

## Publishable Summary for 20FUN05 SEQUME

### Single- and entangled photon sources for quantum metrology

#### Overview

Quantum technologies (QT) are one of the most relevant contributors to innovation and advanced technologies. However, there is an important range of quantum-enhanced measurements that are not yet exploited by national metrology institutes (NMIs) because single-photon and entangled-photon sources with the required performance parameters (e.g. purity for single-photon sources and sub-shot noise quantification for entangled sources), are not readily available. Major advancements have been made in engineering these sources, but if they are to be used in metrological applications, significant further development is necessary. This project will develop high brightness, high-efficiency entangled photon sources (based on semiconductor quantum dots (QDs)) and it will exploit high-purity single-photon sources (based on ion implantation in diamond and on single molecules), to demonstrate the quantum advantage achieved when using these sources for specific measurements.

#### Need

Several international, European and national quantum programmes are currently under way, e.g. the European “Quantum-Flagship”, Quant-ERA, ITN Networks, the German QT initiatives, the UK Quantum Technologies programme (which was recently re-launched), as well as programmes in Australia, Austria, Canada, China, Japan, Russia, Switzerland, and in the US. However, these initiatives do not yet comprehensively address the metrological challenges related to QT, such as traceability, standardisation and optimisation of quantum enhanced measurements. A robust European metrology infrastructure is required that enables the traceable characterisation of entangled-photon and single-photon sources.

The Strategic Research Agenda for Metrology in Europe has identified a need for further work in quantum metrology, to ensure Europe keeps at the leading edge of QT development worldwide. Noise limits must be overcome and the invasiveness of measurements (i.e. disturbance on the tested objects) must be minimised, both of which can be achieved using novel quantum-enhanced approaches. For this, single-photon sources and entangled-photon sources used in traceable quantum-enhanced measurements must be assessed and their performance parameters specified. Additionally, novel validated methods should be developed for the fabrication of single-photon sources and to optimise the sources for highest purity and indistinguishability.

#### Objectives

The overall objective of the project is to develop bright entangled photon sources based on different application-oriented platforms and to exploit high-purity single-photon sources to demonstrate the quantum advantage achievable using these sources for specific measurements.

The specific objectives of the project are:

1. To assess single-photon sources and entangled-photon sources in traceable quantum-enhanced measurements (e.g. quantum calibration at the single-photon level, sub-shot noise measurements, quantum imaging, sub-diffraction imaging and quantum illumination), to overcome classical measurement limits (e.g. noise);
2. To specify the performance parameters of single-photon and entangled-photon sources required to carry out different quantum-enhanced measurements;
3. To develop novel validated methods for the fabrication of single-photon sources and to optimise the sources for highest purity ( $g^{(2)}(t=0)$  close to 0), brightness (photon rate  $> 5 \times 10^6$  photons per second) and indistinguishability (Hong-Ou-Mandel visibility  $> 95\%$ ), according to the performance parameters specified in Objective 2;
4. To develop the European metrology infrastructure required for the traceable characterisation of entangled-photon and single-photon sources, i.e. detectors (including photon-number resolving

detectors), amplifiers, as well as standardised quantum-optical setups for characterisation (in particular entanglement tomography);

5. To facilitate the take up of the technology and measurement infrastructure developed in the project by the EMN Quantum, measurement supply chain, standards developing organisations (e.g. CEN and ISO) and end users (in the fields of quantum technology and nano-photonics).

### **Progress beyond the state of the art**

This project will partially build on previous achievements of EMRP EXL02 SIQUTE and EMPIR 17FUN06 SIQUIST projects and will go beyond the state of the art by developing optimised single- and entangled-photon sources for quantum metrological application and by demonstrating the quantum advantage achieved by using these sources.

