Observation of Quantum Advantage with Squeezed Light for Absorption Measurement

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Absorption measurements are routinely used in science and engineering, it is an exceptionally versatile tool for most applications. For absorption measurements using laser beams of light, the sensitivity is theoretically limited by the shot noise due to the fundamental Poisson distribution of photon number in laser radiation. In practice, the shot-noise limit can only be achieved when all other sources of noise are eliminated. Here, we use bright squeezed light to demonstrate direct absorption measurement can be performed with sensitivity beyond the shot-noise limit. We present a practically realizable scheme, where the bright squeezed light is in the continuous-variable regime generated by the four-wave mixing process in an atomic rubidium vapor. This is a direct sub-shot-noise measurement of absorption, and more than 1 dB quantum advantage for the measurement sensitivity is demonstrated at faint absorption levels. A theoretical model that yields excellent agreements with experimental observations is also provided.

It has been demonstrated that one can improve the sensitivity and precision of many classical measurement techniques using various quantum states of light [1–15]. Most prominently, sub-shot-noise detection of changes in optical phase have been demonstrated in interferometers using quantum light [16–19] and have been implemented for gravitational wave detection [20]. Although a straightforward readily attainable approach to achieve desired performances of a classical measurement is to simply increase the photon flux of the probe light to yield a greater signal-to-noise ratio, it has been proven uneconomical whenever one faces limits on the brightness of the optical probes, for instance, in the case where samples can be altered or damaged by the probe light [13–21]. It is therefore highly desirable to optimize measurement sensitivity with a fixed amount of input photon flux [13]. It is also important to note that for measurement schemes where the sensitivity itself varies with parameters of the measured sample it is possible for the sensitivity to be degraded, potentially requiring either prior knowledge about the optical sample or the addition of a feedback servo loop and control to ensure a sub-shot-noise performance [22–24].

Since the intensity measurement of an idealized laser fluctuates with a Poisson distribution, it is therefore used to define the shot-noise limit (SNL) in optical measurements, and it can only be reached in classical experiments once all other sources of noise are removed. For a direct measurement of optical transmission, the number of photons that pass through a sample is used to estimate the absorption $\alpha$, and thus the estimation sensitivity $\Delta\alpha$ is determined by the SNL. An approach that allows a sub-shot-noise measurement of an unknown sample’s transmission is to use quantum-correlated beams of photons [10, 14, 15]. In particular, such techniques have been implemented in the context of imaging [25, 26]. For practical applications, the reduction of noise between quantum-correlated beams of photons generated with spontaneous parametric down-conversion (SPDC) [27] or four-wave mixing (FWM) [16] is very attractive since correlations between photon pairs are unaffected by optical phase induced by a measured sample. This technique can be transferred to detecting correlated photons altogether in the same image of a charge-coupled-device (CCD) camera to acquire sub-SNL measurements in both the spatial domain [8, 10, 26, 28, 29] and most recently in the temporal domain as well [30]. With the inclusion of a spatially absorbing sample, it has been shown that correlated twin beams can be used to suppress noise in imaging objects to a degree that out-performs classical measurement using an equally efficient detection [10, 31]. Since absorption measurement is the most versatile tool for many applications in spectroscopy, metrology, chemistry and biology, improving the measurement sensitivity is thus indisputably beneficial to both science and engineering communities. It is therefore absolutely valuable for experiments to be performed to observe clear quantum advantages that gained by using quantum states of light in absorption measurements.

