Coherence Between Different Propagating Surface Plasmon Polariton Modes Excited by Quantum Mechanical Tunnel Junctions

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Coherence between different surface plasmon polariton (SPP) modes excited by inelastically tunneling electrons in biased metal–insulator–metal tunnel junctions (MIM-TJs) is demonstrated. By employing a dedicated SPP stripe waveguide with MIM-TJ, an effective double-slit configuration similar to the Young’s experiment is realized for an electrically biased SPP source. The spatial correlation between different SPP modes originates from a single inelastic tunneling event and leads to strong interference in the far-field, observed as alternate bright and dark fringes in the Fourier plane. The measured fringe-spacing inversely follows the stripe waveguide length, with upper limit dictated by the SPP propagation length, confirming the SPP mediated spatial correlation. Finite difference time domain simulations support the experimental findings. Also, the experimental and simulation results unambiguously demonstrate the two-step plasmonic decay process in plasmonic MIM-TJs. The results presented here provide a simple and robust demonstration of the inherent coherence existing between different decay channels (plasmons and photons) of the inelastically tunneling electrons and can be exploited for plasmonic applications with tailored spatial coherence with implications in plasmon amplification and quantum information processing.

1. Introduction

Tailoring the spatial coherence in a source-waveguide interconnect system is an essential step in many state of the art plasmonic applications, for instance, in modulated data transfer for information processing,[1–4] in on-chip platforms for surface plasmon polariton (SPP) mediated interferometric plasmonic imaging[5] and sensing[6] for biological species, and for spatial differentiation and edge detection without optical lenses that perform the Fourier transform,[7,8] to name a few. SPPs are coherent due to the collective nature of free electron oscillations[9] and are bound to the metal–dielectric interfaces which provides unrivalled field confinement at the nanoscale,[10] opening up a plethora of applications ranging from sensing[11] to quantum information processing.[12,13] Plasmonic metal–insulator–metal tunnel junctions (MIM-TJs) offer a unique platform for the electrical excitation of SPPs by low energy (<2.5 eV) tunneling electrons[14–18] because of their nanoscale footprint and potential for device-integration for subwavelength imaging, or nano-optoelectronics.[19,20] In MIM-TJs, the SPP or photon emission originates from a three-step outcoupling process in which, first, the energy quanta (ℏω) given off by inelastically tunneling electrons can couple predominantly to a highly-confined cavity mode (MIM-SPP,[18] Figure 1a), which then outcouples to SPPs and finally to photons.[21] Here we show that this entire three-step process remains coherent as the excitation originates from a single inelastic tunneling event and is important for areas of research where spatially correlated and actively modulated sources are essential, such as, plasmonic interferometry.[22] Plasmonic analogue of quantum optics...
Light emission from planar MIM-TJs was first reported by Lambe and McCarthy in 1976. Since then both photon and SPP emission have been reported from various macroscopic MIM tunneling devices including scanning tunneling microscope (STM), molecular junctions, metallic break junctions, nanowire-metal stripe junctions, and metal-2D material hybrid systems. The past, modulation of the spatial coherence of light by plasmonic near-field has been demonstrated in Young’s interference experiments in metal stripe waveguides supporting SPPs and also in the context of inelastic tunneling, with STM light emission, where the localized plasmonic response of the STM tip plays a crucial role. For large area TJs, the lack of correlation between inelastic tunneling events typically results in spatially incoherent and spectrally broad (lack of temporal coherence) photon or plasmon emission. However, for MIM-TJs with conductance $G < G_0$ (conductance quantum), the tunnel current typically ranges from nA to µA and leads to $10^9$–$10^{12}$ electrons tunneling per second and essentially results in a single MIM-SPP mode excitation during an inelastic tunneling event as the decoherence time of these modes is typically of the order of few fs. It is of fundamental interest to investigate whether this initial coherence, originating from the tunneling event, is preserved as the MIM-SPP outcouples to all available modes (SPPs and photonic modes) leading to a mutual correlation between different SPP modes excited by the MIM-TJ. Even though previous works have clarified the outcoupling mechanisms of the MIM-SPP mode, the correlation between the MIM-SPP and the outcoupling channels (SPPs and photonic modes) for biased MIM-TJs still remains elusive.

