

Publishable Summary for 17FUN06 SIQUEST Single-photon sources as new quantum standards

Overview

The aim of this project is to develop new absolute standard radiation sources, which exploit the discrete and quantum nature of photons, and the necessary metrological infrastructure. These sources will be based on single-photon emitters with a calculable photon emission rate and high purity, i.e. with a very low multiple photon emission probability. Such sources hold promise as new quantum standards which will have a large number of applications, e.g. for use in the calibration of single-photon detectors, for the realisation of the SI base unit candela, for quantum random number generation, for quantum key distribution, for subshot noise metrology, for quantum enhanced metrology, and for photon-based quantum computation.

Need

The need for this project arises from the fact that single-photon sources will be necessary for applications in the field of quantum technologies (e.g. quantum computing, quantum communications, quantum metrology, quantum enhanced imaging and sensing), which are among the most relevant topics with respect to innovation and high technology in Europe and worldwide (Objectives 1 and 2). The use of single-photon sources in quantum computing will lead to much needed increases in computational power, which cannot be achieved by classical computers, and this will enable problems to be solved in physics, biology, chemistry and material science. However, the use of quantum computing will mean that the currently safe cryptographic protocols and procedures that are used in communication and data storage will become obsolete. Hence quantum cryptography, quantum key distribution and quantum communication will also be needed to maintain data safety and secure communication. Quantum metrology is needed to enable the classical shot-noise limit to be undercut and this will result in more precise measurements becoming possible. In addition, quantum imaging and quantum sensing need to be developed to enable high resolution measurements, as well as the possibility of carrying out imaging without the direct detection of photons, which hit the intended target (Objective 3). There is therefore a need to be prepared for when these new challenges, possibilities and technological innovations happen. The importance of this can also be seen from the European 1 billion € “Quantum Flagship” programme that started in 2018. This was triggered by the “Quantum Manifesto” and related European and national programmes, see e.g. the German “QUTEQA” initiative, the UK Quantum Technologies programme (“Quantum Hub”), QuantERA and the expected following programmes. This project is in line with the above mentioned programmes and it will strengthen the metrological basis for the innovations and developments to come in the field of quantum technologies, i.e. it will enable the National Metrology Institutes to meet future requirements. It will also perfectly complement and benefit the objectives of the quantum technology roadmap outlined in the context of the European Flagship initiative, by fulfilling the necessary metrology needs.

An ideal single-photon source emits one photon on demand, at a time chosen by the user with the emitted photons being indistinguishable from one another and having an adjustable repetition rate. Such a photon source would represent a new quantum standard with an extensive range of widely needed applications: for the calibration of single photon counter devices, for the realisation of the SI base unit candela, for quantum random number generation, for quantum key distribution, for photon based quantum computation and subshot noise metrology (Objectives 1, 2 and 3).

Furthermore, the characteristics of the single-photon sources need to be measured with traceability to national standards, therefore the necessary measurement infrastructure needs to be developed. This supporting measurement infrastructure for single photon metrology, i.e. new and better amplifiers and attenuators for the measurement of low optical fluxes, is needed in many fields of application (e.g. biology, chemistry, astronomy, etc.) and not just in the field of single photon metrology (Objective 4).

Also, this project’s results will lead to commercial products and to new standards, which need to be promoted (Objective 5).

Significant progress was made, in the previous EMRP JRP EXL02 SIQUTE, where e.g. for the first time a single-photon source was absolutely characterised metrologically. This was an important step towards the

realisation of a photon standard source. However, the photon fluxes of state-of-the-art technologies are too low ($< 1 \times 10^6$ photons per second) and the emission bandwidths are too broad (> 100 nm) for practical use. Therefore, single-photon sources are still far from being completely predictable, deterministic and indistinguishable. More development is needed to bring close-to-ideal single-photon sources into the NMIs to form the basis for future work in the field of quantum enhanced metrology (Objectives 1, 2 and 3).

