Absorption of a heralded single photon by a nitrogen-vacancy center in diamond

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A simple, room-temperature, cavity- and vacuum-free interface for an efficient photon-matter interaction is implemented. In the experiment a heralded single photon generated by the process of spontaneous parametric down-conversion is absorbed by a single atom-like system, specifically a nitrogen-vacancy color center in diamond. Here phonon-assisted absorption solves the mismatch problem of a narrow absorption bandwidth in a typical atomic medium and broadband spectrum of quantum light. This allows observation of fluorescence only in the spectral range 452 – 575 nm, which overlaps well with the absorption spectrum of nitrogen-vacancy centers. This can also be considered as a useful technique paving the way for development of novel quantum information processing and sensing applications.

Efficient control of light-matter interaction at a single-particle level is a key factor enabling several quantum applications. One of the main goals in quantum technology is to deliver a quantum network platform for secure quantum communication. Atom or trapped ion implementations of quantum memories and quantum repeaters together with photonic information carriers are one of the proposed scenarios [1–3]. Moreover, quantum imaging and sensing benefit from single-photons interaction with matter. In particular, this is apparent when discussing quantum illumination, sub-shot-noise imaging, ghost imaging, and absolute detector calibration [4]. Further, absorption microscopy exhibits higher signal to noise ratio when quantum light is applied [5]. Entangled two-photon absorption is another microscopy technique which has a great potential to outperform its classical counterpart in terms of applicability [6]. It may also serve as a tool for virtual-state spectroscopy [7–9]. However, all the above-mentioned applications require an efficient single-photon and single atomic system interaction, which still remains a challenge. The narrow atom absorption spectrum together with spectrally broad single-photon sources demand cavities to enhance the interaction efficiency [10–12].

Here, for the first time, a different solution is proposed and the mismatch problem is solved by using a spectrally broad absorber, such as a color center in diamond, where a nonresonant excitation is possible due to phononic sidebands [13]. This eliminates the need for excitation with a narrow bandwidth light. A plethora of such systems, like SiV, GeV, SiC or dyes used in biological imaging, is already known and investigated [14–16].

A simple, room-temperature, cavity- and vacuum-free interface for photon-matter interaction is reported. The heralded single-photon source (HSPS) is based on the spontaneous parametric down conversion (SPDC) process, where the detection of an infrared photon is used as a herald for the visible one. In the experiment, a heralded single photon is absorbed by a single atom-like system, specifically a nitrogen vacancy center (NV) in diamond. The NV center then emits another photon, whose arrival time is measured by means of a time-resolved single-photon detection technique.

I. RESULTS

A. Experimental setup

The experimental setup is depicted in Fig. 1 (a). A blue pump photon is converted through the SPDC process into a pair of photons – one in visible and one in infrared spectral range. When the superconducting single photon detector (SSPD) measures the latter, the existence of the former is heralded and the time reference is defined. Subsequently, the heralded visible photon is delivered to the custom-made confocal microscope (CM), where it is reflected off a dichroic mirror (DM) and focused with a microscope objective (MO) onto a high NV−-concentration diamond sample. Next, the photon excites a color centre, which results in a fluorescence photon emission in the range of 600 – 800 nm [17], see Fig. 1 (d). The resulting photon is collected by the same MO and propagates through the DM and a longpass filter. This allows observation of fluorescence only in the spectral range above 700 nm. Finally, its arrival time is measured by a single-photon avalanche diode (SPAD) supported by a digital oscilloscope. The statistics of arrival
FIG. 1. Interaction between photons and NV centers. (a) The experimental setup. Detailed experimental setup description is available in the Appendix and Supplementary Information. (b) Magnification of the diamond surface: One of the NV centers from the illuminated ensemble absorbs heralded green photon and then emits fluorescence photon. (c) Simplified energy level structure of an NV centre: a green photon excites an electron from the $^3A_2$ (ground) state to a $^3E$ (excited) state. The electron then decays non-radiatively to a lower energy excited state, transferring some of its energy to the diamond lattice. Finally, the electron decays radiatively back to the $^3A_2$ (ground) state via the emission of a lower energy red photon. (d) Blue: NV$^-$ fluorescence spectrum with zero phonon line (ZPL) at 637 nm and vibronic sideband up to 800 nm. Red: the transmission range of the spectral filtering setup (MD3 and F3).

times collected by the measurement system are used to build a histogram exhibiting the characteristic fluorescence decay shape.