Figure 1. a) Schematic of the MIM-TJ. Inset gives the schematic representation of the (i) inelastic tunneling, (ii) MIM-SPP, and (iii) SPP mode. The process ‘$1e^-$’ represents the inelastic electron tunneling related to the source current density $J$ (upward vertical arrow represents the tunneling). Photonic outcoupling channels $S_1$ and $S_2$ represent the scattering of the MIM-SPP and SPP respectively and leads to interference in the far-field. $\Omega$ denotes the correlation and $\Delta k$ represents the momentum mismatch between MIM-SPP and SPP. The length of the Au waveguide is given by $L$. b) Topography of the MIM-TJ and associated waveguides obtained by AFM. Representative device for the $L$ dependence study (width $W$) is fixed at 5 µm and $L$ is varied from 2 to 14 µm. The Au waveguides are 150 nm thick in all cases. The Al stripe has a fixed length (20 µm), width (6 µm), and thickness (70 nm). c) Real plane image of the light emission from a representative device for $L$ = 6.5 µm. The image is collected for $V_b = -1.5$ V. d) Intensity profile along the horizontal dotted line through the middle of the Au waveguide in (c). Two peaks are demarcated that represent $S_1$ (TJ right-end) and $S_2$ (waveguide-end) equivalent to the sources represented in (a). The inset shows the emission spectra from MIM-TJs for varying $W$ from 200 to 1000 nm, with an applied bias of -1.5 V. Spectral plots are shifted vertically for better visibility. In both (c) and (d) the rectangular dotted-line box represents the end of the Au stripe where the intensity is rescaled by a factor of 4.
Here we show that the coherence of the MIM-SPP mode with its outcoupling pathways (single interface SPPs and photons) is preserved and leads to strong interference in the far-field. We experimentally demonstrate, for the first time, that the photon emission from the MIM-SPP mode and the SPP scattering share a common source viz. the MIM-SPP mode and we unambiguously prove the role of the MIM-SPP modes in light emission from plasmonic TJs. SPP waveguides are employed to demonstrate the spatial correlation between two mutually correlated photon sources, originating from MIM-SPP ($S_1$; Figure 1a) and SPP ($S_2$; Figure 1a) scattering and results in alternate bright and dark fringes in the Fourier plane (or back focal plane, BFP) of the detection setup. The fringe-spacing provides the direct evidence for the mutual correlation.\(^{[46]}\)

2. Results and Discussion

For this work, we fabricated Al-AO$_x$-Au MIM-TJs which are well-characterized in literature.\(^{[15,18,37,43]}\) Figure 1a shows a cross-section of the TJ used in this study where the Au and Al stripes constitute the electrodes of the TJ and the AlO$_x$ serves as the tunnel barrier. In the linear energy transfer limit, all energy quanta ($h\omega$) in the range of $0 \leq h\omega < eV_b$ (where $V_b$ is the applied bias) are accessible by the tunnelling electrons (indicated as ‘‘$\ell e$’’ process in the inset of Figure 1a).\(^{[46]}\) From the $\ell e$ process, the source current density, $J$, excites the MIM-SPP mode, which subsequently outcouples (via the momentum transfer $\Delta k$) to SPPs and photons. Since the MIM-SPP originates from the $\ell e$ process, an inherent correlation ($\Omega$)\(^{[43]}\) between MIM-SPP and SPP is preserved. This is demonstrated here by investigating the interference between the two mutually correlated sources, $S_1$ and $S_2$, which are the far-field outcoupling of the MIM-SPP and SPP respectively, by varying the length of the Au stripe waveguide ($L$). The MIM-SPP mode scattering at the junction-edge gives $S_y$, where the tunnel barrier ($\text{AlO}_x$) is exposed to the glass medium, and the bound SPPs (Au-glass) outcouple to $S_z$ at the waveguide-end.\(^{[44,45]}\) We chose a Au waveguide thickness of 150 nm so that it is greater than the skin depth of Au ($\sim 25$ nm)\(^{[46]}\) which restricts the excitation to the bound-SPP modes and minimizes any cross-coupling with the Au-air interface. The Al electrode is partially embedded in the glass, and it extends $\sim 20$ µm on each side of the MIM-TJ to avoid any interference effects associated with the SPPs supported by the Al stripe (see Section S1 in the Supporting Information).

