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Aston Institute of Photonic Technologies

WINGS

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PART A

Aston University

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Declaration

I, Giuseppe Marino, hereby declare that

- Both Part A and Part B of this Report and its related appendices have been approved by the Scientist in-charge, Giuseppe Leo, and any other relevant party (for e.g. secondment academic/non-academic organisation).
- The contents of the publishable summary (Section 1 of Part A) do not contain any confidential data, and have been approved by the Scientist in Charge and any other relevant party ((for e.g. secondment academic/non-academic organisation) involved in the generation of the Results.

Signed,

Giuseppe Marino

Paris, 31-08-2019

1. Summary for publication

1.1 Summary of the context and overall objectives of the project

Our project aims at developing novel **nonlinear wave shaping devices based on light structuring at the nanoscale** operating in the visible (VIS) and near-infrared (NIR) spectral regions for nonlinear nano-scale light sources of quantum light. The nonlinear optical waves generated by such components can find a broad range of applications such as three-dimensional optical transport along predefined trajectories [1], hologram formation [2, 3], particle trapping [4], imaging [5, 6], filamentation [7, 8], and spatial multiplexing of several sources of heralded photons [9].m In particular, WINGS addresses the application of nanoscale nonlinear beam shaping on dielectric metasurfaces for the **quantum-state engineering of signal-idler photon pairs generated** by spontaneous parametric down-conversion (SPDC).

Optical metasurfaces are arrays of coupled optical antennas, with sub-wavelength (sub- λ) size and separation. At variance with current optical components, which need propagation distances of several wavelengths to alter the phase front, metasurfaces can mold polarization, amplitude and phase of light at a sub-λ scale. The latter is an intermediate scale between the case where the optical structures are close or larger than λ and the limit case where they are much smaller than it (like in Rayleigh scattering). In such intermediate scale, corresponding to resonant Mie scattering, neither diffracted orders propagate nor the medium can be seen as being homogeneous (as for example in form birefringence).

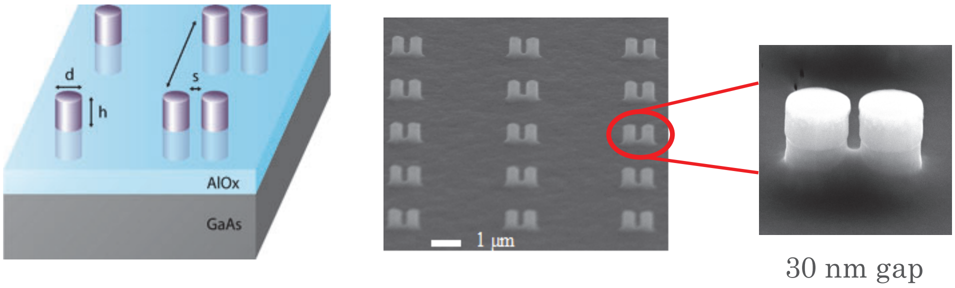
The targeted breakthrough of the WINGS project is the demonstration of **1)** a metasurface implementing beam shaping on the field generated by classical up- or down-conversion; **2)** a metasurface for the quantum-state engineering of signal-idler photon pairs generated by spontaneous parametric down-conversion (SPDC).

1) Via a fundamental understanding of the physical mechanisms associated to sum-frequency generation (SFG) and difference-frequency generation (DFG) in dielectric metasurfaces, we will enhance the conversion efficiency at levels $\geq 10^{-4}$ at pump intensities in the GW/cm^2, i.e. at least one order of magnitude above the state of the art of single nanoantennas. Control and tunability of polarization, frequency, and directionality will be implemented by varying the geometry of the nanoantennas, reaching phase control of the generated field over the 0-2π range. With the 2D amplitude distribution of the generated field set by arranging different nanoantennas in spatially inhomogeneous metasurfaces, an additional control beam will finally be used to dynamically vary the optical properties of the nanoantennas. *All the above degrees of freedom will be exploited to demonstrate beam shaping on the field generated by SFG and DFG.*
2) We will utilize our $\chi^{(2)}$ metasurfaces to tailor quantum optical emission. To this end we will start by performing 3D electromagnetic simulations and compared with spectroscopical measurements. The flexibility of our metasurfaces will allow for an enhanced control of the properties of the generated two-photon states, which will be tested by quantum correlation measurements. Controlling the joint and individual spectra of signal and idler has been a much-researched topic for traditional SPDC sources. This is relevant for entanglement swapping [13], which is part of any quantum repeater protocol. By designing the spatial properties of the metasurface we will be able to generate new forms of spatial entanglement, which can give rise to new quantum imaging functionalities. Finally, by working on the nanoantenna shape we will be able to control the polarization properties and generate hyper-entanglement, i.e. entanglement in multiple degrees of freedom, which enhances quantum applications like dense coding [14]. *This theoretical and experimental study will enable to demonstrate photon-pair generation with controllable spatial, polarization and entanglement properties.*