The HPS used in the experiment is tunable in the range of 452-575 nm [18]. The scenario with a 532 nm single-photon was chosen but other pumping scenarios were tested as well (see Supplementary Information). For the source two pumping power settings, 1 and 5 mW, were used, which are referred to as lower and higher, respectively. Both powers ensure high-quality single photon generation (see Appendix).

B. Fluorescence decay measurement

In Fig. 2 the obtained NV fluorescence decay pumped with heralded single photons is shown for the two pumping power settings. The probability of the fluorescence emission in time, $p(t)$, can be described by a simple exponential model including a non-radiative decay path. The model assumes a fast non-radiative decay followed by a radiative decay, with characteristic times $\tau_N$ and $\tau_R$, respectively. The explicit formula can be obtained by taking a convolution of the two single exponential decays

$$p(t) = \theta(t) \cdot \frac{1}{\tau_N - \tau_R} \left( e^{-\frac{t}{\tau_N}} - e^{-\frac{t}{\tau_R}} \right), \quad (1)$$

where $\theta(t)$ is the step-function (equal to 0 for $t < 0$ and to 1 for $t \geq 0$). The SPDC source is pumped with a laser emitting pulses of 140 fs time duration every $t_0 = 12.5$ ns. Therefore, a photon pair can be generated every multiple of the repetition period $t_0$. Note, that due to the probabilistic nature of the SPDC process not every pulse generates a pair. On top of that due to the losses in the experimental setup not all of the observed coincidences registered in the detection system originate from the same pump pulse. Sometimes the pulse that produces the visible photon exciting the NV center and causing the fluorescence is different than the pulse from which the detected heralding photon is created. Such coincidences are referred to as accidental, while the coincidences caused by a single pump pulse are called proper. The ratio of accidental to proper coincidences, $r$, can be also used as a parameter of the fluorescence model given by the formula below

$$P(t) = a \left( p(t) + r \sum_{n=1}^{\infty} [p(t + nt_0) + p(t - nt_0)] \right) + b, \quad (2)$$
Two possible explanations for this discrepancy are proposed. Firstly, the sample is illuminated with single photons resulting in the NV center not being spin-polarized, as opposed to the case of the excitation with a high power laser [19, 20]. Secondly, the experiment is performed on a very dense sample (see Appendix), where the shorter decay time can be attributed to the Förster resonance energy transfer (FRET). It makes the fluorescence emission faster when the emitters are close to each other [21].

The fitted ratio, r, takes the value of 0.233(9) (0.0225(81)) in the higher (lower) pumping power setting. The difference reflects the fact that for the higher pump power setting of the HSPS the ratio of accidental to proper coincidences is higher. It can be clearly seen when comparing black histograms in Fig. 2. Details on r estimation for other pumping wavelengths can be found in Supplementary Information.

C. Signal to noise ratio (SNR)

Due to the heralding scheme applied in the experiment the signal to noise ratio (SNR) analysis is done in a more sophisticated manner. Instead of comparing the total number of real fluorescence counts and dark counts registered on the single-photon detector, only heralded photons are taken into account. For experimental conditions in the higher (lower) power setting this results in 2.96(27)% (1.42(16)%), of the real fluorescence and 0.446(12)% (0.172(22)%), respectively. The difference is due to the fact that the detection window for the fluorescence monitoring detector is being opened only when a heralding photon was already detected. Therefore it is closed most of the time and opened only when there is an increased probability of detecting a fluorescence photon, which is heralded by heralding photon detection. Hence, the effective detection probability of a fluorescence photon is higher than of a dark count within a given detection window. This leads to the increase of SNR for the higher (lower) power setting from 0.643(46) (0.154(16)), for the non-heralding scenario, to 4.3(8) (1.3(6)), for the heralding one.