Objectives

This project focuses on the development of single-photon sources as new quantum standards. The specific objectives are:

1. To develop single-photon sources as new quantum standards in the visible, near infrared and telecom wavelength range, based on optically and electrically driven impurity centres in nano and bulk diamonds, quantum dots in semiconductor structures and molecules having, simultaneously, photon rates $> 1 \times 10^6$ photons per second, emission bandwidths < 2 nm and high purity emission indicated by $g^{(2)}(t=0)$ values < 0.05 .
2. To assess new materials and concepts for single-photon sources, such as 2D materials (e.g. hexagonal boron nitride and thin transition metal dichalcogenides) and coupling designs to optimise the collection efficiency (e.g. micro-resonators, waveguides, optical antennas). To assess the impact of excitation schemes on the quantum optical properties of single-photon sources.
3. To establish sources of indistinguishable and entangled photons based on near infrared ($< 1 \mu\text{m}$) quantum dot single-photon sources with a visibility $> 90\%$ and novel sensing and measurement techniques based on these sources.
4. To develop metrology infrastructure for traceable single-photon source characterisation, i.e. detectors, amplifiers, single-photon spectroradiometers.
5. To promote the results, to trigger commercialisation of products, and to deliver input to standardisation organisations.

Progress beyond the state of the art

In recent years, there has been significant progress in the field of single-photon sources and their use in metrology. This also includes the progress made by the previous EMRP JRP EXL02 SIQUTE. In this previous project, leading European research institutes and NMIs worked closely together to bring quantum technology at the single-photon level into the NMIs. Within EMRP JRP EXL02 SIQUTE, the first single-photon source absolutely calibrated with respect to its total optical radiant flux and spectral power distribution, traceable to the corresponding national standards via an unbroken traceability chain, was realised along with the pump laser providing optical excitation for the single-photon sources at varying repetition rates. The total photon flux rates were between 190 000 photons per second and 260 000 photons per second, respectively and the single-photon emission purity was indicated by a $g^{(2)}(0)$ value, which was between 0.10 and 0.23, depending on the excitation power. Although this might be considered as a huge step towards the realisation of a new photon standard source, the photon fluxes are still too low ($< 1 \times 10^6$ photons per second), the emission bandwidth is too broad (> 100 nm) and the purity is still insufficient ($g^{(2)}(t=0) > 0.1$) for real practical use.

Photonic structures are required in order to enhance the photon collection efficiencies. Typical collection efficiencies from solid state materials (diamond, quantum dots) using high numerical aperture objectives are within the few percent range due to the high material refractive index and the resulting large fraction of light captured by total internal reflection. Photonic structures typically employed for enhancing the photon collection efficiency are solid immersion lenses and waveguiding structures (e.g. nanopillars), resulting in typically 30 % collection efficiency. Even larger efficiencies (50 % - 100 %) are expected for schemes based on coupling to micro-resonators or planar optical antenna and micro-lens structures where the latter combines high extraction efficiency and ease of use in NMI applications.

Results

1. *To develop single photon sources as new quantum standards in the visible, near infrared and telecom wavelength range, based on optically and electrically driven impurity centres in nano and bulk diamonds, quantum dots in semiconductor structures and molecules having, simultaneously, photon rates $> 1 \times 10^6$ photons per second, emission bandwidths < 2 nm and high purity emission indicated by $g^{(2)}(t=0)$ values < 0.05 .*

Type-IIa diamond bulk crystals have been implanted with the following ions, which are supposed to be an efficient single-photon emitter, these are: Sn, Pb, He, F, Cl, Er. The fabricated samples were then spectroscopically characterised at room temperature, where the following main emission lines were observed: 535 nm and 560 nm for He, 621 nm for Sn, 538 nm and 552 nm for Pb, 600 nm to 800 nm for F, thus proving the successful implantation. For Cl and Er, photoluminescence in the visible range was not observed. Diamond samples implanted with Sn and Pb demonstrated single-photon emission at room temperature under 520 nm and 532 nm laser excitation. The SnV centre exhibited $g^2(0)$ values down to 0.29 ± 0.02 with a saturation photon rate of approx. 1.4×10^6 photons/s. Single-photon purity can be enhanced to a $g^2(0)$ down to 0.05 on the cost of saturation photon rate, which is in this case only 0.15×10^6 photons/s. The PbV centre exhibited a $g^2(0)$ value < 0.5 after background noise correction, which originated from first and second-order Raman scattering of diamond. Preliminary experiments also indicated anti-bunching behaviour from F-related centres.