Figure 1b shows a representative atomic force microscopy (AFM) image of the fabricated devices. From the AFM images a root-mean-square (over $1 \times 1$ µm$^2$ area) surface roughness of $8 \pm 2$ nm and $1 \pm 0.2$ nm is obtained for Al and Au metal stripes respectively. We also extracted the thicknesses of the stripes, where the bottom Al electrode is $68 \pm 5$ nm, with $30 \pm 4$ nm above the glass substrate and $38 \pm 1$ nm sunken into the glass, and the thickness of the top Au electrode is $150 \pm 2$ nm. For the charge transport studies, a negative bias was applied to the Al electrode with the Au grounded to avoid the breakdown of the TJs at positive bias, as was previously shown.\(^{[48]}\) We observe an exponential behavior of the $I(V)$ curves and a positive parabolic shape of the differential conductance (see Section S2 in the Supporting Information) which are signatures that the MIM-TJs are dominated by tunneling and do not suffer from pinholes, showing that the characteristics of the TJs investigated in this work agree with previous findings.\(^{[18,19,37]}\)

We recorded the photon emission from the TJs using an inverted optical microscope equipped with a spectrophotograph for collecting the emission spectra (400–1100 nm spectral range) and with an electron multiplying charge-coupled device (EMCCD) camera to record the real and BFP images (see Section S3 in the Supporting Information). The emitted light is collected from the glass-side of the TJs using a 100x oil immersion objective of numerical aperture (NA) = 1.49. Figure 1c shows the real plane image of the light emission for a representative device with $L = 6.5$ µm and Figure 1d shows the intensity profile along the horizontal dotted line in Figure 1c through the middle of the waveguide. The rectangular box represents the end of the Au stripe where the intensity is rescaled by a factor of 4, to enhance the visibility of the scattering at the end of the waveguide as compared to the intense light emission from the junction area. The first peak of the intensity profile corresponds to $S_1$, and the second peak represents $S_2$. The inset in Figure 1d shows the emission spectra from TJs with Au stripe width varied from 200 to 1000 nm, with an applied bias $V_b = -1.5$ V. For all cases, the spectra show a peak around wavelength $\approx 900$ nm with a full width at half maximum of $\approx 140$ nm, independent of the waveguide width.

The far-field intensity ($I_{ph}$) recorded in the BFP can be represented as

$$ I_{ph} = \frac{1}{A} \left( E_{S_1} + E_{S_2} \right)^2 = \sum \left( E_{S_1}^2 + E_{S_2}^2 + E_{S_1} \times E_{S_2} + E_{S_2} \times E_{S_1} \right) \text{ (1)} $$

