Publishable Summary for 17FUN06 SIQUST
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 can be used as new quantum standards for 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

Single-photon sources are needed for applications in quantum technologies such as quantum computing, quantum communications, quantum metrology, quantum enhanced imaging and sensing), which are amongst the most relevant topics with respect to innovation and technology in Europe. This importance can be seen in the EC's 1 billion € "Quantum Flagship" programme that started in 2018. This programme was triggered by the "Quantum Manifesto" and related European and national programmes, e.g. the German "QUTEGA" initiative, the UK Quantum Technologies programme ("Quantum Hub"), and QuantERA.

The use of single-photon sources in quantum computing should lead to much needed increases in computational power, however, the same use of quantum computing will also mean that the currently safe cryptographic procedures that are used in communication and data storage will become obsolete. Thus, quantum cryptography, quantum key distribution and quantum communication need to be developed to maintain data safety and security. Further to this, quantum metrology is needed to enable the classical shot-noise limit to be undercut in order to support an increased precision in measurements. In addition, quantum imaging and quantum sensing need to be developed to enable high resolution measurements, and the ability to perform imaging without the direct detection of photons.

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 is greatly needed as it 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.

Furthermore, the characteristics of the single-photon sources need to be measured with traceability to national standards, therefore the necessary measurement infrastructure for traceable single-photon source characterisation needs to be developed.

Significant progress was made in the previous EMRP project EXL02 SIQUTE, where e.g. for the first time a single-photon source was metrologically characterised. 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 of this single-photon source. Therefore, further work is needed to develop close-to-ideal single-photon sources

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 (QD) in semiconductor structures and molecules having,

simultaneously, photon rates > 1 × 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 µm) QD 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 project EXL02 SIQUTE. In this previous project, 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. This was done along with the development of 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 × 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.

To go beyond the current state of the art, photonic structures are required in order to enhance the photon collection efficiencies. Currently, typical collection efficiencies from solid state materials (diamond, QD) 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 used for enhancing the photon collection efficiency are solid immersion lenses and waveguiding structures (e.g. nanopillars), resulting in approximately 30 % collection efficiency. However, even larger efficiencies (50 - 100 %) can be expected for schemes based on coupling to micro-resonators or planar optical antenna and micro-lens structures and should provide beyond the state of the art high extraction efficiency combined with 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, QD in semiconductor structures and molecules having, simultaneously, photon rates > 1 × 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 were implanted with ions (i.e. Sn, Pb, He, F, Cl, Er) that are supposed to be an efficient single-photon emitter. The samples were then spectroscopically characterised at room temperature in order to demonstrate the successful implantation of the ions.

Diamond samples implanted with Sn and Pb demonstrated single-photon emission at room temperature under 520 nm and 532 nm laser excitation. The Sn Vacancy (SnV) centre of the samples implanted with Sn exhibited single-photon purity ($g^2(0)$ values) down to 0.29 ± 0.02 with a saturation photon rate of approx. 1.4 x 10^6 photons/s. Single-photon purity could be enhanced to a $g^2(0)$ down to 0.05 but this was at the expense of the saturation photon rate, which was only 0.15 x 10^6 photons/s. The Pb Vacancy (PbV) centre of the samples implanted with Pb exhibited a single-photon purity $g^2(0)$ 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.

A diamond sample with Ge Vacancy (GeV) colour centres was characterised in terms of its fluorescence mapping, photon rate, single-photon purity ($g^2(0)$-value) and spectral characteristics. Three GeV centres were characterised and their spectral line emission (zero phonon line) was located at 604 nm. The single-photon purity obtained was approx. 0.5, but the $g^2(0)$ value was strongly affected by the high background emission of the samples. The maximal count rate measured with a single-photon avalanche diode (SPAD) detector was approx. 120000 count/s (background = 30 kilo counts per second; kcps), thus further background reduction of the samples is needed.

In order to better understand the emission characteristics of impurity centres in nanodiamant, back focal plane imaging of the emission of a nitrogen-vacancy (NV)-centre at a dielectric interface was compared to theoretical models for the calculation of the angular emission patterns. The orientation of the NV-centers was obtained from measurements of the fluorescence intensity independently of the polarisation angle of the linearly polarised excitation laser. The collection efficiency was calculated to be higher than 80 % using the model of the angular emission of the NV-centres. This method is also applicable to other emitters like Si-vacancy centres, tin-vacancy centres and hexagonal boron-nitride (hBN).