First single-photon sources based on InGaAs QDs in micro-structured GaAs were fabricated and used for a relative calibration of two Silicon single-photon avalanche diode detectors. The photon rate measured for a specific QD was around 3×10^5 photons/s with a $g^2(0)$ -value of 0.23 at a wavelength of (922.37 ± 0.02) nm. The measurement uncertainty for this relative calibration is about 0.7 %. To improve the photon flux, structures with so-called “Bull’s eyes cavities” (BECs) are being designed as well as structures with a bottom gold mirror.

Several single-photon emitters based on a single molecule (dibenzoterrylene in anthracene, DBT:Ac) were designed and fabricated. In order to efficiently collect the emission, a planar antenna design which is simple and cheap to fabricate, and which provides hundreds of source candidates per fabrication run, was developed. A bright source was metrologically characterised, the parameters are, simultaneously: photon flux up to 1.4×10^6 photons/s at the wavelength of (785.6 ± 0.1) nm with a $g^2(0)$ -value < 0.1 and a spectral bandwidth (FWHM) < 2 nm. The photon flux was measured with a traceable low-noise analogue reference Si detector. This source was used to directly calibrate a Silicon single photon avalanche diode detector against a Silicon photodiode, which is in turn traceable to the cryogenic radiometer.

2. *To assess new materials and concepts for single-photon sources, such as 2D materials (e.g. hexagonal boron nitride and thin transition metal dichalcogenides) and coupling designs to optimise the collection efficiency (e.g. microresonators, waveguides, optical antennas). To assess the impact of excitation schemes on the quantum optical properties of single-photon sources.*

Thus far, room temperature and low temperature (~ 70 K) spectral fluorescence mapping of hexagonal boron nitride (hBN) flakes has started. A confocal system capable of Hanbury Brown and Twiss interferometry, which is suitable for use with hBN samples, is fully functional. These samples have been spectrally characterised using photoluminescence spectroscopy at room and low temperature, as well as atomic force microscopy measurements. Approx. 10 % of the emitters are stable under extended optical pumping. Two samples have been produced, each with around 10 stable emitters, which will now be further investigated with respect to single-photon emission purity.

With respect to an optimisation of the coupling efficiency, spin-coating, micro-infiltration and drop-casting approaches for depositing nanocrystals were evaluated and the trade-off between cleanliness and success rate makes micro-infiltration more effective than the drop-casting technique.

The impact of different excitation schemes on the quantum optical properties of single-photon sources is being investigated. For InGaAs QDs (emitting around 930 nm) in a GaAs matrix, optical excitation via the GaAs matrix, the wetting layer and the p-shell are ready-to-use for HBT and HOM measurements, a setup for strict resonant excitation of microlenses is under development. For the 1.5 μm region, similar investigations are currently being carried out with InGaAs QDs in GaAs. A strong linewidth decrease is observed from above-band ($\Delta\lambda \approx 11$ GHz) via phonon-assisted ($\Delta\lambda \approx 7.1$ GHz) to direct resonant excitation ($\Delta\lambda \approx 3.1$ GHz). Furthermore, two-photon excitation was applied and high purity ($g^{(2)}(0) = 0.072 \pm 0.104$) and highly indistinguishable ($V = 0.894 \pm 0.109$) single photons were observed. Electro-optical pumping schemes have been implemented which allow the driving of the emission and the tuning of the wavelength of a single quantum dot by electrical means in a monolithic device. A single photon rate of ~ 0.5 MHz in the first lens has been attained with $g^{(2)}(0) = 0.093 \pm 0.123$ at injection currents < 10 μA .

3. *To establish sources of indistinguishable and entangled photons based on near infrared (< 1 μm) quantum dot single-photon sources with a visibility > 90 % and novel sensing and measurement techniques based on these sources.*

The indistinguishability of photons emitted by a single molecule has been demonstrated. Setup upgrades to enhance emission purity and spectral filtering, as well to refine indistinguishability measurement are in progress.

The setup for ODMR measurements was upgraded resulting in an increase of magnetic field sensitivity of ODMR-based magnetometry protocols ultimately for applications of sensing in biological systems. Specifically, an optimised diamond sample with a very narrow layer of NV centres is being used and the possibility to drive simultaneously all three hyperfine peaks corresponding to one spin orientation was implemented. A new state-of-the-art experimental setup for the implementation of single-photon confocal photoluminescence and ODMR measurements down to 4 K temperature was established.