#### *Assessment of single-photon sources and entangled-photon sources in traceable quantum-enhanced measurements (objective 1)*

This project will investigate new fields of quantum metrology and quantum-enhanced measurements, which have not been investigated thus far. Much progress has been made on developing highly efficient and highly pure single-photon sources and entangled-photon sources with high indistinguishability in terms of high Hong-Ou-Mandel (HOM) visibility. The necessary and straightforward next step is to use these sources in optical quantum-enhanced metrology. In particular, the project will carry out a metrological assessment of single-photon sources and entangled-photon sources in traceable, quantum-enhanced measurements, e.g. quantum calibration at the single-photon level, sub-shot noise measurements, quantum imaging, sub-diffraction imaging and quantum illumination, to overcome classical measurement limits (e.g. noise); to date these measurements have never been performed by NMIs. To achieve these measurements, the project will design, develop and implement high performance single-photon and entangled-photon light sources for applications in enhanced quantum imaging with the target of beating the quantum noise limit. New paradigms of quantum measurements, such as weak values (i.e., the amount of correlation between measured and measuring system) and correlated measurements in Hong-Ou-Mandel (HOM) configurations will be explored as a tool for enhancing the resolution and sensitivity of microscopes and spectroscopic tools. Furthermore, the NMIs will address the completely new aspect of traceability in quantum enhanced measurements.

#### *Performance parameters of single-photon and entangled-photon sources (objective 2)*

This project will improve the performance parameters (e.g., purity and sub-shot noise quantification) for quantum light sources, which will significantly advance their application readiness. The main thrust of this project, however, is to explicitly determine the performance parameters, which are necessary for different quantum-enhanced measurements. This will be achieved by identifying both consolidated and recently proposed tools and physical observables, suitable for characterising the relevant “quantumness” properties, such as e.g., entanglement verification, sub-shot noise quantification, squeezing measurements for entangled sources; and purity, Glauber correlation functions, and photon flux variance measurements for single-photon sources.

#### *Development of novel validated methods for the fabrication of single-photon sources and optimisation of the sources (objective 3)*

Within this project, the partners will advance methods for the fabrication of single-photon sources and optimise the development of sources with the highest purity ( $g^{(2)}(t=0)$  close to 0 (i.e. target value  $< 0.001$  in the context of this project), brightness (photon rate  $> 5 \times 10^6$  photons per second) and indistinguishability (Hong-Ou-Mandel visibility  $> 95\%$ ). This will include fabrication of i) single-photon sources based on single defects in bulk diamond and on single molecules, and ii) single and entangled photon sources based on InGaAs/GaAs quantum dots (QDs).

#### *Development of a European metrology infrastructure for the traceable characterisation of entangled-photon and single-photon sources (objective 4)*

To meet the accelerating range of demands with respect to the upcoming new challenges in quantum technology, the metrological infrastructure in Europe needs to be continuously improved. This project will address the traceable characterisation of single-photon sources and especially of entangled-photon sources with respect to quantum technological applications, which has not yet been achieved. To carry out this traceable characterisation, low-photon flux detectors and corresponding amplifiers, photon-number resolving detectors will be developed, which will go beyond the state-of-the-art in terms of detection noise, measurement

uncertainty and stability.

## Results

### *Assessment of single-photon sources and entangled-photon sources in traceable quantum-enhanced measurements (objective 1)*

So far, the single photon purity of our fabricated telecom QDs was evaluated under the non-resonant, quasi-resonant (p-shell) and resonant excitation schemas. The experiments clearly show that p-shell and resonant excitation can be used to produce highly pure single photons with a two-photon probability as low as 0.005 at a record fibre coupled single photon count rate of nearly 14 Mcps.

A Hong-Ou-Mandel setup as basis for the planned experiments has been developed. It will be extended for cascaded HOM measurements to prepare the N00N states in the course of the project.

The characterisation of the mid infrared source emission spectrum and emission wavelengths of the entangled photons generated by spontaneous parametric down-conversion (SPDC) were carried out. In order to gain insight into the quantum advantage gained by excitation with MIR/NIR entangled beam, the microscope for conducting low flux illumination of polystyrene samples was set up. The SPDC source is being investigated with respect to its usefulness for two photon absorption measurements.

A Quantum Interference Imaging Mid-IR system for biological samples has been set up, and low flux illumination of polystyrene samples is being conducted to gain insight into the quantum advantage gained by excitation with a MIR/NIR entangled beam. Furthermore, the microscope is being tested for losses, collection rate, scanning speed and spatial resolution. Also, traceability is being investigated for the Quantum Interference Imaging Mid-IR system.

