Modeling roughness for highly confined plasmonic systems is notoriously complex due to nanoscale mode profiles.[66,64] Here, we only aim to show how a variety of local features in the electrode topography can affect the MIM-SPP outcoupling pathways and improve the outcoupling efficiencies to the bound-SPP modes. Figure 4 shows simulation results for MIM-TJs with different roughness conditions and $t_{\text{Al,eff}}$, where all thicknesses are indicated and the electric field intensity is normalized uniformly for all panels. Figure 4a shows the simulation results for a planar MIM-TJ without roughness, and Figures 4b–d show the results for MIM-TJs with surface defects with heights on the order of $\sigma_p$. Figure 4b shows the results for a $\sigma_p = 25$ nm, Figure 4c shows an MIM-TJ where $\sigma_p$ is close to $t_{\text{Al,eff}}$, and Figure 4d shows an MIM-TJ where $\sigma_p$ is $\approx 3.5x$ smaller than $t_{\text{Al,eff}}$. Figure 4b shows the enhancement induced by the roughness of the outcoupling of the MIM-SPP through the Al electrode by pathway 2 relative to the smooth electrodes in Figure 4a, where outcoupling is dominated at the MIM-TJ edges via pathway 3. The presence of the roughened interface significantly increases outcoupling to all bound-SPPs, supporting our observations in Figure 3i. Figure 4c,d shows the effect of thickness on pathway 2, where $t_{\text{Al,eff}} >> \delta_{\text{MIM}}$ significantly reduces the mode overlap of the MIM-SPP and SPP$_{\text{Al,glass}}$. This effect supports the observation in Figure 3c where only light from the MIM-TJ edges can be observed. These mode profiles indicate the enhancement effect of both $\sigma_p$ and $t_{\text{Al,eff}}$ on outcoupling via pathway 2, as well as demonstrate that pathway 3 remains relatively unaffected by either parameter.

2.6. MIM-SPP Outcoupling Efficiencies

The outcoupling efficiencies of the MIM-SPP mode to the bound-SPP modes can be estimated by placing dipoles at
different spatial positions inside MIM-TJs of different thickness and roughness. Figure 5 shows the coupling efficiency of the MIM-SPP to SPP\textsubscript{Au-glass} for an MIM-TJ with $\sigma_m = 0$ nm (dashed lines) and $\sigma_m = 5$ nm (solid lines) for each corresponding MIM-TJ thickness combination from Figure 3. Figure 5a,c shows the coupling efficiency between the two modes via pathway 2 for different dipole positions (indicated in Figure 5a) inside the insulator of the MIM-TJ from the edge of the MIM-TJ ($x = 0 \mu m$) to the center ($x = 2.5 \mu m$). Figure 5b,d shows the coupling efficiency for dipoles placed within 100 nm of the MIM-TJ edge (indicated in Figure 5b), where edge effects begin to dominate due to contributions from pathways 2 and 3, as the distance from the edge approaches $x \approx \Lambda\text{MIM-SPP}$ which we calculate to be $\Lambda\text{MIM-SPP} \approx 50$ nm for $\lambda = 900$ nm (see Figure S4, Supporting Information). Since $\Lambda\text{MIM-SPP}$ is a function of wavelength, this also implies that the contribution from pathway 3 will depend significantly on wavelength, and by extension a total outcoupling efficiency will also be wavelength dependent. We have chosen $\lambda = 900$ nm as this is the spectral peak of the emitted light from our MIM-TJs (as reported previously\cite{17}).