The new paradigm dubbed the genetic quantum measurement, specifically the possibility to exploit this new approach with quantum dots and colour centres in diamond, has been investigated to understand if there is some peculiar observable of these quantum systems that can take advantage of this novel technique.

4. To develop metrology infrastructure for traceable single-photon source characterisation, i.e. detectors, amplifiers, single-photon spectroradiometers.

A cooled Predictable Quantum Efficient Detector (PQED) in a liquid nitrogen operated cryostat for use in performing SI traceable measurements of low power sources was constructed and characterised. The device uses electronics developed within the project and it is capable of detecting 1 000 000 photons/s with an uncertainty of $< 1\%$ in the wavelength range between 650 nm and 750 nm. New absorber structures for the Inductive Superconducting Transition Edge Detector (ISTED) were modelled and designed. Currently, each 5.5 mm chip contains 15 ISTED devices with absorber structures of different sizes (15 mm, 20 mm and 25 mm) and also a test pad for measuring the transition temperature T_c . It was observed, that a variation of the Nb absorber thickness by 2 nm provides a small change in T_c of ~ 0.4 K, whereas doping the Nb thin film with Si from 97.1 % to 95.4 % Nb changes T_c by 1.2 K.

A portable single-photon source was designed and constructed and currently undergoes compaction to reduce the size to approx. only 40 cm x 27.5 cm x 20 cm. This source delivered with a molecule emitter (terrylene in p-terphenyl) approx. 2×10^6 photons/s at room temperature. The $g^{(2)}(0)$ value was approx. 0.2 and the emission covered the spectral region between 550 nm and 700 nm with emission peaks at approx. 580 nm, 630 nm and 680 nm.

A novel criterium for detecting non-classical behaviour in the fluorescence emission of ensembles of single-photon emitters was investigated. In particular, the so-called Filip's θ function was identified as a useful parameter, which is defined as the ratio between the probability of detecting zero photons in coincidence at the output of an n -dimensional Hanbury Brown & Twiss interferometer, divided by the product of the no-click probabilities at the individual channels. Experimental data clearly demonstrate two advantages of the tested non-classicality criterion with respect to the well-known $g^{(2)}$ -characterisation, i.e. the resilience to Poissonian noise and an increasing deviation from classicality at an increasing number of single photon emitters. A feasibility study on the exploitation of the Filip's θ function, for optical modes reconstruction was performed. It is expected that exploiting higher order Filip's θ functions will allow mode reconstructions with improved accuracy. First simulations have been performed, which indeed shows promising evidence.

First results were achieved using techniques for measuring the $g^{(2)}(t=0)$ value. A pilot study on the characterisation with respect to the $g^{(2)}(t=0)$ value of a pulsed-pumped single-photon source, based on a NV centre in nanodiamond, was performed by INRIM, NPL and PTB. The main result of this study was the development of a standardised measurement technique as well as an uncertainty estimation procedure. The validity of the technique (system-independent and unaffected by the non-ideality of the apparatus) was demonstrated by the results which yielded estimated values of $g^{(2)}(t=0)$ that were compatible within the uncertainty ($k=2$) for all of the participants. This study will greatly benefit the single-photon metrology community, as well as rapidly-growing quantum-technology-related industries.

A strategy was developed to properly evaluate the multi-photon component of a CW light source when applied to a low-noise prototype of a fibre. This heralded a single photon source operating at 1550 nm. This source will be particularly suited for metrological and quantum-communication-related purposes and it will be adaptable to a large variety of detectors and other devices. The results of the whole measurement campaign, carried out with different measurement setups and data collection methodologies, are all in agreement within the experimental uncertainties, even with a coverage factor of $k=1$. This strategy may enable the standardisation of the characterisation of single-photon sources, a task of the utmost relevance for the current and future metrology of quantum technologies.

5. To promote the results, to trigger commercialisation of products, and to deliver input to standardisation organisations.

Currently, the project promotes the results via publications (16 open access publications in peer reviewed journals) and in conferences (> 90 conference contributions). It triggers special sessions on single-photon

sources and on single-photon metrology in workshops and conferences. The project also delivers input to the Industry Specification Group on Quantum Key Distribution for Users of the European Telecommunications Standards Institute (ETSI ISG-QKD).

Impact

Thus far, there are 16 peer reviewed, open access scientific publications and > 90 conference presentations and posters within the SIQUEST project. Furthermore, there are 7 standards and regulatory activities, all within the European Telecommunications Standards Institute (ETSI).

