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Quantum plasmonics: quantum information at the nanoscale 122054

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### Executive summary of our findings

Highlights of our research programme include the demonstration of Hong-Ou-Mandel interference of surface plasmon polaritons [1], proving directly the bosonic nature of surface plasmons. This constitutes a first step towards active nanoscale quantum information processing devices based on surface plasmon polaritons. We demonstrate that despite the high losses in surface plasmon waveguides, which we investigated using single-photon to single-plasmon conversion [3], single surface plasmon polaritons can interfere quantum mechanically and show the characteristic bunching at the output ports of a four-terminal device. The second highlight is the publication of a roadmap for the field of quantum plasmonics [2], published in *Nature Physics* with cover page of the journal. In total this project has thus far resulted in six journal articles. We are currently writing up an additional work, on direct quantum tomography on state entanglement in quantum interferometers, which could form the basics of quantum sensing [4].

### Context

The two main goals of this project are i) the investigation of the physical nature of single surface plasmon polaritons (SPPs), focusing particularly on their quantum nature and quantum statistical behaviour, and ii) the demonstration of switching and modulation of single SPPs based on quantum interference phenomena.

Of particular importance is the careful assessment of the issue of optical losses on the quantum properties: for SPPs, there exists a fundamental trade-off between localization and loss – the tighter confined the SPP modes are, enabling sub-diffraction-limit waveguide modes, the more optical losses due to Ohmic damping is encountered. In this project we conducted fundamental quantum optical experiments in such a highly lossy environment in order to assess the feasibility of nanoscale quantum optics based on SPPs.

### Description of main results

## I. Demonstration of loss-robustness and interference of single SPPs

Early in the project we already demonstrated that the quantum statistics of single surface plasmon polaritons are not negatively affected by high optical losses [3]. Namely, we showed that the quantum statistics of single photons converted into single SPPs stay unaffected by the high optical losses of a plasmon waveguide channel, during a photon-SPP-photon conversion experiment. This demonstrated the Markovian nature of the optical losses encountered by SPPs at the single quantum level.

The next significant step, which we worked on during calendar year 2013 and the early months of 2014, was the direct demonstration of the bosonic nature of single SPPs utilizing a plasmonic version of the well-known Hong-Ou-Mandel interference experiment (Figure 1). This experiment forms the basis of our understanding of single SPPs, and further more will be enabling for modulation of single SPPs via quantum interference experiments. The work has now been published [1].

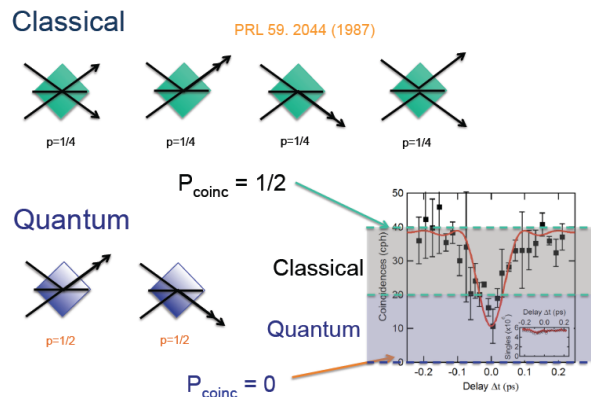


Figure 1. Bunching of photons at the output port of a 4-port beam splitter due to quantum interference. In order to reach the quantum regime, the coincidence count dip (the “visibility”) must be higher than 50%.

The basis of Hong-Ou-Mandel interference is the observance of bunching of photons at the output ports of a 4-port beam splitter, as shown in Figure 1. Quantum interference manifests itself via both photons detected in the same output port according to

$$|1\rangle_A |1\rangle_B \rightarrow \frac{1}{\sqrt{2}} (|2\rangle_{B_1} |0\rangle_{B_2} + |0\rangle_{B_1} |2\rangle_{B_2})$$

This can be detected using an interferometric setup, basically counting coincidences of photon detection at the two output ports. For zero time delay, a dip in coincidences will be observed, which for unambiguous observation of quantum interference needs to be larger than 50%.