First single-photon sources based on indium gallium arsenide (InGaAs) QDs in micro-structured GaAs were fabricated and used for a relative calibration of two Si SPAD detectors. The photon rate measured for a specific QD was approx. 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 was about 0.7 %. Finally, Allan deviation analysis was performed giving an optimal averaging time of 92 s for the photon flux. Improvements were made to increase the collection efficiency of the QD sample, i.e. the setup transmission was improved by a factor of 2 and the QD (Mesa-structures) were characterised again. The maximal photon rate measured after the improvements was 2.5 Million photon/s with $g^2(0)$ value < 0.23. For lower excitation values (approx. 10 % of the excitation power), a photon rate of 307 kcps was obtained with a $g^2(0)$ value of 0.09.

Several single-photon emitters based on a single molecule (dibenzoterrylene in anthracene, DBT:Ac) were designed and fabricated and a planar antenna was also designed to efficiently collect their emissions. A bright source from the DBT:Ac emitters was characterised using a traceable low-noise analogue reference Si detector: it had 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 (Full Width Half Maximum) < 2 nm. The bright source was also used to calibrate a Si SPAD detector against a Si photodiode, which in turn is traceable to a cryogenic radiometer.

2. *To assess new materials and concepts for single-photon sources, such as 2D materials and coupling designs to optimise the collection efficiency. To assess the impact of excitation schemes on the quantum optical properties of single-photon sources.*

hBN samples have been spectrally characterised using Atomic Force Microscopy (AFM) and Photoluminescence (PL) spectroscopy at both room and low temperatures. The results showed that 80 % of the emitters quench rapidly, while 10 % survived extended optical pumping. Two hBN samples were produced, each with around 10 stable emitters; one was sent to PTB, and one retained at NPL, where one of the sample's stable emitters exhibited $0.5 < g^2(0) < 1$.

A focused ion beam (FIB) method for generating hBN defects was explored, including attempts to use patterned substrates and Transmission Electron Microscopy (TEM) grids to ameliorate FIB damaged substrate fluorescence. A re-exfoliation technique was developed which allowed the FIB exposed flakes to be studied without the overwhelming background fluorescence of the FIB damaged substrate. Exposure to FIB doses was shown to increase fluorescence in exfoliated hBN crystals, but so far this has lacked any hallmarks of single-photon emission.

The optimisation of methods for coupling efficiency, spin-coating, micro-infiltration and drop-casting for depositing DBT:Ac nanocrystals were investigated, and results showed that the trade-off between cleanliness and success rate makes micro-infiltration more effective than the drop-casting technique. The first evidence of fluorescence coupling into the waveguide mode inside a cryostat has also been reported by the consortium.

The effect of different excitation schemes on the quantum optical properties of single-photon sources is being investigated. 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 be used for Hanbury Brown-Twiss (HBT) and Hong–Ou–Mandel (HOM) measurements, and a new setup for strict resonant excitation is under development. Two-photon resonant excitation was successfully performed on QD-micro-lenses and fidelities of up to 80 % were obtained, however non-resonant excitation was found unsuitable for reaching similar values.

Similar investigations are currently being carried out with InGaAs QDs in GaAs in the 1.5 µm region, and a strong linewidth decrease has been 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). Pulsed excitation under different excitation conditions has also been carried out and high pure triggered single photon emission at 1550 nm was achieved under different excitation emission (g(2)(0) = 0.023 ± 0.019 for resonant excitation).

Finally, electro-optical pumping schemes have been developed that allow the driving of the emission and the tuning of the wavelength of a single QD using electrical means in a monolithic device. A single photon rate of ~0.5 MHz in the first lens was attained with $g^{(2)}(0) = 0.093 \pm 0.123$ at injection currents <10 µA. The next step is to fabricate new devices with new epitaxial designs in order to improve wavelength tuneability.

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

Indistinguishability of triggered single photon emission from InGaAs quantum dots in InAs in the 1.5 µm spectral region was measured, i.e. 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. For single molecules, indistinguishability of emitted photons was also demonstrated and setup upgrades to enhance emission purity and spectral filtering, as well to refine indistinguishability measurement, are in progress.

The impact of pulsed non-resonant and pulsed quasi-resonant excitation on photon indistinguishability of QD emission in the 930 nm wavelength range was investigated and it was found that resonant excitation is required to obtain visibilities that exceed 90 %. For the generation of entangled photon pairs, two-photon excitation as the spectral extinction of the exciting laser light was found to be more effective than single-photon resonant excitation.

Optical Detection of Magnetic Resonance (ODMR) sensing protocols were developed with NV centres showing 40 nT/Hz^1/2 magnetic sensitivity in continuous excitation (70 nT/Hz^½ in biocompatible conditions). This is in line with the current state of the art for sensing volume, thus further work to increase the sensitivity and pulsed measurements is on-going. So far Rabi oscillations and Ramsey fringes have been observed and the first preliminary results of pulsed ODMR have shown improved linewidth of the resonance dip.