### *Performance parameters of single-photon and entangled-photon sources (objective 2)*

So far, a theoretical model for the emission dynamics of a solid-state single-photon source has been developed, including to continuous wave (CW) and pulsed excitation and emission. Second- and higher-order Glauber ( $g^{(n)}$ ) and Theta ( $\Theta^{(n)}$ ) parameters were identified as complementary suitable tools for the qualification of non-classical photon emission. The investigation in the properties and features of  $g^{(n)}$  and  $\Theta^{(n)}$  parameters for sources assessment has started. A theoretical model was developed to exploit a combination of both parameters for source mode reconstruction. The model is being validated against available experimental data sets.

Models of quantum dot based single-photon sources have been developed, parameter optimization in order to maximize Purcell factor and extraction has been performed. Different designs for vertical extraction have been compared in terms of their performance, designs for on-chip extraction have been optimized, designs with multiple, interfering resonances have been investigated. Modelling and optimization methods and scripts have been made available in an open access data publication (Zenodo 6565850, 2022).

The respective experimental key parameters of entangled photon sources that need to be measured to assess their non-classicality performances were collected. Among them are intrinsic parameters (generation probability / rate of two-photon state, decay times of biexciton and exciton, entanglement fidelity, fine-structure splitting, spin precession of biexcitonic/excitonic states, and background luminescence) as well as external parameters with noticeable impact on measurement results (photon-extraction efficiency of DUT, temporal resolution and efficiency of detectors, and dark counts / noise of detection system). The setup for quantum state tomography including two-photon resonant excitation is ready to use.

A SPDC model for different types of nonlinear crystal materials has been developed based on SPDC Green functions. The model considers experimental parameters, such as nonlinearity, phase-matching condition, losses, crystal type. The parameters to be calculated are flux rate, joint spectral amplitude, purity of the generated photon pairs.

A waveguide-integrated photonic cavity has been investigated using finite-element modelling. A surrogate model, trained with a machine learning algorithm (Bayesian optimization) has been developed and used to provide quantitative estimates of sensitivities to fabrication tolerances. A method for design optimization of emitter-to-fibre coupling, considering the impact of fabrication tolerances, has been developed and applied. Furthermore, models for studying impact of NV-centre placement and polarization in diamond nano-resonators on optical properties of emitted single photons have been developed and applied for analysing experimental results. Scripts and data for modelling these systems have been made available in an open access data publication.

A QD-based single-photon source was used to drive the biexciton-exciton emission cascade resonantly. It was shown that dephasing of the biexciton and exciton states have a severe impact on the visibility which is reduced below 50 % when the coherence time drops below 300 ps.

*Development of novel validated methods for the fabrication of single-photon sources and optimisation of the sources (objective 3)*

So far, single SnV centres were identified and tested: pure single photon emission was detected with a  $g^{(2)}(0)$ -value  $< 0.1$ , where the deviation from zero is entirely due to detector dark counts (noise limit  $g^{(2)}(0) = 0.093$ ). The emitters are almost strain-free, showing ideal ground state splitting and narrow resonance lines, when annealed at 2100°C and 8 GPa. The charge transition cycle of the SnV was identified and used to devise a method for charge state stabilization, i.e., by illumination with a laser in the blue spectral range. The charge state can be initialized and results in very narrow photoluminescence excitation lines for over 1 hour of experiment, i.e., the cumulative linewidth is 33 MHz at a Fourier-limit of 25 MHz. A Hong-Ou-Mandel interferometer for determining the single photon indistinguishability was set up with a classical visibility of  $> 99.5\%$ . Furthermore, high-purity single-crystal diamond samples were implanted with alternative ion species for the formation of different emitters: F, Cl, Fe, O, Mg, Mn, Ar, Ge and Pb at fluences between  $10^{10} \text{ cm}^{-2}$  -  $10^{15} \text{ cm}^{-2}$ , followed by post-implantation annealing (T: 900 °C - 1200 °C).

