The absolutely characterized nitrogen vacancy center-based single-photon source – measurement uncertainty of photon flux and angular emission properties

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Abstract. Recently, a single-photon source based on the nitrogen vacancy center in a nanodiamond with a traceable spectral photon flux was realized [1]. In this article, we report on the determination of the measurement uncertainty of the spectral photon flux (about 4 %) and discuss its different components. Furthermore, the angular distribution of the emission of an NV center in a nanodiamond located in the vicinity of a dielectric boundary is calculated based on the model of Lukosz and Kunz. The agreement between the angular distribution calculation and the first-ever measurement of the angle-dependent emission in nanodiamond NV centers is satisfactory.

1. Introduction

Single-photon sources (SPSs) are highly relevant for several forms of quantum-based technology such as quantum communication and quantum key distribution [2], [3], [4]. In radiometry, SPSs with a negligible background and a pure single-photon emission show promise as standard sources [1], [5] that can efficiently calibrate single-photon detectors; however, this is because the influence of the photon statistics on the calibration results is omitted [1], [6]. Currently, single-photon detectors are calibrated by means of attenuated lasers, resulting in an unavoidable measurement correction caused by the photon statistics and the dead time of the detector [7]. Using a pure, on-demand single-photon source would make such a measurement correction unnecessary [6], thus leading to lower uncertainties. A possible candidate for such a single-photon source is the nitrogen vacancy (NV) center in nanodiamond, which photon emission has shown to be stable at room temperature (i.e., it does not bleach or show photo-blinking) [4].

In this paper, we briefly summarize the metrological characterization of a single-photon emitter with respect to its total spectral photon flux, as presented in detail in Ref. [1]. Furthermore, we present the detailed estimation of its measurement uncertainty and report on the calculation and measurement of the angular emission characteristic of an NV center.

2. The single-photon source

The single-photon source consists of the emitter and the setup that excites the emitter and collects and analyzes its emission. A nanodiamond doped with a nitrogen vacancy center (NV center) was used as the emitter. The nanodiamond had a size of approximately 75 nm and was placed on a cover glass by
means of a spin coating procedure. The sample was fabricated by Friedrich Alexander University (FAU).

The setup used is a confocal microscope developed in-house [1]. Excitation takes place via a laser beam at a wavelength of 532 nm, which is reflected by a dichroic beam splitter and focused by means of an oil-immersion microscope objective (numerical aperture of 1.45). The light emitted passes the microscope objective and the dichroic beam splitter as well as some edge filters in order to ensure that only the NV center emission is collected, and in order to prevent any stray light from being transmitted. The emission by the NV centers is coupled into an optical fiber that is connected to several devices (i.e., a calibrated low optical flux detector (LOFD), a spectroradiometer and a Hanbury-Brown and Twiss interferometer (HBTI)) in order to determine the photon flux and the spectral distribution, and in order to check the single-photon emission, respectively [1]. All measurements are carried out at room temperature.

3. Absolute metrological characterization

The absolute metrological characterization of the NV center-based single-photon source was described in detail in [1]. The absolute photon flux per wavelength and per second is described by the following equation [1]:

\[
N_{ph,\lambda}(\lambda) = \frac{\Phi_\lambda(\lambda)}{e_{ph}(\lambda)} = \frac{\lambda}{hc} \frac{U_{out}}{t_{int}} \int S_{rel}(\lambda) s_{det}(\lambda) d\lambda = \frac{\lambda}{hc} \frac{U_{out}}{t_{int}s_{spec}} S_{rel}(\lambda),
\]

where the factors \( h \) and \( c \) are the Planck constant and the speed of light, respectively. The measurands of the low-optical flux detector (LOFD) [8] are the output voltage \( U_{out} \), the integration capacity \( c_{int} \) and the integration time \( t_{int} \). The effective spectral responsivity \( s_{spec} \) is defined as the combination of the relative spectral distribution of the NV center emission \( S_{rel}(\lambda) \) and the spectral responsivity of the detector \( s_{det}(\lambda) \). \( S_{rel}(\lambda) \) was determined by calibrating the spectrometer with respect to its spectral irradiance responsivity (i.e., traceable to the blackbody radiator) [1]. For clarification, the traceability chain for the determination of the spectral photon flux is shown in Figure 1a. The absolute spectral photon flux (i.e., the absolute number of photons per wavelength and per second) is shown in Figure 1b.