II. DISCUSSION

SNR can be improved by replacing the fluorescence monitoring detector with a smaller one. The detector currently used has an active area with 50 µm diameter, exhibiting 65(8) dark counts per second. The detector could be replaced by one with 30 µm diameter giving 1 dark count per second with the same photon detection efficiency of the order of 15 – 28 % for the spectral range of interest, which is 700 – 800 nm [22]. The expected SNR would increase by a factor of 65. Also, it is estimated, when switching from pulsed to continuous-wave laser pumping of the SPDC source, the power can be safely increased up to 200 mW with single-photon characteristics of the HSPS maintained [23]. This should increase the SNR by a factor of 40. All together, the
low dark count rate SPAD detector along with 200 mW continuous-wave-laser-pumped HSPS should lead to SNR on the level of $10^4$.

Now the problem of converting a heralded photon generated by HSPS to the fluorescence photon emitted by an NV center is addressed. This quantity can be estimated based on the observed count rates to be $1.46(32) \cdot 10^{-4}$ ($2.1(8) \cdot 10^{-5}$) for higher (lower) power setting. This is the efficiency of the sample, meaning the experimental setup imperfections are excluded. The setup sets additional limits resulting in the observed conversion efficiency of $7.0(7) \cdot 10^{-6}$ ($1.01(25) \cdot 10^{-5}$) for higher (lower) pumping setting. This is due to experimental factors including: efficiency of extracting fluorescence photons from the diamond, losses in optical elements, fiber coupling efficiencies and quantum efficiencies of the detectors. It is worth mentioning that while the experiment was performed with the aid of an oil-immersion MO, it is also possible to conduct it with a dry air MO. Such configuration would result in lower SNR but would enable cryogenic experiments.

In conclusion, a simple, room-temperature, cavity- and vacuum-free single-photon single atom-like system interaction was demonstrated. The experiment was performed on a high NV$^-$-concentration diamond sample with a tunable, SPDC-based heralded single-photon source. Interaction efficiency, usually limited by the very small spectrum overlap between the broadband quantum light and narrow atomic transitions, was significantly enhanced thanks to phonon-broadened absorption in nitrogen-vacancy centers in diamond.

The result can also be considered as a useful technique paving the way for development of novel quantum information processing and sensing applications, including quantum networks, quantum microscopy [5, 6] or virtual-state spectroscopy [7–9]. In particular, the ability of interaction of NVs with statistics-controlled quantum light enables two-photon processes, such as ionization of negatively-charged NV centers, to be eliminated.

Moreover, it can be extended to the scenario of a low NV$^-$ concentration sample, which would enable addressing of a single color centers. It would facilitate testing of the fundamental properties of quantum light interacting with atomic systems. As a further step, several applications, such as quantum microscopy, can be enhanced when non-classical properties of quantum light are exploited.

III. APPENDIX

A. Experimental setup

The experimental setup is depicted in Fig. 1 (a). The infrared light from the 80 MHz pulsed laser pumps a BiBO crystal to generate blue photons by second harmonic generation (SHG). The infrared photons are filtered out with dichroic mirrors (DM1). The blue pump photons undergo SPDC process in the PPKTP crystal to produce a pair of photons, one visible and one infrared [18]. The photons are separated by a dichroic mirror (DM2) and further the blue pump photons are filtered out (F1 and F2). The superconducting single photon detector (SSPD) measures the arrival time of the heralding (infrared) photon. The heralded (visible) photon is coupled using a fibre coupler FC1 and travels through a fibre (SMF780) to the confocal microscope (CM). The photon is reflected off a dichroic mirror (DM3) and focused with a high NA= 1.45 oil-immersion microscope objective (MO) onto a high NV$^-$ concentration diamond sample. The fluorescence photon is collected by the same MO and propagates through DM3 and a longpass filter (F3). A single photon avalanche diode (SPAD) detects the fluorescence photon and basing on the SSPD and SPAD signals the fluorescence decay histogram is built on the oscilloscope.

B. HSPS

The visible range photon, generated by HSPS, was characterized in both pumping power settings, for which the $g^{(2)}(0)$ correlation function [24, 25], describing the goodness of the single-photon source, was measured. For lower power pump it took the value of $g^{(2)}_{\text{h}}(0) = 0.0011(2)$, whereas for higher pump power $g^{(2)}_{\text{b}}(0) = 0.0111(5)$. The corresponding heralded photon count rates were 4.5 kcps (lower power) and 40 kcps (higher power). The HSPS characterization was done for 532 nm.