From Figure 5c,d we can see that for each MIM-TJ, introduced surface roughness in the simulation increases the coupling efficiency of pathway 2 by $\approx 70\times$. This is due to the momentum mismatch in both pathways being reduced by scattering from the roughened surfaces. Furthermore, the thicker Al electrodes give weaker coupling efficiencies for both pathways, due to the reduction in spatial overlap of the modes. For pathway 2, as the dipole source approaches the edge of the waveguide ($x = 0 \mu m$) the distance the excited SPP\textsubscript{Al-glass} has to propagate is reduced, reducing propagation loss and increasing coupling efficiency. With these simulations we compute a total MIM-SPP out-coupling efficiency by all pathways of 0.62% for the thinnest Al electrode MIM-TJ by integrating all contributions from pathways 2 and 3 across the MIM-TJ, and then normalizing by MIM-TJ area. Taking into consideration the proposed inelastic tunneling excitation efficiency of the MIM-SPP mode of 10%\cite{24} we estimate a total electron-to-SPP efficiency of 0.062%. This efficiency is 4–6 orders of magnitude larger than recent publications\cite{14,27} and within an order of magnitude of our experimental findings\cite{17}. We note that the improvements in the coupling efficiencies in the calculations only serve as estimates given our ad hoc roughness profile, currently we are conducting more comprehensive calculations with rigorously derived, statistically modeled surface topographies.

3. Conclusions

In this work, we have used imaging techniques coupled with FDTD simulation to understand SPP mode excitation...
mechanisms in MIM-TJs, as well as developed basic structural understanding of MIM-TJs to expose the influence of topographical surface roughness on MIM-SPP outcoupling and discrete control over electrode thickness to "turn on and off" excitation of various bound-SPPs as well as light emission. We demonstrate that in MIM-TJs, inelastically tunneling electrons excite the MIM-SPP mode, which then outcouples to electromagnetic modes by three pathways: i) outcoupling to photons by interface roughness-induced momentum matching, ii) outcoupling to bound-SPPs due to surface roughness both momentum matching and reducing the effective electrode thickness to increase spatial mode overlap, and iii) outcoupling directly to bound-SPPs at the edge of the MIM-TJ. We demonstrate that the efficiencies of pathways 1, 2, and 3 depend on the electrode thickness and that the contributions from each pathway can be controlled; when \( t_{\text{Al,eff}}, t_{\text{Au,eff}} \approx \delta_{\text{MIM}} \) the coupling of the momentum-matched, scattered MIM-SPP mode is relatively efficient (pathways 1 and 2 contribute alongside pathway 3), and when \( t_{\text{Al,eff}}, t_{\text{Au,eff}} > \delta_{\text{MIM}} \) the coupling is small (only pathway 3 contributes).

This MIM-SPP out-coupling can have efficiencies as high as 0.62%, confirming what we have claimed in previous works\[17\] that this process is similarly efficient as other on-chip SPP excitation sources. We note that these estimates are conservative since we only modeled roughness coarsely as the main aim of this paper was to investigate the outcoupling mechanisms of the MIM-SPP mode and the effect of different topographical defects. By changing the bottom and top electrode thickness in MIM-TJs we can control the dominating mode and re-direct outcoupling of the MIM-SPP mode to the bound-SPP mode either at the metal–glass or metal–air interfaces. Contrary to the recent arguments offered by Parzefall et al.,[14,27] we find that the contributions from the different MIM-SPP outcoupling pathways vary depending on electrode thickness and interface roughness. Not only that, but we demonstrate that the majority of the light emanating from a statistically roughened MIM-TJ area is indeed due to MIM-SPP scattering from roughness at the electrode interfaces and outcoupling into photons, resolving a long-standing question in MIM-TJ literature. This research confirms the efficiency of MIM-TJs as on-chip SPP sources. Therefore we believe that MIM-TJs, especially when combined with optical elements such as gratings,\[65\] antennas,\[66\] or waveguides,\[17\] are potentially interesting candidates for applications in sensing and opto-electronics.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.
Author Contributions
K.S.M., T.X.H., and T.J.D. contributed equally to this work. K.S.M. and A.R. fabricated the samples. K.S.M. performed the experiments and analysed the data. T.X.H. and H.-S.C. performed theoretical calculations. V.K. provided theoretical support. T.J.D. assisted with measurements. C.A.N. conceived and designed the experiments. All authors discussed the results and commented on the manuscript.

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inelastic electron tunneling, light emission, roughness, surface plasmon polaritons, tunnel junctions

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