Figure 1: a) Traceability chain for the metrological characterization of a single-photon source in terms of its absolute spectral photon flux [1]; b) Absolute spectral photon flux of the single-photon source (blue curve). The calculation of the measurement uncertainty presented (red stripe) is described in Section 4.

4. Measurement uncertainties

The determination of the measurement uncertainty for the absolute spectral photon flux \( N_{ph,\lambda}(\lambda) \) follows the model shown in equation (1) and is carried out in accordance with the procedure described in the Guide to the expression of uncertainty in measurement (GUM) [9].
Several parameters have to be taken into account in order to determine the measurement uncertainties of the absolute spectral photon flux of the single-photon source. Three groups of parameters can be identified: uncertainties related to the error contribution from the determination of the spectrum; those related to the detection of the absolute photon flux; and some other, minor contributions.

The error caused by the spectrum is the largest. It mainly arises from the resolution error, which depends on the entrance slit and the grating used. These lead to smoothed structures that have to be corrected. Based on a measurement obtained by means of a narrow-linewidth laser, a triangular slit function with a bandwidth of 1 nm is assumed. In principle, this leads to a correction to the measured spectrum of about 3.8 %; for simplification, this is taken into account as a measurement uncertainty. Further analysis requires a more detailed characterization of the spectroradiometer, especially of the slit function that has to be carried out in the future. Furthermore, the effect of the spectrometer calibration has to be considered. Calibration with a standard tungsten lamp leads to an uncertainty in the relative spectral responsivity of about 1 %. Stray light and second-order effects are negligible: in the measurement with a narrow bandwidth laser, neither of these effects had a significant influence. This gives a total measurement uncertainty for \( S_{rel} \) of 3.9 %.

In addition to the spectral contributions, the detector also has an impact on the measurement uncertainty. The uncertainty sources associated with the LOFD are obtained from Ref. [8] and are thus mentioned only briefly below. The standard uncertainty of the effective spectral responsivity is 0.3 %. The integration time has a standard uncertainty of 0.3 %. The uncertainties related to the outcome voltage and the capacity are 0.5 % and 0.014 %, respectively [3]. All uncertainty sources and their contributions to the measurement uncertainty of the source photon flux for a wavelength of 770 nm are listed in Table 1.

| Measurand       | Value     | Standard uncertainty (%) | Distribution | Contribution (%) |
|-----------------|-----------|--------------------------|--------------|------------------|
| \( U_{out} \)   | 0.01854642| 0.5                      | Standard     | 9.97             |
| \( C_{int} \)   | 1.0046E-12| 0.014                    | Standard     | 0.27             |
| \( S_{rel} \)   | 0.0047    | 3.9                      | Square       | 77.79            |
| \( t_{int} \)   | 1         | 0.3                      | Standard     | 5.98             |
| \( s_{spec} \)  | 0.338088662| 0.3                      | Standard     | 5.98             |
| \( N_{ph3}(\lambda) \) |           | 3.95                     | Standard     |                  |

As expected, the largest contribution of 77.8 % is caused by the measurement of the relative spectral power distribution \( S_{rel} \). Overall, the relative standard uncertainty was calculated as being 4.0 % \((k = 1)\), giving an expanded uncertainty \((k = 2)\) of 7.9 %.

5. Angular emission of the NV center emission

The emission of the NV center in a nanodiamond can be considered a dipole emission into the 4\( \pi \) solid angle. However, it can be modified by placing the NV emitter in dielectric structures that alter the emission characteristic – for example, within the near-field distance of an antenna that could then improve the collection efficiency [10], [11].

The angular emission of the NV center in a nanodiamond is influenced by the cover glass, which is a dielectric structure and therefore acts like an antenna. This implies that the NV center emission is redirected in a preferred direction. The calculation regarding the angular emission is carried out in accordance with reference [12]. The field of illumination is divided into three areas (see Figure 2a) and the results regarding the illumination intensity are rotationally symmetric around the z-axis. The
first partition belongs to the transition between the air and the cover glass. Moreover, the illumination in the cover glass is divided into two areas that are separated by the critical angle (i.e., the angle of total reflection). Below, the angular emission calculations for the three areas in accordance with Ref. [12] are presented.

![Diagram](image)

Figure 2: a) Schematic representation of the field of illumination divided into the three calculation areas; the calculated angular emission is presented in blue; b) Calculated angular emission of an NV center-doped nanodiamond on a cover glass (blue curve); the collection efficiency is presented in green and the opening angle of the microscope objective used is indicated in orange.