where $E_{S_1}$ and $E_{S_2}$ represent the electric field ($E_{S_i}$ is the complex conjugate of $E_{S_i}$) corresponding to $S_1$ and $S_2$ respectively. The cross-terms between $E_{S_1}$ and $E_{S_2}$ represent the interference contribution and the summation over wavelength ($\lambda$) takes the broadband nature of the MIM-SPP into account.\(^{[13]}\) The left panels of Figure 2a–d show the experimental BFP images corresponding to MIM-TJs with Au waveguides of $L = 2.4$ µm (Figure 2a), 3.5 µm (Figure 2b), 6.5 µm (Figure 2c), and 8.5 µm (Figure 2d). The Fourier axes are restricted to the positive $k_x/k_0$ from 0 to 1.5, as the features in the BFP images are prominent in this range and the color scales are adjusted to give the best possible contrast for the fringes (see Section S3.3 in the Supporting Information for the full data set). We make the following observations from the BFP images which are the main results of this Communication: 1. Alternate bright and dark intensity bands (fringes) are observed in the range $1 \leq k_x/k_0 \leq 1.5$ (where $k_x = \sqrt{k_x^2 + k_0^2}$) and are centered around the $k_x$ axis; 2. As $L$ is increased from 2.4 to 8.5 µm, more fringes are visible with a reduced spacing (fringe periodicity, $\Delta k_x/k_0$) between them.

Observation 1 clearly indicates the dominance of the light emission through glass at high angles ($1 \leq k_x/k_0 \leq 1.5$). Fringes with regular spacing in the Fourier plane are a characteristic feature of the interference between two coherent sources.\(^{[45]}\) The dependency of the spacing between the bright/dark bands on $L$ strongly suggests the interference between $S_1$ and $S_2$ as the increase in $L$ essentially increases the distance between the two sources, inducing an additional phase offset and thereby decreasing $\Delta k_x/k_0$. Furthermore,
the orientation of the fringes is mainly around the $k_x$ axis which also is a strong indication that the Au waveguide controls the interference between $S_1$ and $S_2$ as the $k_x$ axis coincides with the waveguide alignment in the experimental setup.

To confirm the experimental observations, we performed 3D finite difference time domain (FDTD) simulations using Lumerical.\textsuperscript{[48]} Inelastic tunneling in MIM-TJs is represented by a dipole source with a broadband spectral response (similar to the spectra shown in Figure 1d inset; see Section S4 in the Supporting Information for more details) and the BFP images are obtained from the near-to-far-field transformation of the near-field for the MIM-SPP and SPP scattering through glass. As the inelastic tunneling is spatially delocalized over the MIM-TJ area, more than one dipole is used to simulate the far-field light emission. The location of the sources is restricted along the edges of the MIM-TJ because the propagation length of the MIM-SPP is $\approx 50$ nm\textsuperscript{[18]} and sources positioned beyond this limit from the junction edge will have negligible contributions to the outcoupling as photons or plasmons (see Section S4 in the Supporting Information for more details). However, since the tunneling events are assumed to be completely noncorrelated,\textsuperscript{[49]} an incoherent summation is applied to the far-field intensity ($\sum E_i^2$, where $E_i^2$ is the far-field intensity from a dipole located at $(x_i, y_i, z_i)$ within the tunnel junction) to obtain the effective BFP images (see Section S4 in the Supporting Information). Simulated BFP images (right panels of Figure 2a–d) clearly reproduce the experimental observations. In particular, they reproduce the evolution of the fringe patterns with $L$. The fringe periodicity $\Delta k_x/k_0$ also shows a consistent trend and is further analyzed in Figure 3.

To gain deeper insight into the spatial correlation between $S_1$ and $S_2$ and to further compare the experimental and

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure2.png}
\caption{BFP images of the light emission from the TJs for $L = \text{a) } 2.4 \, \mu\text{m}, \text{ b) } 3.5 \, \mu\text{m}, \text{ c) } 6.5 \, \mu\text{m}, \text{ and d) } 8.5 \, \mu\text{m}. \text{ All images are collected for } V_b = -1.5 \, \text{V. Left panels show the experimental BFP images, and the right panels show the BFP images from FDTD simulations. Fourier axes are restricted to the positive $k_x/k_0$ (from 0 to 1.5), as the fringe patterns are dominant in this range. } \Delta k_x/k_0 \text{ represents the fringe periodicity. Color scales of the BFP images are adjusted to give the best possible contrast for the fringes.}}
\end{figure}
simulation results, we evaluated the BFP emission intensity along the horizontal dotted lines shown in Figure 2a–d and plotted in Figure 3a. To visualize the evolution of the fringes with L (=2.4, 3.5, 6.5, and 8.5 μm), the line profiles are normalized to unity and are shifted vertically in the vertical axis. The dotted lines are from the experimental BFP images and solid lines are from the FDTD simulations. For L = 6.5 μm (Figure 3a) the fringe peak positions (upward vertical arrows) from the experiments are in excellent qualitative agreement with the simulations results (in terms of kx/k0).