Single-mesa quantum dot structures and circular Bragg grating (CBG-) structures of high structural quality are fabricated that provide up to 450 kcounts/s under CW excitation in the 910 nm – 930 nm wavelength range. The metamorphic buffer design for the deposition of the InAs telecom QDs on GaAs substrates was modified in such a way that single-lambda cavities can be built around selected QDs. With this approach, samples were fabricated and evaluated which exhibit narrow linewidth of below 40  $\mu\text{eV}$ , a fine-structure splitting below 20  $\mu\text{eV}$  and a single photon purity as low as 0.005. With these structures and applying in-situ lithography, monolithic micro lenses and two-dimensional Bragg cavities (“Bulls-eye” cavities) were fabricated to enhance the brightness of the samples. A fibre coupled single photon rate up to nearly 14 Mcps at a single photon purity of 0.005 was achieved. This is at the moment worldwide the highest count rate value of a semiconductor quantum dot emitting in the telecom C-band.

Methods for modelling and optimizing nanophotonic structures incorporating point-like single-photon emitters have been developed. This includes circular Bragg gratings as high-Q resonators, and their coupling to fibres (Opt. Express 30, 15913, 2022 & Rickert, et al., arXiv:2212.04883). Data and parameterized modelling scripts have been made available as open-access data publications to allow for a straight-forward application to all material and source systems of this resonator class (Zenodo 7360516, 2022).

*Development of a European metrology infrastructure for the traceable characterisation of entangled-photon and single-photon sources (objective 4)*

The cryo-optic facility for the transition edge sensor (TES) has been set up successfully. Temperatures  $< 100 \text{ mK}$  were observed, as well as temperature-stabilized operation at 100 mK. Optical simulations have shown the possibility to reduce the TES reflectance losses embedding the TES in an optical cavity with an antireflection coating.

Two photonic integrated circuits (PICs), i.e., six cascaded 1:10 multimode interference splitters, have been designed, produced, packaged and tested for 850 nm. It has been shown that cascading the two PICs can also be made to achieve higher split ratios than  $1:10^6$  and down to  $1:10^{10}$ .

A new front-end electronic, based on switched integrator principle, has been designed, manufactured, and assembled. The new amplifier has dual channel capabilities that will be used to measure split ratios of up to  $1:10^6$  by measuring the photocurrent of two fiber coupled photodiodes simultaneously.

A low noise low photon flux fiber coupled detector has been constructed. Its responsivity has been calibrated in the spectral range from 580 nm to 970 nm traceable to the primary standard for optical power, the cryogenic radiometer. Preliminary tests performed on a fiber coupled quantum dot single photon source emitting at 933 nm showed measurement of photon flux as low as  $4 \times 10^5$  photons/s with noise at the level of 1 %.

For the portable semiconductor quantum dot system, the supporting system (19” housing including a Stirling cryocooler and fiber-coupled output) is ready to use. First tests were performed based on a free-space as well as a fiber-based configuration. Fiber-coupling efficiencies of up to 24 % in combination with vanishing multi-photon emission event was observed. For the room temperature portable source, suitable samples were identified and investigated showing count rates exceeding  $10^6$  counts/s.

## Impact

The project webpage was created (<https://seume.cmi.cz/>) and had about 3000 views so far. In the first 18 months of the project, 27 presentations were given at conferences, seminars and workshops. Additionally, 17 papers have been published in open access peer-reviewed journals, in addition, 5 further publications were submitted are already accepted and will be published soon. The consortium has further delivered 3 external and 2 internal training activities. The external trainings dealt with the fabrication and characterisation of quantum dots in semiconductor structures, with deterministic ion implantation topics and with the detection of entangled photons from spontaneous parametric down-conversion sources. These trainings were predominantly for the scientific community and had around 30, 25 and 1 (one-to-one training) attendee(s), respectively. The internal trainings had up to 5 attendees from the consortium and dealt with the handling, operation and measurements of single-photon sources.