C. Fluorescence decay model

When the NV centre absorbs a photon, it non-radiatively decays to a lower energy excited state, before decaying further to the ground state and emitting a photon. Since the two processes occur consecutively, the fluorescence emission probability over time, $p(t)$, can be calculated as a convolution of the non-radiative, $p_N(t)$, and radiative, $p_R(t)$, decay probability distributions

$$
p_N(t) = \frac{1}{\tau_N} e^{-t/\tau_N},
$$

$$
p_R(t) = \frac{1}{\tau_R} e^{-t/\tau_R}.
$$

The resulting formula is as follows

$$
p(t) = \int_0^t p_N(t')p_R(t-t')dt' = \frac{1}{\tau_N - \tau_R} (e^{-t/\tau_N} - e^{-t/\tau_R}).
$$
Only the probability of the decay for \( t > 0 \) is considered since the NV centre must be excited before it can decay. Thus, the probability distribution \( p(t) \) is redefined to

\[
p(t) = \theta(t) \cdot \frac{1}{\tau_N - \tau_R} \left( e^{-\frac{t}{\tau_N}} - e^{-\frac{t}{\tau_R}} \right),
\]

where \( \theta(t) \) is the step-function equal to zero (one) for the negative (positive) values of \( t \) such that the probability of decay is 0 for \( t < 0 \).

### D. Diamond

The sample used in the experiment is a high-pressure high-temperature (HPHT) diamond with a dense concentration of negatively charged nitrogen-vacancy (NV\(^-\)) centres, approximately 18 ppm. Full sample characterization and preparation description, including spectrum and ODMR measurement, is given in Refs. \([26, 27]\).

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I. FLUORESCENCE DECAY MODEL

When the NV centre absorbs a photon, it non-radiatively decays to a lower energy excited state, before decaying further to the ground state and emitting a photon. Since the two processes occur consecutively, the fluorescence emission probability over time, \( p(t) \), can be calculated as a convolution of the non-radiative, \( p_N(t) \), and radiative, \( p_R(t) \), decay probability distributions.

\[
\begin{align*}
p_N(t) &= \frac{1}{\tau_N} e^{-t/\tau_N}, \\
p_R(t) &= \frac{1}{\tau_R} e^{-t/\tau_R}.
\end{align*}
\]

The resulting formula is as follows

\[
p(t) = \int_0^t p_N(t')p_R(t - t')dt' = \frac{1}{\tau_N - \tau_R} \left( e^{-\frac{t}{\tau_N}} - e^{-\frac{t}{\tau_R}} \right).
\]

Only the probability of the decay for \( t > 0 \) is considered since the NV centre must be excited before it can decay. Thus, the probability distribution \( p(t) \) is redefined to

\[
p(t) = \theta(t) \cdot \frac{1}{\tau_N - \tau_R} \left( e^{-\frac{t}{\tau_N}} - e^{-\frac{t}{\tau_R}} \right),
\]

where \( \theta(t) \) is the step-function equal to zero (one) for the negative (positive) values of \( t \) such that the probability of decay is 0 for \( t < 0 \).

The detailed version of the experimental setup scheme for the heralded measurement illustrated in Fig. 1 (a) in the main text is given in Fig. S1. In the SPDC process, a pair of photons is produced in a nonlinear crystal. Since the photons are created together and the chromatic dispersion in the setup in negligible, the time delay between their detection events is constant and depends on the difference in path lengths of the photons and detection signal. In a histogram of time delays, Fig. S2 (b-c), this is visible as a peak in coincidences, further called proper coincidence peak. However, due to setup imperfections, one or both of the photons from a given pair can be lost. When this occurs, the coincidence may instead happen between photons from different pairs, as shown in Fig. S2 (a). This results in a time delay that is a multiple of the rate of generation of the photon pairs, which is the time between laser pulses \( t_0 \). These events, called accidental coincidences, can be seen as smaller peaks in the number of coincidences in Fig. S2 (b-c).