Minor offsets in the peak positions are observed for L = 2.4, 3.5, and 2.4 μm (Figure 3a) between the experiment and simulation and can be explained as follows. In general, the phase offset between S1 and S2 dictates the peak positions of the fringes and in our case, it crucially depends on the SPP propagation losses of the Au stripe waveguide. Apart from the material absorption, these losses are at a minimum in the simulations due to the lack of the roughness factor and the discrepancy of the FDTD with the experiments will be higher for longer Au stripes, particularly for L = 8.5 μm. On the other hand, SPP reflection at the stripe-end may not be completely neglected due to the impedance mismatch, especially for L = 2.4 and 3.5 μm and can lead to the observed discrepancy between experiment and simulation. For L = 6.5 μm, a trade-off between these two effects can be observed, resulting in a good agreement between the FDTD simulation and the experiment.

From Figure 3a, the fringe periodicity (Δkx/k0) is calculated as a function of L and is plotted in Figure 3b. Open rhombs represent the experimental data; open circles are the results from the 2D FDTD simulations (see Section S4.3 in the Supporting Information). From the phase offset between S1 and S2, Δkx/k0 can be written as[34]

$$\frac{\Delta k_x}{k_0} = \frac{\lambda_0}{L}$$  \hspace{1cm} (2)

where λ0 represents the excitation wavelength. The solid line in Figure 3b is a fit to Equation 2 with λ0 = 900 nm (peak position in the emission spectra, see Figure 1d inset). Good agreement is observed between the experiment, FDTD results, and the model given by Equation 2, which further confirms the proposed interference model in the present study.

As L increases beyond the propagation length of SPPs, the propagation losses of the Au waveguide will further increase the phase offset between S1 and S2, eliminating any interference effects. In Figure 4a, we show the real plane (left) and BFP (right) of a MIM-TJ with an Au waveguide of L = 13.5 μm. We observe that, even though S1 and S2 are both visible in the real plane image, the BFP image does not reveal any fringe patterns, which indicates the uncorrelated nature of the two sources leading to the absence of interference fringes.

We further discuss the BFP images from the devices with S1 selectively masked by a spatial filter (pinhole).[50] Figure 4b shows the real plane (left) and BFP (right) images for the case of L = 6.5 μm (Figures 1c and 2c). The real plane image is overlaid with the bright-field image of the device from the EMCCD to explicitly show the device-outline and the position of the pinhole (dotted-line circle with radius = 6 μm). So, only the far-field emission from the exposed region (stripe-end) is recorded in the BFP with a pinhole inserted to the image plane[51] (see Section S3 in the Supporting Information). The source S1 (Figure 1) from the junction edge is mainly directed towards the forward propagation direction (k0) of the SPP modes in the Au stripe waveguide. As S1 is blocked, the far-field interference between S1 and the forward scattering SPPs is completely absent and thereby the fringe patterns are not observed in the BFP which only shows the SPP scattering from the stripe-end. We contrast this with our recent work,[18] where S1 is inherently absent due to the geometric orientation of the TJ (in-plane, vertical to the glass substrate) and therefore fails to generate distinct fringes in the BFP. Similarly, we blocked S2 and recorded the BFP image, which only shows the light scattering...
off the surface roughness in the MIM-TJ area, with no distinct fringes as the far-field interference is absent in this case as well (see Section S3 and Figure S6 in the Supporting Information for the recorded images).