A schematic of the plasmonic Hong-Ou-Mandel experiment conducted is shown in Figure 2, utilizing a plasmonic beam splitter designed for a 50-50 splitting ratio (Figure 3), which is required by the Hong-Ou-Mandel interference scheme. In order to overcome the detrimental influence of optical losses on the splitting process, we decided against simple co-directional coupling, but instead employed scattering of a specially designed nanoparticle Bragg grating in the centre part of the plasmonic beam splitter (Figure 3a) to give us the desired splitting ratio.

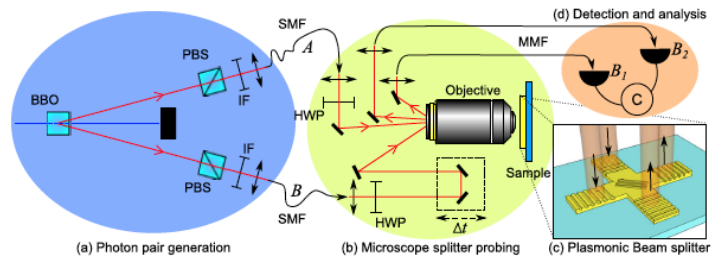


Figure 2. Schematic of our plasmonic Hong-Ou-Mandel interference experiments utilizing a 4-port plasmonic beam splitter.

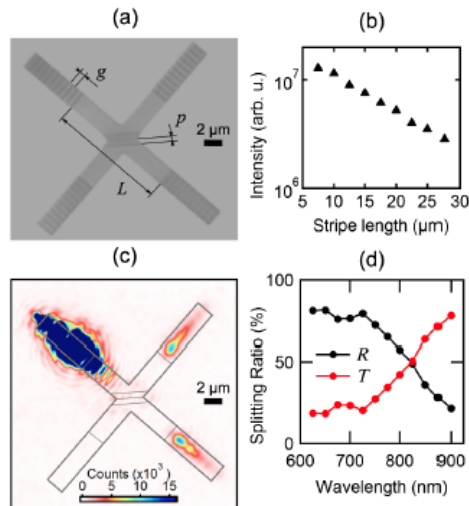


Figure 3. Optical micrograph of the plasmonic beam splitter (a), loss characterization (b), and demonstration of the 50-50 splitting ratio (c, d) necessary for Hong-Ou-Mandel interference.

The central result of this study is shown in Figure 4 – the visibility (depth of the coincidence dip) is well below the 50% required for demonstration of quantum interference (72%). This constitutes direct evidence of the bosonic nature of SPPs, despite the hybrid photon-electron character of this excitation. Furthermore, the successful interference now puts active modulation of single SPPs in the realm of possibility, for example in quantum sensing experiments based on additional phase shifts induced by molecular binding in one of the plasmonic beam splitter arms.

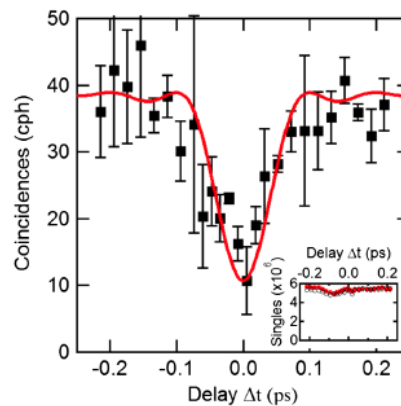


Figure 4. Coincidence count versus time delay for the two output ports, demonstrating quantum interference with a visibility of 72% [1].

The fact that quantum plasmonic waveguides show both a robustness of their quantum statistics to loss [3] and allow for interference of surface plasmons [1] should allow for their incorporation into sensing devices for low level of molecules. We have investigated the limitations in terms of attainable signal-to-noise ratio and compactness for a variety of waveguide geometries [4], concluding that indeed careful preparation of so-called noon states should allow for a lowering of the signal to noise ratio below the traditional shot-noise-limit, despite the high losses of these compact waveguides. A generic schematic of such a sensing scheme is shown in Figure 5.