The project has investigated a new paradigm dubbed the genetic quantum measurement. Specifically, the use of the genetic quantum measurement approach with QD and colour centres in diamond (objective 1) was studied in order to determine whether there is a specific observable in these quantum systems that can take advantage of the new genetic quantum measurement approach. The results showed that the polarisation degree of freedom is suitable and that the most suitable source for the polarisation degree of freedom is the single-photon source, since it naturally emits photons in a defined linearly polarised state. Therefore, such a single-photon source has been used to investigate different regimes in terms of interaction strengths and polarisation states.

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 was constructed and characterised for traceable measurements of low power sources. The PQED device uses electronics developed by the project and it is capable of detecting 1,000,000 photons/s with an uncertainty of < 1 % in the wavelength range 650 - 750 nm.

New absorber structures for an Inductive Superconducting Transition Edge Detector (ISTED) have been designed. Each 5.5 mm chip contains 15 ISTED devices with absorber structures of different sizes (i.e. 15, 20 and 25 mm) and a test pad for measuring the transition temperature T_c. Results have shown that a variation of Nb absorber thickness by 2 nm produces a change in T_c of ~ 0.4 K, whereas doping the Nb absorber structure with Si (i.e. 97.1 - 95.4 % Nb) changes T_C by 1.2 K.

A portable single-photon source was constructed and is currently undergoing further work to reduce the size to approx. 40 cm x 27.5 cm x 20 cm (i.e. to make it more portable). The portable single-photon source delivered a molecule emitter (terrylene in p-terphenyl) of approx. 2 x 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 and 700 nm with emission peaks at approx. 580 nm, 630 nm and 680 nm.

The project has also investigated a novel criterium for detecting non-classical behaviour in the fluorescence emission of ensembles of single-photon emitters. The Filip's θ function was identified as a useful parameter and is defined as the ratio between the probability of detecting zero photons in coincidence at the output of an *n*-dimensional HBT interferometer, divided by the product of the no-click probabilities at the individual channels. Experimental data demonstrated two advantages of the Filip's θ function over well-known $g^{(2)}$-characterisation; (1) the resilience to Poissonian noise and (2) an increasing deviation from classicality at an increasing number of single photon emitters. A feasibility study on the use of the Filip's θ function, for optical modes reconstruction is being performed and it is expected that the use of higher order Filip's θ functions will allow mode reconstructions with improved accuracy. Indeed, the first simulations have shown promising results.

A pilot study was performed by INRIM, NPL and PTB on the characterisation of the $g^{(2)}(t = 0)$ value of a pulsed-pumped single-photon source, based on a NV centre in nanodiamond (from objective 1). From the results a standardised measurement technique for the characterisation of the $g^{(2)}(t = 0)$ and an uncertainty estimation procedure were developed. The validity of the measurement technique (i.e. system and/or apparatus-independence) was demonstrated in the pilot study and the results gave estimated values of $g^{(2)}$ $(t = 0)$ that were compatible within the uncertainty ($k = 2$).

Finally, a strategy was developed to evaluate the multi-photon component of a continuous wave (CW) light source when applied to a low-noise prototype of a fibre. The strategy produced a single photon source operating at 1550 nm. The results of a measurement campaign using the strategy but carried out with different measurement setups and data collection methods, showed agreement within the experimental uncertainties (i.e. a coverage factor of $k = 1$). These positive results suggest that this strategy may enable the standardisation of the characterisation of single-photon sources.

Impact

This project will benefit end-users such as manufacturers of quantum communication systems, by giving them access to new and improved single-photon sources. NMIs can also use them as new standard sources for radiometry and photometry, enabling the NMIs to provide the measurements required to certify new quantum technologies based on the discrete and quantum properties of photons.

So far, the project has produced 27 peer reviewed, open access scientific publications and 113 conference presentations and posters, as well as 3 press releases. In addition, the project website is available at https://www.siqust.eu/

Impact on industrial and other user communities

The technological developments within this project will support innovation in the field of quantum technologies, through the development of quantum devices for use in quantum communication and quantum metrology. More specifically, the single-photon sources (objectives 1, 2 & 3) and the supporting measurement infrastructure (objective 4) will help to develop the necessary measurement infrastructure for low photon flux measurements, i.e. new and better amplifiers and attenuators, new optical single photon excitation sources, which can be used in quantum communication, medicine, biology, and astronomy. The developed (standard) single-photon sources also have the potential to become a commercial product useful for companies active in the fields of quantum technology. This is particularly important as currently; the lack of useful single-photon sources hinders the development of quantum technology fields such as quantum cryptography and quantum metrology.