Finally, to further validate the insights gained from the fringe-analysis, we discuss the experimental and simulation results from the width dependence study (Figure 4c) where the width ($W$) of the Au stripe is changed whereas $L$, which crucially affects the periodicity of the interference fringes, is kept constant at 3.5 µm. In Figure 4c we plot the BFP intensity along the $k_x/k_0$ axis (see Figure S5 in the Supporting Information for the real plane and BFP images), where the dotted lines are from the experimental BFP images and solid lines are from the FDTD simulations. As expected, the number of fringes remains constant in the BFP images with the change in $W$, which further corroborates the interference model. The experimental and simulation results are in excellent agreement. We also note that for $W = 0.8$ µm, the fringes show a straight-line character (Figure S5, Supporting Information) as compared to the case of $W = 2$ µm where the fringes are semi-circular in shape with respect to the $k_x/k_0$ axis.

### 3. Conclusions

The study presented here demonstrates the coherence in plasmonic outcoupling channels from biased MIM-TJs. As the MIM-SPP excitation corresponds to a single inelastic tunneling event, its coherence with the outcoupling channels (photons and SPPs) is preserved and by a systematic control of the SPP stripe waveguide length, the spatial coherence between MIM-SPP and SPP/photons can be controlled. Alternate bright and dark fringes observed in the Fourier plane images are the indicators of the strong interference in the far-field photon outcoupling, and the calculated fringe-spacing as a function of the stripe-length follows the phase correlation relation between two coherent sources separated by the stripe length $L$. Numerical FDTD simulations support the findings and the results from long SPP stripe waveguides (> 8.5 µm) and spatially filtered BFP images (where one of the sources is selectively masked) further corroborate the spatial correlation picture. This correlation further unambiguously confirms the two-step plasmonic decay process in biased plasmonic TJs where an electron first excites the MIM-SPP junction mode which then excites other available optical modes. For the broadband excitation from inelastic tunneling, the SPP stripe waveguide crucially maintains the correlation, leading to a coherent superposition in the far-field between individual frequency components. Furthermore, the electrical excitation scheme employed here readily excites bound-SPP modes as compared to the optical schemes where leaky-SPP modes are mainly excited,[52] thereby minimizing the SPP leakage losses in spatial correlation studies.

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**Figure 4.** a) Real plane (left) and BFP image (right) for the light emission from the junction with an associated Au waveguide of $L = 13.5$ µm, where the BFP image does not show a fringe pattern. The images are recorded for $V_b = -1.5$ V. Intensity is rescaled by a factor of 8 at the Au stripe-end (rectangular dotted-line box in left panel). b) Real plane (left) and BFP image (right) for the light emission from the junction with a spatial filter introduced for the $L = 6.5$ µm case (Figures 1c and 2c). The real plane image is overlaid with the bright-field image of the device from the EMCCD. The dotted-line circle represents the pinhole with radius $\approx 6$ µm. c) BFP emission intensity showing the evolution of the fringe patterns with the waveguide width ($W = 0.8, 2, \text{ and } 4$ µm). For the Au stripes, $L = 3.5$ µm and thickness $= 150$ nm. Line profiles are normalized to the maximum value and are shifted vertically for better visibility. Dotted lines are from the experimental BFP images and solid lines are from the FDTD simulations.
The inherent coherence associated with the electrically excited SPPs demonstrated here with a simple and efficient experimental design points to the potential of these systems towards the plasmonic computation at the nanoscale where the spatially correlated and actively modulated sources are essential. Our study also indicates that the plasmon excitation from tunneling can be further exploited for plasmonic applications with tailored spatial coherence, especially for phase correlated measurements in quantum information processing that benefit from spatially coherent sources, particularly with electrical and optical control over the excitation and propagation. Moreover, the composite device structure of a TJ and a plasmonic waveguide used here can be further extended for temporal coherence studies in mode outcoupling from tunnel junctions which support resonant MIM-SPP modes.

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.

Data Availability Statement
The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords
back focal plane imaging, coherent tunnel junctions, electrical excitation of plasmons, interference, near-field coupling, optical coherence

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