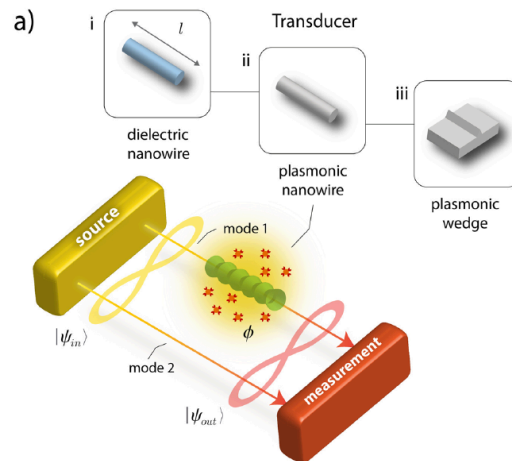


Figure 5. Schematic of a quantum sensing device allowing detection beyond the limits of shot noise and diffraction [4].

## II. Quantum tomography of single SPPs

As mentioned in the first part of this report, we obtained direct evidence of the bosonic nature of SPPs by observing a two-SPP interference, the plasmonic version of the Hong-Ou-Mandel experiment [1], in a scattering based waveguide beam splitter. In this seminal experiment, Hong, Ou and Mandel showed that two single photons arriving simultaneously at the two input ports of a beam splitter will always leave the beam splitter together at one of the two output ports. This is a completely non-classical effect and was considered one of the first direct proofs that the electromagnetic field needs to be quantized, photons are bosons. We reported an interference visibility of 74 %, a clear breach of the classically possible maximum visibility of 50 %, SPPs are thus bosons. For clarity the results are additionally summarized in Figure 6.

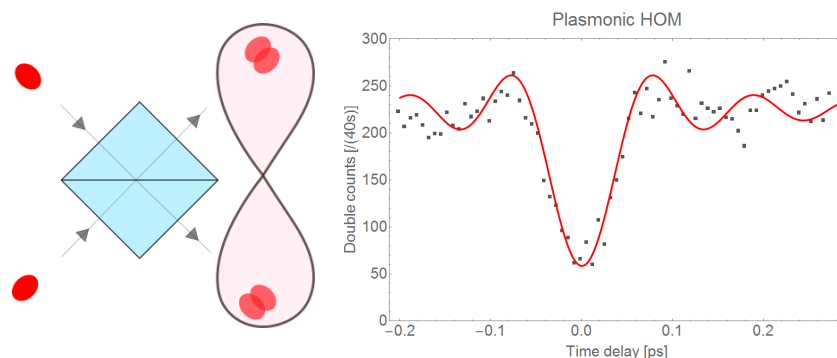


Figure 6: Left, visual representation of a HOM interference, the two photons leave the beamsplitter in a superposition (entangled) state of both photons being together at the upper and lower output, this is known as the 2 photon NOON state:  $|20\rangle + |02\rangle$ . Right, experimental result of this interference with SPPs in a plasmonic beam splitter, a strongly non classical visibility of 74 % was obtained.

The beam splitter consists of two crossing gold waveguides with a Bragg reflector consisting of a low amount of gold ridges patterned on the crossing, see inset figure 7. The total size of the structure is only several microns large, with an interaction region of a wavelength, showing the miniaturization potential of plasmonic structures for integrated -quantum-photonic. The optical set-up has been completely rebuilt since the first publication [1], in order to get higher and more reliable visibilities. This allowed us to attribute the non-perfect interference to parameters like the multimodal behavior of the waveguides and the deviation of the beam splitter output from the well-studied lossless case.