The electric dipole component of the emission into air (Area I in Figure 2a), $P_{e,L}$, is given by the following equation [12]:

$$
P_{e,L} = \frac{3}{8\pi} \frac{n^2 \sin^2(2\alpha_{L1})}{(n \cos(\alpha_{L1}) + \cos(\alpha_{L2}))^2}.
$$

(2)

The factor $n$ is the ratio of the refractive index of the cover glass ($n_2 = 1.5255$) and the refractive index of air ($n_1 = 1$). The angles $\alpha_{L1}$ and $\alpha_{L2}$ are defined as follows [12]:

$$
\frac{\pi}{2} \leq \alpha_{L1} \leq \pi
$$

(3)

and

$$
\alpha_{L2} = \arcsin\left(\frac{\sin(\pi - \alpha_{L1})}{n}\right).
$$

(4)

The second partition occurs in the cover slip itself at the angle of total reflection. For angles larger than the critical angle (Area II in Figure 2a), the equation for the electric dipole emission is [12]:

$$
P_{e,G,2} = \frac{3}{8\pi n^2 - 1} \frac{n^5 \sin^2(\alpha_{G2})}{(n^2 + 1)^2 \sin^2(\alpha_{G2}) - 1)}
$$

(5)

with

$$
\arcsin\left(\frac{1}{n}\right) \leq \alpha_{G2} \leq \frac{\pi}{2}.
$$

(6)

For angles smaller than the critical angle (Area III), the electric dipole emission is defined as [12]:

$$
P_{e,G,1} = \frac{3}{8\pi} \frac{n^5 \sin^2(2\alpha_{G1})}{(n \cos(\alpha_{G1}) + \cos(\alpha_{G1}))^2},
$$

(7)

where

$$
0 \leq \alpha_{G1} \leq \arcsin\left(\frac{1}{n}\right)
$$

(8)
and

\[ \alpha_{1G2} = \arcsin(ns\sin(\alpha_{1G1})) . \]  

(9)

The results of the calculations are presented in Figure 2b (blue curve). The highest amount of the emission is directed into the cover glass at an angle of 41°. The calculated collection efficiency (i.e., the integration over the signal normalized to the total signal) is also illustrated as a green curve. Here, 87% of the illumination is emitted into the cover glass (angles from 0° to 90°) and only approx. 13% (angles from 90° to 180°) of the photons are directed into the air. The microscope objective used has a numerical aperture corresponding to a half opening angle of 72.8° (orange line), thus allowing 85% of the emitted photons to be collected.

In the experiment, the angular emission is measured under the application of a back focal plane lens and an sCMOS camera. The resulting image is shown in Figure 3a. The pattern presented corresponds to two times the opening angle of the microscope objective. The intensity of the emission is color coded (i.e., the bright ring belongs to the strongest emission). The emission intensity profile was determined by taking the signal of the camera along the white line. As can be seen, the resulting plot (green curve in Figure 3b) agrees well with the calculations (blue curve). The deviation observed between the calculated and measured intensity profile is assumed to be due to distortion arising from the back focal plane lens.

Furthermore, the emission shows a circular intensity modulation, as can be clearly seen in the bright circle. This pattern indicates the dipole characteristic of the NV center emission and is observed for all NV centers under investigation. However, the orientation differs from one NV center to another, according to the orientation of the nanodiamond and the NV center.

![Figure 3: a) Measured picture of the angular emission of an NV center; b) Measured (green) and calculated (blue) intensities of the angular emission for the opening angle of the microscope objective in the range of -80° to 80°.](image)

6. Conclusion

In this article, the metrological characterization of a single-photon source based on a nitrogen vacancy center-doped nanodiamond was presented in terms of its absolute spectral photon flux. The relative standard uncertainty of the spectral photon flux was determined to be 4.0%. To reduce the uncertainty, it is planned to optimize the resolution of the spectroradiometer in order to lower the highest uncertainty contribution.

Furthermore, investigations on the angular emission of an NV center were presented. The results of the measurement correspond to the calculations and the dipole characteristic of the NV emission was observed. The measurement carried out was for a half sphere only; for this reason, further investigations are planned in which the angular emission in a full sphere will be measured by means of a double-confocal microscope [13].

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