In this letter, we report an experimental scheme for a direct absorption measurement using bright squeezed light as the probe to demonstrate clear quantum advantage over the SNL. The bright squeezed light is generated with the FWM process in an atomic $^{85}$Rb vapor cell [32–35]. The experimental setup and the respective atomic level structure are shown in Fig. 5(a) and (b). The medium possesses a large third-order electric susceptibility $\chi^{(3)}$ and is pumped by a strong ($\sim 500$ mW) narrow-band continuous-wave (CW) laser at frequency $\nu_1$ ($\lambda = 795$ nm) with a typical linewidth $\Delta\nu_1 \sim 100$ kHz.
Applying an additional weak (~10 nW) coherent seed beam at frequency \( \nu_p = \nu_1 - (\nu_{HF} + \delta) \), where \( \nu_{HF} \) and \( \delta \) are the hyperfine splitting in the electronic ground state of \(^{85}\text{Rb}\) and the two-photon detuning respectively in Fig. 5(b) (further experimental details can be found in the Supplementary Material), two pump photons are converted into a pair of twin photons, namely ‘probe \( \nu_p \)’ and ‘conjugate \( \nu_c \)’ photons, adhering to the energy conservation \( 2\nu_t = \nu_p + \nu_c \) (see the level structure in Fig. 5(b)). The resulting bright twin beams are strongly quantum-correlated and are also referred to as bright two-mode squeezed light [39]. As can be seen from Fig. 5(c), the twin beams exhibit a intensity-difference squeezing of 6.7 dB measured by balanced photodiodes, which is indicative of strong quantum correlations [39] (see the Supplementary Material for further details on the squeezing measurement).

After the \(^{85}\text{Rb}\) vapor cell, the pump and the bright twin beams are separated by a second polarizer, with \( \sim 2 \times 10^5 : 1 \) extinction ratio for the pump. The probe beam transverses through a combination of a \( \lambda/2 \) plate and a PBS, acting as an absorption sample, while the conjugate beam serves as a reference. The twin beams are then focused onto an electron-multiplying charge-coupled-device (EMCCD) camera (Andor iXon Ultra 897). The EMCCD camera is enclosed in a light-proof box with filters installed at the entrance to block ambient light photons from entering the camera. The acousto-optic modulator (AOM) on the probe beam path is used to pulse the beam with 2 \( \mu \)s duration (FWHM) and duty cycle of 1/12. Since the CW pump beam is present all the time, the conjugate beam is therefore also pulsed as a result of the FWM process. The time sequencing of the pump and the twin beams are shown in Fig. 2(a) as the red strap, and the blue and green squares respectively.

We acquire the temporal quantum noise reduction of the bright twin beams through the use of the kinetic mode of the EMCCD camera [30]. The EMCCD has 512 \times 512 pixels with each pixel size of 16 \( \mu \)m\( \times \)16 \( \mu \)m. We focus the twin beams on the camera with an \( 1/e^2 \) beam diameter of \( \sim 50 \mu \)m, occupying roughly 3 pixels as shown in Fig. 2(b). The temperature of the EMCCD is kept low (\( < -65^\circ \text{C} \)) to curb the thermal noise contributions. We adopt the same method developed in Ref. [30] to capture images of the bright twin beams containing the desired absorption information. The rest of the EMCCD camera settings can be found in the Supplementary Material.

For each absorption \( \alpha \) (acquired by changing the angle of the \( \lambda/2 \) plate), we capture 200 kinetic series (i.e., 200 frame sequences), with each frame containing 35 pairs of probe and conjugate images. For the measurement of the quantum noise reduction between the bright twin beams, we adopt an algorithm originally developed in the spatial domain [28,29] but re-deriving it in the temporal domain [30]. In brief we crop a 10\( \times \)10 pixel region around the maximum-intensity area in each probe and conjugate images, large enough to enclose their respective full beam profiles (see Fig. 2(b)), we then are able to obtain the temporal photon counts fluctuations of the probe \( N_p(t) \) and conjugate \( N_c(t) \) as shown in Fig. 2(c) by integrating the photon counts in the cropped regions. As expected, strong correlations between the photon counts fluctuations of the bright twin beams can be observed in Fig. 2(c) and manifested in Fig. 2(d) through the subtraction and addition of these two modes. The quantum noise reduction characterization, \( \sigma_t \), in the temporal do-