In recent years, metasurfaces have gained enormous momentum because of their promise for ultrathin devices compatible with planar fabrication technology that can potentially replace bulky optical components limited by diffraction[10, 11]. Many plasmonic metasurfaces with beam bending [12, 13], beam focusing [14, 15], hologram formation [2, 3], and beam shaping [9, 16] capabilities were developed in the last five years only. At the same time, large absorption losses and complex fabrication processes associated with metals have also fueled research into dielectric based metasurfaces with light shaping capabilities such as hologram formation [17, 18], beam generation [17, 19], and strong directional emission by either exploiting magnetic and electric dipole resonances [20, 21] or the geometric phase originating from space-variant polarization manipulations (Pancharatnam–Berry phase approach [17]). So far, wavefront manipulation has been performed mainly using linear phenomena. However, this functionality can also be achieved by exploiting nonlinear optical processes [22-30]. This can open the way to many new possibilities that go beyond the current state-of-the-art functionalities of linear beam shaping techniques. **Nonlinear beam shaping** would allow the **generation of shaped light at new frequencies,** by increasing the possibilities for ultrafast **all-optical control of the generated wavefront,** and **reducing costs and**

size as compared to other setups [26, 27]. Recently, plasmonic metasurfaces showed the potential for smaller device size and higher spatial resotion and accuracy, thanks to a continuous control over the amplitude of the local nonlinear response [24, 26]. However, frequency generation in plasmonic nanostructures usually suffers from low conversion efficiency. Even though this could, in principle, be increased by using hybrid metal-dielectric resonators or intersubband transitions in multi quantum well structures [25, 26], **all-dielectric metasurfaces** provide a much simpler and effective solution for nonlinear beam shaping. **WINGS aims at achieving the independent control of phase and amplitude of the second harmonic (SH) optical response** based on the manipulation of magnetic and electric dipole resonances at the scale of macro-pixels which are filled with a certain number of weakly interacting AlGaAs nanoantennas. We will control its amplitude and phase responses via the number of nanoantennas contained in it and its geometrical parameters. Such a flexibility has several advantages compared to previous methods, making the direct near field generation of the desired beams possible with two degrees of freedom for the independent shaping of the trajectory and the amplitude of the beam [1]. In this context, as a benchmark to show their potential, the WINGS project will tackle the generation of nonlinear metasurfaces light sources of quantum light which offers an unexplored potential for highly indistinguishable and reconfigurable spatial photonic modes [3], as well as for spatial multiplexing of several sources of heralded photons [9].

In the perspective of developing on chip beam shaping devices, **the WINGS material system of choice** is **AlGaAs-on-oxide**. AlGaAs exhibits a high refractive index in the VIS and NIR spectral regions; moreover, this material platform will enable the fabrication of monolithic flat optical components in a single lithographic step. With respect to silicon on insulator (SOI) which is used for example in [18], AlGaAs is promising under several standpoints: monolithic **integration** with GaAs-based light sources; broader spectral range of operation thanks to the control of the electronic band gap energy via Al molar concentration; **stronger nonlinear optical response** with the possibility of **minimizing nonlinear absorptions** at VIS and NIR wavelengths; lack of **centrosymmetry**, resulting in strong second-order nonlinearities. In recent experimental demonstrations conducted at a pump wavelength of 1550 nm a conversion efficiency was found up to 7×10^{-5} for a 1.6 $\mathrm{GW/cm^2}$ pump [31]. This result supported our theoretical investigations of SHG from single AlGaAs nanoantennas [32, 33] which predicted a **conversion efficiency** 6 orders of magnitude higher than in gold nanoantennas [34]. Recent experiments [35-37] show the huge potential of AlGaAs nanostructures for nonlinear nanophotonics. More recently, the WINGS team has focused its investigations on AlGaAs nano-dimers shown in figure 1 [38], which can be seen as a first step towards the demonstration of a $\chi^{(2)}$ metasurface.