The presence of accidental coincidences has a direct influence on the heralded measurement and hence further modificaions of the analytical model are required. As a result the histogram is formed as a sum of the proper decay, originating from the proper coincidence visible photons, and accidental decays, originating from the accidental coincidence photons. The corresponding formula for the probability distribution function for the time of fluorescent photon detection can be written as

\[
P(t) = A \left( p(t) + r \sum_{n=1}^{\infty} [p(t + nt_0) + p(t - n \cdot t_0)] \right) + b,
\]
**FIG. S1. Detailed experimental setup.** Infrared light from a Ti:Sapphire pulsed laser is focused by lens L1 (plano-convex, \( f = 7.5 \) cm) onto a nonlinear BiBO crystal. The resulting blue photons are collimated by another lens L1. Two dichroic mirrors, DM1 (Chroma AHF T425LPXR), separate unconverted laser light. The halfwave plate HWP rotates the laser beam polarization before it is focused by lens L2 (plano-convex, \( f = 10.0 \) cm) into the type-II PPKTP nonlinear crystal (10 \( \times 4 \times 2 \) \( \mu m \)). Dichroic mirror DM2 (Chroma AHF 76−875−LP) separates the visible and infrared photons. Lens L3 (plano-convex, \( f = 15.0 \) cm) collimates the visible photons and then filters F1 (ET500 and FF550/88) remove unconverted laser light before photons are coupled into a single mode fibre (SMF780) by fibre collimation package with focal length either \( f = 1.51 \) cm or \( f = 1.1 \) cm (FC1). Infrared photons are transmitted through DM2, collimated by lens L4 (plano-convex, \( f = 10.0 \) cm), filtered by filter F2 (1319 LP), and coupled into a single mode fibre (F1-2000-FC-1) using mirror M and fibre collimator FC1 (\( f = 1.51 \) cm). Superconducting nanowire single photon detector, SSPD, detects the IR photons. Visible photons (532/565/575 nm) travel through fibres and are collimated by collimation package FC2 (F671FC−405) to reflect off dichroic mirror DM3 (Semrock FF573-DIO1). Microscope objective MO focuses green photons on diamond sample and collects red fluorescence. Red photons pass through DM3 and remaining green photons are removed by filter F3 (Thorlabs FELH550/FELH700/BP650−40). For tests with 532 nm, 565 nm, and 575 nm green photons, fluorescence photons with wavelengths below 550 nm or 700 nm, below 700 nm, and between 610 nm and 690 nm were filtered respectively. Lens L5 (plano-convex, \( f = 3.0 \) cm) focuses photons onto avalanche photodiode SPAD. Oscilloscope detects signals from SSPD and MPD.

**II. EXPERIMENT**

**A. Heralded Fluorescence Measurement - Varying Excitation Wavelengths**

Several measurements with different HSPS and fluorescence filter settings were performed. The combinations of the central wavelength for the visible photons generated in SPDC process, \( \lambda_e \), the filter cut-off wavelength, \( \lambda_f \), and the pump power setting used in the experiment are listed in Tab. S1.

Tab. S1 shows the goodness of the model fit used to determine the decay time from the data (Eq. (S3)). The goodness of the fit is described by the p-value from the \( \chi^2 \) test where a larger p-value indicates a higher probability that the model fits the data. The data analysis was done

\[
P(t) = \frac{A}{\tau_N - \tau_R} \left[ r \left( e^{-\frac{t}{\tau_N}} - e^{-\frac{t}{\tau_R}} \right) + \theta(t) \left( e^{-\frac{t}{\tau_N}} - e^{-\frac{t}{\tau_R}} \right) + \theta(t-t_0) r \left( e^{-\frac{t+t_0}{\tau_N}} - e^{-\frac{t+t_0}{\tau_R}} \right) + \theta(t-2t_0) r \left( e^{-\frac{t+2t_0}{\tau_N}} - e^{-\frac{t+2t_0}{\tau_R}} \right) \right] + b. \quad (S3)
\]
FIG. S2. Accidental coincidences. (a) When one of the photons in the SPDC photon pair is lost, this can result in coincidences between photons in neighbouring pairs, accidental coincidences. (b),(c) Time delay between visible and infrared SPDC photons for higher and lower power settings, respectively. The accidental coincidences almost disappear in the lower power setting.

in Mathematica and Python. Both, the data and the codes for the data analysis, are available upon request.