![FIG. 1. (a) Experimental setup in which a seeded \(^{85}\text{Rb}\) vapor cell produces strong quantum-correlated twin beams via FWM. The twin beams are separated from the pump by a \( \sim 2 \times 10^7 : 1 \) polarizer after the cell. The probe beam passes through an absorption ‘sample’ (i.e., a combination of a \( \lambda/2 \) plate and a PBS) while the conjugate beam serves as a reference, before they are focused onto an EMCCD camera. The camera is enclosed in a light-proof box with filters mounted to block ambient light. The AOM on the probe beam path is used to pulse the twin beams with 2 \( \mu \)s FWHM and duty cycle of 1/12. PBS: polarizing beam splitter. PM fiber: polarization-maintaining fiber. (b) Level structure of the D1 transition of \(^{85}\text{Rb}\) atom. The optical transitions are arranged in a double-L configuration, where \( \nu_p, \nu_c \) and \( \nu_t \) are the probe, conjugate and pump frequencies, respectively, fulfilling \( \nu_p + \nu_c = 2\nu_t \). The width of the excited state in the level diagram represents the Doppler broadened line. \( \Delta \) is the one-photon detuning, \( \delta \) is the two-photon detuning, and \( \nu_{HF} \) is the hyperfine splitting in the electronic ground state of \(^{85}\text{Rb}\). (c) Measured intensity-difference noise power spectrum for the squeezed twin beams (red line) and for the SNL (blue line), obtained with a radio frequency spectrum analyzer (with resolution and video bandwidth of 300 kHz and 100 Hz, respectively). A squeezing of 6.7 dB is achieved.](http://example.com/fig1)
FIG. 2. (a) Time sequencing of the pump and the twin beams. The pulse duration of 2 µs and the duty cycle of 1/12 is realized by pulsing the probe beam with an AOM. The CW pump beam is present all the time. (b) Typical images of the twin beams with absorption $\alpha = 1.1\%$ captured by the EMCCD camera with four consecutive pulses. (c) Temporal photon counts fluctuations of the probe $N_p(t)$ and conjugate $N_c(t)$ obtained by integrating the photon counts in the cropped regions in (b). Clear similarities can be observed between the twin beams. (d) The strong noise reduction in the subtraction as opposed to the summation of the $N_p(t)$ and $N_c(t)$ depicted in (c) showcases strong correlations between them.

The main reads

$$\sigma \equiv \frac{\langle \Delta^2(N_p(t+\delta t) - N_p(t)) - \eta(N_c(t+\delta t) - N_c(t)) \rangle_t}{\langle N_p(t+\delta t) + N_p(t) + \eta N_c(t+\delta t) + \eta N_c(t) \rangle_t}$$

(1)

where $N_p(t+\delta t) - N_p(t)$ and $N_c(t+\delta t) - N_c(t)$ are the subtractions of photon counts in the cropped regions in two successive probe and conjugate images with time interval of $\delta t = 24$ µs. Since the intensities of the twin beams are inherently imbalanced due to the seed power and different transmissions through the vapor cell $\text{[29]}$, a scaling factor $\eta = 0.8$, which is obtained by taking the ratio between the conjugate and probe photon counts in the analysis regions without the presence of the absorption sample, is applied to the conjugate mode to rescale its photon count before the two modes are subtracted. The scaling factor effectively eliminates the DC portions of the Gaussian profiles of the probe and conjugate images. The subtraction of the two successive images leads to the cancellation of the low-frequency portion of the classical noise as well as the individual common Gaussian profiles of the probe and conjugate images $\text{[28, 29]}$. The numerator of Eq. (1) represents the temporal variance of the intensity-difference noise between the probe and conjugate pulses. The denominator gives the mean photon counts for the probe and conjugate pulses used for the analysis and represents the shot noise. For coherent state pulses $\sigma = 1$, which corresponds to the SNL, while for thermal light or other classical states $\sigma > 1$. Temporally quantum-correlated beams, like the bright twin beams generated in our experiment, will result in $\sigma < 1$, with a smaller $\sigma$ corresponding to a larger degree of quantum correlations (i.e., two-mode squeezing).