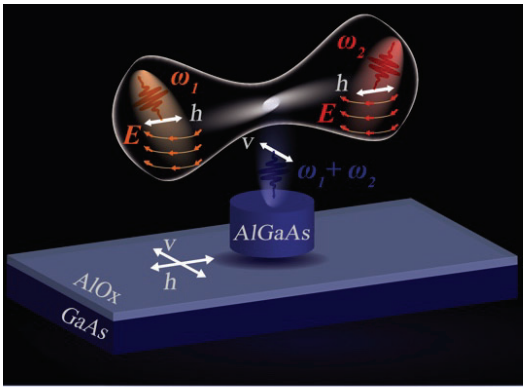


a) b) c)

Figure 1. *From AlGaAs pillars to dimers: a) scheme; b) electron-microscope picture; and c) zoom on a single dimer.*

The latter is a two-dimensional arrangement of a large number of nanoresonators with subwavelength distances, whose nonlinear emissions interfere coherently. The WINGS metasurface, with a 1 mm^2 footprint and internal distances smaller than 25 nm, will bridge the gap between the nanometer and millimeter scales.

The WINGS project is original in several respects. The only SPDC preliminary experiment at the nanoscale has been carried out by us on a single nanocylinder [39]. However more efficient spatial and polarization correlations of photon-pairs generated through SPDC in metasurfaces made of an array of single nanocylinders is still unexplored. In our work [39], we demonstrated experimentally nonlinear AlGaAs nano-disk sources of efficient sum-frequency light and high rate heralded photons with non-classical



correlations via SPDC, establishing a quantum-classical correspondence between these two processes (see figure 3). While most nonlinear frequency conversion applications to date rely on bulk nonlinear crystals, there is continuing quest for miniaturization of the nonlinear light sources to micro and nano-scale dimensions. Such miniaturization would allow denser integration of nonlinear light sources and ultimately smaller and functional nonlinear devices for classical and quantum applications. The success for these applications requires the development of highly efficient mixing of different wavelengths in nano-scale elements, which are scalable to large area fabrication. The realization of metasurfaces of nonlinear nano-scale light sources of quantum light therefore offers an unexplored potential for highly indistinguishable and reconfigurable spatial photonic modes [3], as well as for spatial multiplexing of several sources of heralded photons [9]. Our project encompasses theoretical and numerical **modelling**, III-V semiconductor **fabrication** processes, linear and nonlinear optical **characterization** of nanostructures, including optical trapping experiments. A close international collaboration with Brescia University (UNIBS), ANU and Politecnico di Milano (POLIMI) completes the MULTIPLY triple–I paradigm, where the above interdisciplinary and intersectoral actions perfectly marry with a high-level international dimension of the project.

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1.2 Work performed from the beginning of the project to the end of the period covered by the report and main results achieved so far