B. Heralded Fluorescence Measurement - accidental coincidences

Tab. S1 compares the ratio between the coincidence peaks to the factor $r$ fitted in the decay model (Eq. (S3)) for different excitation wavelengths ($\lambda_e$), fluorescence photons’ wavelengths ($\lambda_f$), and higher or lower power settings. The columns ratio 1 and ratio 2 are related to the first and second accidental peaks in the histogram shown in Fig. S2, respectively.

C. Experimental imperfections

The heralded fluorescence system conversion efficiency was estimated to be $7.0(7) \times 10^{-6}$ ($1.01(25) \times 10^{-5}$) for higher (lower) power setting. It includes pump 532 nm to NV fluorescence photon conversion efficiency, efficiency of extracting a fluorescence photon from the high refractive index diamond, optics losses, fiber coupling efficiency and spectrum weighted average quantum efficiency of the SPAD detector in the spectral range of interest for the higher. The corresponding values are listed in Tab. S2.

D. Signal to noise ratio (SNR)

The heralded measurement scheme was applied. The heralded signal counts were estimated by multiplying the observed fluorescence signal counts, $n_f$, by the coincidence, $n_{\text{coinc.}}$, to visible, $n_{\text{VIS}}$, count rates ratio, giving the rate of heralded fluorescence counts per second

$$n_{\text{sh}} = \frac{n_{\text{coinc.}}}{n_{\text{VIS}}} \cdot n_f.$$ 

The heralded dark counts were estimated as follows. The detected amount of dark counts (DC), $n_{\text{DC}}$, was multiplied by the probability of detecting a heralded DC, $p_{\text{herald.}}$,

$$n_{\text{DCh}} = p_{\text{herald.}} \cdot n_{\text{DC}},$$

$$p_{\text{herald.}} = \eta_{IR} \eta_{SSPD} \eta_{SPDC},$$
where $\eta_{IR}$ denotes the infrared photon fiber coupling efficiency, $\eta_{IRSSPD}$ stands for the SSPD quantum efficiency for the IR photon and $\eta_{SPDC}$ for the SPDC process efficiency per pulse. Hence, finally the SNR was calculated as a ratio

$$SNR = \frac{\eta_{sh}}{\eta_{DCch}}.$$ 

### TABLE S1. Decay time and accidental coincidence ratio at varying wavelengths.

| $\lambda_e$ (nm) | $\lambda_f$ (nm) | power | decay time | p-value | ratio 1 | ratio 2 | $r$  |
|-----------------|-----------------|-------|------------|---------|---------|---------|------|
| 532             | >550            | higher| 7.4(2)     | 0.48    | 0.2333(7) | 0.1741(6) | 0.192(7) |
| 532             | >550            | lower | 7.7(2)     | 0.20    | 0.0349(7) | 0.0345(7) | 0.055(6) |
| 532             | >700            | higher| 7.68(23)   | 0.46    | 0.241(1)  | 0.224(1)  | 0.233(9) |
| 532             | >700            | lower | 7.17(14)   | 0.57    | 0.024(1)  | 0.025(1)  | 0.0225(81) |
| 565             | >700            | higher| 7.6(2)     | 0.23    | 0.178(2)  | 0.177(2)  | 0.198(6) |
| 565             | >700            | lower | 7.5(2)     | 0.39    | 0.0434(4) | 0.0398(4) | 0.067(6) |
| 575             | 610-690         | higher| 6.9(4)     | 0.61    | 0.15(2)   | 0.133(2)  | 0.18(2) |

### TABLE S2. Experimental imperfections.

| Contributing factor                                                                 | higher         | lower         |
|-------------------------------------------------------------------------------------|----------------|---------------|
| pump to fluorescence photon conversion efficiency                                    | $1.46(32) \cdot 10^{-4}$ | $2.18(8) \cdot 10^{-4}$ |
| fiber coupling efficiency                                                             | 0.34(5)        | 0.352(54)     |
| efficiency of extracting a fluorescence photon from the diamond                      | 0.09(1)        |               |
| optics losses                                                                        | $4.8(6) \cdot 10^{-2}$ |               |
| SPAD quantum efficiency                                                              | 0.220(5)       |               |