This project will create impact for end-users (e.g. manufacturers of quantum communication systems) by giving them access to new and improved single-photon sources. High technology companies working in the field of quantum technologies will be able to accelerate innovation. National Metrology Institutes will use these as new standard sources for radiometry and photometry, enabling them to provide the measurements required to certify new quantum technologies based on the discrete and quantum properties of photons. Schools and academia will benefit from the development of easy-to-use and affordable devices for education in quantum technologies.

Impact on industrial and other user communities

The technological developments within this project will trigger and speed up innovation 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 single-photon sources and the supporting measurement infrastructure will impact on the development of measurement infrastructure for low photon flux measurements, i.e. new and better amplifiers and attenuators, new optical single photon excitation sources, which will be used in different fields where low optical fluxes need to be measured, e.g. quantum communication, but also in medicine, biology, astronomy and research in general. The developed (standard) single-photon sources have the potential to become a commercial product useful for companies active in the fields of quantum technology, e.g. the telecommunication industry, metrology equipment companies, and academic spin-off companies. They will also be very useful for educational purposes, both in academia and schools, as cheap and easy-to-handle single-photon sources will be available. Up to now, the lack of useful single-photon sources hinders the development of quantum technology fields such as quantum cryptography and quantum metrology. The development of single-photon sources with higher flux rates and indistinguishability, will lead to the removal of these road blocks.

Impact on the metrology and scientific communities

The development of new standard sources based on single-photon emitters will create impact for the realisation of optical radiant flux scales in the low-photon-flux region and it will be the basis for the definition of the optical radiant flux in terms of photon rate, i.e. the countable number of photons over time, with selectable emission rates. As this field develops, it is foreseen that quantum standards, based on counting photons, will also enter the metrological area of radiometry and photometry. The results will therefore impact on metrology committees like the Consultative Committee for Photometry and Radiometry (CCPR) or the technical committees of the regional metrology organisations (EURAMET, COOMET), where the partners are active. The SI base unit for luminous intensity, covering the areas of photometry and radiometry, is the candela, which is currently realised in purely classical ways, i.e. by incandescent standard lamps carrying and maintaining the luminous intensity scale or by photometers, which are radiometrically calibrated in a way which is traceable to the primary standard for optical radiant flux. However, in the current *mise en pratique* for the candela, the possibility for a photon-number-based realisation is explicitly formulated. Therefore, there will be a direct impact on the *mise en pratique* and thus on the metrology community in general. Furthermore, these sources would be ideal for the calibration of single-photon detectors, especially if a multiple wavelength source is available. Also, the sources developed and characterised in this project can emit entangled photons, thus opening new fields in quantum metrology. The scientific results of this project will lead to high impact publications, which will act as further multipliers for research fields which are not even anticipated yet.

Impact on relevant standards

The development of a standard single-photon source will impact the National Metrology Institutes by giving them access to photon sources that are useful for a variety of applications, as mentioned above. Furthermore, new documentary standards based on the results of this project in the field of low-flux radiometry are expected, such as ETSI standards on quantum communication and quantum key distribution, and the *mise en pratique* for the candela. As mentioned above, the current *mise en pratique* for the candela allows the photon number based (and thus quantum based) realisation of photometric and radiometric units.

Longer-term economic, social and environmental impacts

This project will have a significant economic impact on the European market, because it will strengthen the European position in the field of quantum technologies. The sources and metrological infrastructure developed within the project may give rise to commercialisation and thus lead to employment in high technology areas, due to the development of highly innovative devices for commercialisation by companies followed by their implementation in commercial products. Another aspect to be considered is the field of data safety, guaranteed by secure quantum communication, for which single-photon sources can be exploited. So, in the longer-term perspective, end-users from the fields stated above will benefit from the outcomes of this project.

List of publications

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1 PTB, Germany	8 CNR, Italy	
2 Aalto, Finland	9 CSIC, Spain	
3 CMI, Czech Republic	10 FAU, Germany	
4 INRIM, Italy	11 INFN, Italy	
5 Metroserf, Estonia	12 TUB, Germany	
6 NPL, United Kingdom	13 UdS, Germany	
7 VTT, Finland	14 UNITO, Italy	
	15 USTUTT, Germany	
RMG: -		