WINGS focus of attention has been put on understanding and controlling the interactions among resonances at a level of individual nanoantennas for the development of appropriate design rules for their ultimate applications, especially with regards to nonlinear metasurfaces. In order to achieve the above task, we have performed cathodoluminescence imaging spectroscopy on both single AlGaAs nanoantennas and their hybridization in a dimer configuration [40]. We have also exploited the effective coupling of electric and toroidal modes in AlGaAs nanodimers to locally enhance both electric and magnetic fields while minimizing the optical scattering, thereby optimizing their second harmonic generation (SHG) efficiency with respect to the case of a single isolated nanodisk [38]. We have further presented a nonlinear hybrid antenna based on an AlGaAs nanopillar at coincidence with the anapole mode, surrounded by a gold ring, which merges in a single platform the strong field confinement typically produced by plasmonic antennas with the high nonlinearity and low loss characteristics of dielectric nanoantennas. This platform allows boosting by more than one order of magnitude both second- and third-harmonic generation conversion efficiencies [41]. Then, we have demonstrated, both experimentally and numerically, the control over the angular distribution of the second harmonic radiation pattern from a monolithic AlGaAs-on-AlOx nanodisk by varying the polarization and the angle of incidence of the pump beam. By tuning the angle of incidence from 0° to 45°, the detected second harmonic signal is monotonically increased up to over an order of magnitude thanks to the strong illumination dependence of the nonlinear radiation pattern when *s*-polarized light is employed. Our results demonstrate that precise angular measurements of the pump inclination can be performed with a sensitivity of up to 0.25 $(°)^{-1}$ and polarization discrimination, thus establishing a new technique for background-free nanoscale nonlinear goniometry [42] We have also demonstrated the shaping of the second-harmonic (SH) radiation pattern from a single AlGaAs nanodisk antenna using coplanar holographic gratings. The SH radiation emitted from the antenna towards the - otherwise forbidden - normal direction can be effectively redirected by suitably shifting the phase of the grating pattern in the azimuthal direction. The use of such gratings allows to increase the SH power collection efficiency by two orders of magnitude with respect to an isolated antenna and show the possibility of intensity-tailoring for an arbitrary collection angle. Such reconstruction of the nonlinear emission from nanoscale antennas represents the first step towards the applications of all-dielectric nanostructures for nonlinear holography [43]. Finally, nanoantennas can further open opportunities for multi-photon nanoscale light sources, where novel effects of quantum entanglement and interference at the nanoscale can be explored. Thus, we have demonstrated experimentally an AlGaAs optical nanoantenna, where the high $\chi^{(2)}$ allows the generation of spontaneous photon pairs [44]. The demonstrated nanoscale two-photon emitter allows flexible quantum state engineering by shaping of the spectral and radiation pattern of the nanoantenna and paves the way to more efficient photon pair-generation rates, by either tuning the incident wavelength and angle, or using a doughnut-like cylindrical vector pump beam, or optimising the nanoantenna geometry, or performing transmission detection. For example, in recent works [45,46], we propose meta-atoms and metasurfaces with engineered SHG phase and efficiency from 24 to 550 times bigger than the simple nanoantenna.

1.3 Progress beyond the state of the art, expected results until the end of the project and potential impacts (including the socio-economic impact and the wider societal implications of the project so far)

In recent years metasurfaces have gained enormous momentum because of their promise for ultrathin devices compatible with planar fabrication technology that can potentially replace bulky, diffraction-limited optical components [11]. Many plasmonic metasurfaces with beam bending [12], beam focusing [14], hologram formation [3], and beam shaping [9] capabilities have been developed. So far, however, wavefront manipulation has been performed mainly using linear phenomena, although the exploitation of nonlinear optical processes [23] would open the way to many new possibilities beyond the functionalities of linear beam shaping. Nonlinear beam shaping would allow the generation of shaped light at new frequencies, by increasing the possibilities for ultrafast all-optical control of the generated wavefront, and reducing costs and size. While most nonlinear frequency conversion applications to date rely on bulk nonlinear crystals, there is continuing quest for miniaturization of the nonlinear light sources to micro and nano-scale dimensions. Such miniaturization would allow denser integration of nonlinear light sources and ultimately smaller and functional nonlinear devices for classical and quantum applications. The success for these applications requires the development of highly efficient mixing of different wavelengths in nano-scale elements, which are scalable to large area fabrication. In this context, the work on SHG from AlGaAs nanoantennas that I had the privilege to carry out during the Fellow has greatly contributed to the engineering of nonlinear light at the nanscale [38,41-43]. Then, we have demonstrated experimentally an AlGaAs optical nanoantenna for efficient generation of two-photon quantum states based on the process of spontaneous parametric down-conversion[44]. Our experimental results open the way for a sub-wavelength arrangement of multiple nonlinear nanoantennas in a designed metasurface array, to yield a coherent generation of multi-photon quantum states with complex multi-mode spatial entanglement for a range of applications, including free-space quantum communications and sensing. In more recent works [45,46], we have proposed original meta-atoms and metasurfaces with engineered SHG phase and efficiency up to 550 times bigger than the simple nanoantenna.