The project has compiled written training material for external audiences on “Ion-beam Engineering of Materials for Quantum Technologies”, which will soon be available online at the International Atomic Energy Agency (IAEA) E-portal. The IAEA fosters international collaboration to help close currently existing gaps in physics, technology and regulation and the project's information in the IAEA portal is freely available to end users such as those from industry.

In addition, press releases from the project have highlighted (i) the discovery of new properties of colour centers in diamond by partner UNITO researchers, (ii) the quantum revolution enabled by diamond defects and (iii) Metrology Common ground https://www.nature.com/articles/s41567-019-0432-9.pdf. The latter article discussed the development of the first standardised technique to characterise single-photon sources and how to provide common uncertainty estimation procedures to estimate the second-order correlation function, a parameter commonly used to describe single-photon sources.

Impact on the metrology and scientific communities

The project's development of new standard sources based on single-photon emitters (objectives 1, 2 & 3) will support the realisation of optical radiant flux scales in the low-photon-flux region. It can also be used as 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 expected that quantum standards, based on counting photons, will be used in radiometry and photometry and hence will impact metrology committees such as the Consultative Committee for Photometry and Radiometry (CCPR), EURAMET and COOMET.

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. This classical method involves either incandescent standard lamps carrying and maintaining the luminous intensity scale or photometers, which are radiometrically calibrated in a way which is traceable to the primary standard for optical radiant flux. In contrast, in the current *mise en pratique* for the candela, the possibility for a photon-number-based realisation is explicitly formulated. Therefore, this project will contribute to the improvement of the *mise en practique* for the candela.

This project's standard sources will also emit entangled photons (objective 3), thus supporting development in new fields in quantum metrology.

Currently, 1 PhD and 6 master theses were completed within the project, demonstrating transfer of knowledge to the next generation of scientists and metrologists.

Five training courses between the consortium have also taken place to transfer knowledge between project partners. These training course topics included (i) single-photon sources based on quantum dots at 1550 nm (ii) single-photon sources based on quantum dots at 930 nm, (iii) single-photon sources based on single molecules, (iv) the installation of the portable single-photon source, and (v) nitrogen-vacancy-assisted magnetic/electric field sensing. Further to this the project has provided 3 training courses to the external scientific community on, (i) single-photon sources based on defects in solid state, (ii) optical quantum metrology sensing and imaging in general and (iii) quantum encryption.

Finally, the project has engaged with the scientific community by establishing 3 new collaborations, with Ruđer Bošković Institute (Croatia), the University of Leipzig (Germany) and the CNR-ISC (Italy).

Impact on relevant standards

The results of this project will provide input to new documentary standards in the field of low-flux radiometry, 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 practique* for the candela allows the photon number based (and thus quantum based) realisation of photometric and radiometric units.

Recently, the project provided input to the CEN/CENELEC Focus Group on Quantum Technologies, in establishing a roadmap for standardisation in the field of quantum technologies. Further to this, the project has provided input to ETSI's Industry Specification Group on Quantum Key Distribution (ISG-QKD), where it contributed to the draft of ETSI GS QKD 013 "Characterisation of Optical Output of QKD transmitter modules", the DGS/QKD-0010_ISTrojan (GS QKD 010) "Implementation security against Trojan horse" and to an internal ISG Working Document to support the development of ETSI GS QKD 00.

Longer-term economic, social and environmental impacts

This project will have significant impact on the European market, by strengthening Europe's position in the field of quantum technologies. The single-photon sources and metrological infrastructure for traceable single-photon source characterisation developed within the project should support commercialisation and thus lead to better employment in high technology areas, due to the development of highly innovative devices for use in quantum technology fields (e.g. quantum cryptography), quantum communication, medicine, biology, and astronomy. In particular the field of data safety, guaranteed by secure quantum communication, will become ever more increasing important in all aspects of life for European citizens.

List of publications

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<table>
<tr><td colspan="2">Project start date and duration:</td><td>01 June 2018, 42 months</td></tr>
<tr><td colspan="3">Coordinator: Stefan Kück, PTB   Tel: +49 531 5924010   E-mail: stefan.kueck@ptb.de
Project website address: https://www.siqust.eu/</td></tr>
<tr><td>Internal Funded Partners:
1 PTB, Germany
2 Aalto, Finland
3 CMI, Czech Republic
4 INRIM, Italy
5 Metrosert, Estonia
6 NPL, United Kingdom
7 VTT, Finland</td><td>External Funded Partners:
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14 UNITO, Italy
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<tr><td colspan="3">RMG: -</td></tr>
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