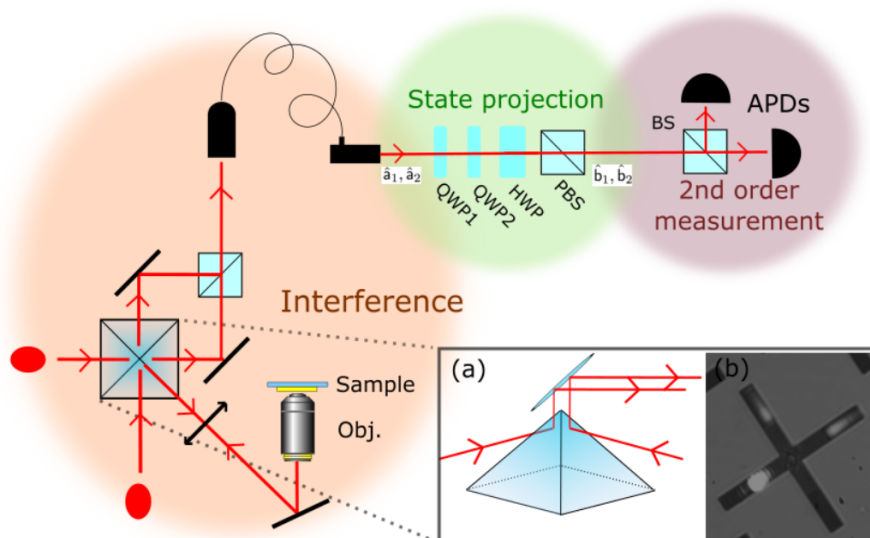


Figure 7: Simplified schematic drawing of the set-up. With a complex image system, the photons are focussed on the input gratings of the plasmonic sample (inset b) with an objective. The SPPs scattered into free space at the two outputs are collected by the same objective and superimposed on a polarization maintaining beam splitter. This allows us to perform quantum state tomography in the polarization basis by a combination of state projections and two-photon measurements.

It also provided the opportunity for in-depth investigations of the generated quantum state of light by quantum state tomography. This would allow to prove and quantify entanglement generated in the interference. To reach these goals, a new scheme was developed and set-up built in which the path-entanglement of the output state is transferred to the polarization basis, where well-established techniques allow us to fully reconstruct the density matrix. This contains all the possible information of the quantum state. In figure 8 below, a result is shown of this measurement on the output of the plasmonic interference. The density matrix allows us to calculate specific measures like the Entanglement of Formation, where our result of  $0.63 \pm 0.09$  reliably shows that the SPP interference generates entanglement, as a non-entangled state produces an Entanglement of Formation of 0. Experimental prove of the generation of entanglement in a plasmonic system has, to our knowledge, not been done before. This further opens the way to more fundamental studies of the influence of SPPs on the quantum information of states and shows that plasmonics could become a viable route for quantum technologies.

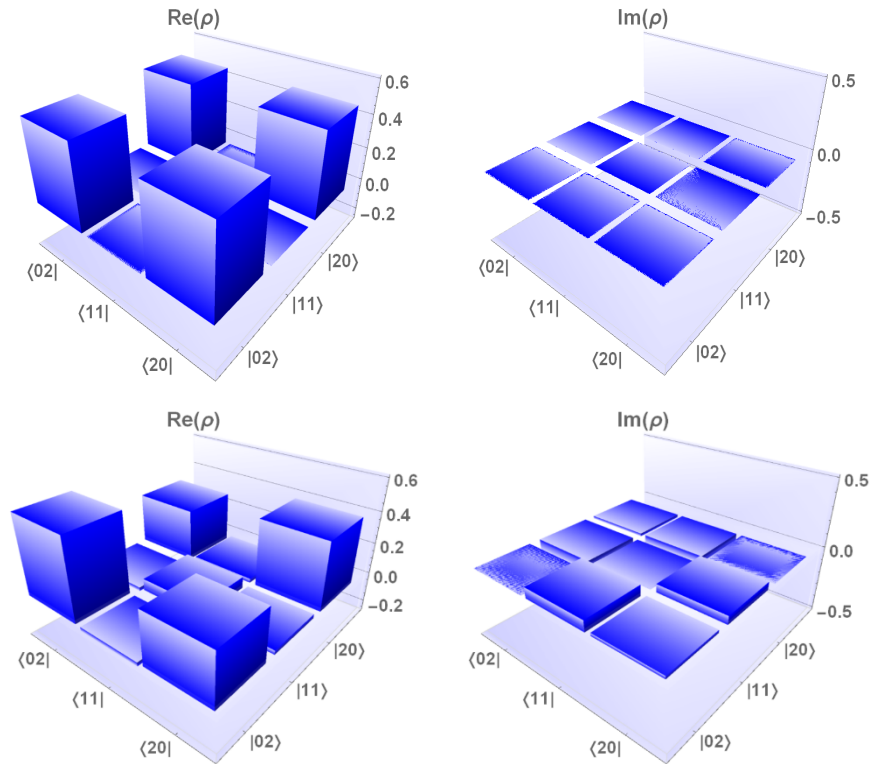


Figure 8: Top: Real and imaginary components of the theoretically expected density matrix of the 2 photon NOON state  $|20\rangle + |02\rangle$ . Bottom: Experimentally obtained result with our quantum tomography set-up on the output of the HOM interference in the plasmonic beam splitter. A good agreement between the two is clearly visible, quantified as the Fidelity of the state, here  $0.77 \pm 0.04$ . (A Fidelity of 1 means perfect overlap)