### *Impact on industrial and other user communities*

This project will trigger and accelerate progress in the field of quantum technologies, due to the development of high-end, highly innovative quantum devices for use and application in science, quantum communication and quantum metrology. More specifically, the entangled and single-photon sources will impact the development of the associated measurement infrastructure for quantum enhanced metrology and for low-flux measurements, i.e., new and better amplifiers, new optical single photon excitation sources, which e.g., can be used in different fields where low optical fluxes need to be measured. The developed sources have the potential to become a commercial product useful for companies active in the field of quantum technology. They will also be very useful for educational purposes, both in academia and schools. This will be achieved through academic partners, and demonstrations at universities and schools. Furthermore, the lack of useful entangled and single-photon sources hinders the development of quantum technology fields such as quantum cryptography and quantum metrology; the developments within this project with respect to higher single photon flux rates, photon indistinguishability and entanglement, will remove these roadblocks. To support knowledge transfer to the industrial community, a workshop on the newly developed devices will be organised and held to which representatives of industry (both manufacturers of and users of photon sources) will be invited.

### *Impact on the metrology and scientific communities*

This project will have high impact on the metrology and scientific communities through the development of entangled and single-photons sources, based on single emitters. Specifically, the knowledge about entangled photon sources and their use at NMIs are scarce and therefore opportunities for applications with quantum light sources have not been exploited so far. It is anticipated that this project will create rather fast, new prospects and applications for quantum enhanced metrology, e.g., sub-shot noise metrology, quantum imaging of biological systems and quantum illumination. In addition, the project will further accelerate the development of a characterisation infrastructure for low flux measurements, e.g., in new amplifiers, as well as in practical fibre-coupled single-photon sources, where low optical fluxes need to be measured.

The project partners are members of the EMN Quantum and will present the project and its results at the meetings of this network and its corresponding network project. Also, an annual report is issued, in which all QT-related projects are listed. The results of this project will be disseminated to the scientific community predominantly via high impact publications on sources of single, indistinguishable and entangled photons, low flux and single-photon radiometry, low-noise amplifiers and excitation sources, therefore paving the way new and unforeseen research fields. To further support knowledge transfer, representatives of academic organisations and NMIs will be invited to a workshop that the project will organise.

Within the project, a software to simulate the performances of TES detectors were developed, available under <https://tes.inrim.it/>.

### *Impact on relevant standards*

New documentary standards based on the results of this project in the field of low-flux radiometry are expected, as a result of input to different standardisation bodies. The current *mise en pratique* for the candela allows the photon-number-based (and thus quantum-based) realisation of photometric and radiometric units. Furthermore, in the long term, a photon-based definition of the SI base quantity candela might be envisaged. Members of the consortium are active in the European Telecommunications Standards Institute (ETSI) and in CEN-CENELEC with respect to standardisation in (quantum) communication and in quantum technology, respectively. Results from this project will be implemented in documentary standards generated by these standards developing organisations. In the first half of the project, a new standard for quantum key distribution has been drafted, i.e., ETSI GS QKD 013 entitled "Characterization of assembled QKD transmitter and receiver

modules”, which will take up results from the project especially with respect to single-photon sources. In CEN/CENELEC, the Focus group on quantum technologies (FGQT) drafted a roadmap including sections on single- and entangled photon sources and on single-photon detectors. The work led to the formation of a Joint TC on quantum technologies (CEN/CENELEC JTC22).

#### *Longer-term economic, social and environmental impacts*

The project has the potential to have a significant economic impact on the European market, because it will strengthen Europe’s position in the field of quantum technologies. The sources and metrological infrastructure developed within the project may lead to their implementation in commercial products and the development of highly innovative commercial devices, respectively, and thus stimulate new high-tech jobs in Europe. This project may also have a long-term impact on the field of data safety, guaranteed by secure quantum communication, for which single-photon sources can be exploited.

#### **List of publications**

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This list is also available here: <https://www.euramet.org/repository/research-publications-repository-link/>

Project start date and duration:		1 June 2021, 36 months
Coordinator: Stefan Kück, PTB		Tel: +49 531 592 4010
Project website address: <a href="https://sequme.cmi.cz/">https://sequme.cmi.cz/</a>		E-mail: <a href="mailto:stefan.kueck@ptb.de">stefan.kueck@ptb.de</a>
Internal Funded Partners:	External Funded Partners:	Unfunded Partners:
1. PTB, Germany	8. CNR, Italy	18. INRIM, Italy
2. Aalto, Finland	9. CSIC, Spain	
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	15. UNITO, Italy	
	16. USTUTT, Germany	
	17. ZIB, Germany	
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