## II. A roadmap for the further development of quantum plasmonics

The second highlight of the current performance period was the development and high profile publication of a roadmap for the further development of the field of quantum plasmonics [2]. The respective review and roadmap article was published in *Nature Physics* and got highlighted via a cover picture (Figure 9). Apart from surveying the current state of the field and explaining the basic nature of the quantum properties of SPPs, we outlined challenges and opportunities for the field and defined a set of milestones for its further development.

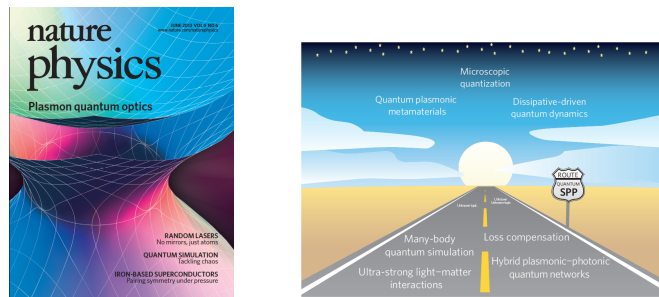


Figure 9. Publication of a review article of quantum plasmonics [2] including a roadmap for the further development of the field

### III. Additional research topics

During the last 24 months we obtained two additional results not envisaged in the original research proposal. The first one concerns the development of a negative permeability quantum metamaterial based on the coherent coupling between quantum dots and metallic nanoparticles [5] (Figure 10). Such a material might prove a route towards switchable artificial magnetic media exploiting quantum effects, for example for the generation of perfect lens imaging modalities or optoelectronic devices with tunable gain profile.

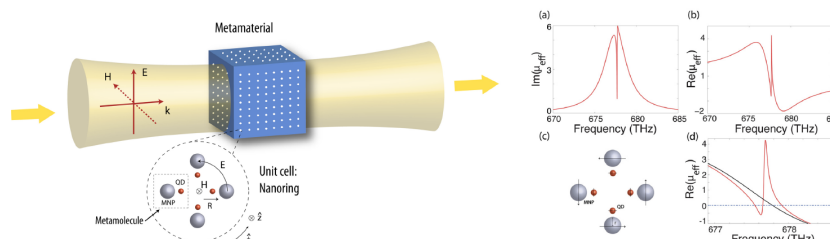


Figure 10. A quantum plasmonic metamaterial consisting of quantum dots coupled to metallic nanoparticles in order to obtain negative permeability in the optical range [5].

The second side project was inspired by our work on quantum statistics preservation under high loss [3] and details a way of entanglement generation between two quantum dots utilizing a chain of metallic nanoparticles as the coupling channel [6] (Figure 11). As we show, this particular entanglement scheme is robust to the loss occurring in the system, thus providing a route towards entanglement generation in highly confined waveguides.

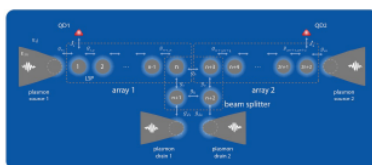


Figure 11. A study of entanglement preservation between two quantum dots coupled to a plasmonic nanoparticle waveguide [6]. We demonstrate that this particular scheme of entanglement is robust to the high losses occurring due to the strong confinement in these waveguides.

### Conclusions

We have demonstrated robustness to loss and entanglement of surface plasmon polaritons at the single plasmon level. This opens up opportunities for the usage of quantum plasmonics in sensing, modulation and switching devices, as well as for fundamental investigations of quantum state manipulation on the nanoscale. Our work will give a boost to the field of quantum plasmonics, and we have in August 2016 secured a three-year research programme by the Leverhulme Trust to continue our work.

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