In Fig. 3 we plot $\sigma$ as a function of absorption $\alpha$ for the bright squeezed light (red dots) and coherent light (blue dots). Solid lines are the theoretical predictions. The quantum advantage is only significant for small values of $\alpha$, so the data was taken only for faint absorption levels. For the virtue of completeness we plot the theoretical curve to include large values of $\alpha$ particularly to demonstrate that for $\alpha > 50\%$ there is no quantum advantage.

FIG. 3. Temporal quantum noise reduction $\sigma$ as a function of absorption $\alpha$ for the bright squeezed light (red dots) and coherent light (blue dots). Solid lines are the theoretical predictions. The quantum advantage is only significant for small values of $\alpha$, so the data was taken only for faint absorption levels. For the virtue of completeness we plot the theoretical curve to include large values of $\alpha$ particularly to demonstrate that for $\alpha > 50\%$ there is no quantum advantage.
FIG. 4. Quantum advantage as a function of absorption $\alpha$. Solid red line is the theoretical prediction.

The final operators before the EMCCD camera can therefore be expressed as

$$\hat{a}_f = \sqrt{1 - \alpha} \left( \sqrt{\eta_a} (\cosh \nu_a) \hat{a} + e^{i \phi} (\sinh \nu_a) \hat{b} \right) + i \sqrt{1 - \eta_a} \nu_a \hat{a}_a$$

$$\hat{b}_f = \sqrt{\eta_b} (\cosh \nu_b) \hat{b} + e^{-i \phi} (\sinh \nu_b) \hat{a} - i \sqrt{1 - \eta_b} \nu_b \hat{b}_b,$$

where $r$ is the squeezing parameter of the FWM, $\theta$ is the relative phase between the twin beams (approximately, $\theta \approx 2 \pi \nu_{HF} \times L/c$, where $2 \nu_{HF}$ is the frequency difference between the twin beams and $\nu_{HF}$ is the hyperfine splitting in the electronic ground state of $^{85}$Rb shown in Fig. 3(b)), $L$ is the vapor cell length and $c$ is the speed of light), $1 - \eta_a$ and $1 - \eta_b$ are the optical losses including imperfect detection quantum efficiencies on the probe and conjugate beam paths respectively, $\alpha$ is the absorption we are interested in measuring, and $\nu_a$, $\nu_b$ and $\nu$ are the vacuum/noise operators. When a coherent state $|\beta\rangle$, $\beta = |\beta|e^{i \phi}$, where $\phi$ is the input phase, seeds mode $a$, and only vacuum fluctuations $|0\rangle$ seed mode $b$, then the input state can be written as $|\beta, 0, 0, 0, 0\rangle$, where the third, fourth and fifth zeros are the inputs for the vacuum/noise operators $\nu_a$, $\nu_b$ and $\nu$ respectively. Although not trivial, it is fairly straightforward to calculate the number operators $\hat{N}_a = \hat{a}^\dagger \hat{a}_f$ and $\hat{N}_b = \hat{b}^\dagger \hat{b}_f$ for the probe and conjugate beams before the EMCCD camera. Since the sample is placed in the probe beam, and the conjugate beam is used as a reference, we adopt the photon counts difference $\langle \hat{N}_a \rangle = \langle \hat{N}_a - \hat{N}_b \rangle$ as our measurement signal. Note that this double-beam approach is commonly implemented in imaging and spectroscopy applications involving weak absorptions [10 20], because it enables the cancellation of classical super-Poissonian noise and provides a direct measurement of the absorption by instantaneous comparison with the unperturbed reference beam. The measurement sensitivity,

$$\Delta \alpha = \sqrt{\frac{\langle \Delta^2 \hat{N}_a \rangle}{\partial_\alpha \langle N_\alpha \rangle}},$$

can then be readily obtained. We define the quantum advantage in this letter as the ratio of the sensitivity enabled by the squeezed light, $\Delta \alpha_{sqz}$, to the one acquired from the coherent light, $\Delta \alpha_{snl}$, with the same amount of average photon numbers $\langle N_a \rangle$ and $\langle N_b \rangle$ as the bright twin beams:

$$\text{Quantum Advantage [dB]} = 10 \times \log_{10} \frac{\Delta \alpha_{sqz}}{\Delta \alpha_{snl}}$$

$$= 10 \times \log_{10} \sqrt{\frac{\langle \Delta^2 \hat{N}_a \rangle_{snl}}{\langle \Delta^2 \hat{N}_a \rangle_{sqz}}} = 10 \times \log_{10} \sqrt{\frac{2T}{\sigma}}.$$  

The theoretical predictions for the temporal quantum noise reduction characterization $\sigma$ and the quantum advantage as a function of absorption $\alpha$ are plotted in Fig. 3 and Fig. 4 respectively as solid lines, where excellent agreements between experiment and theory can be seen.

In conclusion, our experiment realizes a practical scheme that allows the SNL in the direct absorption measurement to be overcome. We demonstrate that by using the bright squeezed light more than 1 dB quantum advantage is achieved for the measurement sensitivity at faint absorption levels. We thus experimentally demonstrate the advantage of quantum light for measurements on open systems. We use FWM in an atomic $^{85}$Rb vapor cell to generate the quantum-correlated twin beams of light. It is also the first experiment that uses quantum light generated with FWM instead of SPDC to demonstrate a sub-shot-noise absorption measurement. Major advantages of this FWM-based quantum light generation scheme include an ultra-high photon-pair flux up to $10^{16}$ photons/s, which is a few orders of magnitude higher than the fluxes produced by SPDs [31 43], and narrowband probe and conjugate beams ($\sim 20$ MHz) [38 40], which can be readily integrated into quantum networks through coupling with micro-resonators/cavities. Moreover, the FWM process offers sufficient gains in a single-pass configuration producing bright quantum-correlated beams of light without a cavity, making it possible to preserve the multi-spatial-mode nature of the bright twin beams [44 45]. Our quantum light generation together with the direct absorption measurement scheme reported here can be therefore greatly beneficial to many applications involving characterizing chemical and biological samples, where the sub-SNL absorption measurements are highly desirable [40 47].

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In this work an external cavity diode laser and a tapered amplifier (a combo system manufactured by Topica Photonics with item number TA Pro 795) is used as the laser source with a typical linewidth of 100 kHz (5 μs), to generate a strong (∼ 400 – 800 mW) pump beam near the D1 line of Rb (795 nm). A weak seed beam is diverted from the pump and ∼ 3 GHz red-detuned
by double-passing an 1.5 GHz acousto-optic modulator (AOM1 in Fig. 5(a)) (Brimrose TEF-1500-100-795 driven by a RF synthesizer manufactured by Hewlett-Packard with item number 8642B). This results in a very good relative phase stability of the seed with respect to the pump. The pump and seed beams are combined in a Glan-Taylor polarizer and directed at an angle of 0.3° to each other into a 12.5 mm long vapor cell filled with isotopically pure $^{85}\text{Rb}$ (Precision Glassblowing TG-ABRB-185-Q) (see Fig. 5(a)). The pump and seed beams are collimated with waists at the position of the cell center of 700 µm and 400 µm $1/e^2$ radius, respectively. The cell, with no magnetic shielding, is heated to 112°C. The windows of the cell are antireflection coated on both faces, resulting in a transmission for the seed beam of ~98% per window.

After the cell, the pump and the twin beams (‘probe’ and ‘conjugate’) are separated by a second polarizer, with $\sim 2 \times 10^5 : 1$ extinction ratio for the pump. The pump at frequency $\nu_p$ is blue-detuned by a ‘one-photon detuning $\Delta$’ of 900 MHz with respect to the $^{85}\text{Rb}$ $5S_{1/2}, F = 2 \rightarrow 5P_{1/2}$, D1 transition (see Fig. 5(b)). The seed at frequency $\nu_s$ is red-detuned from the pump by (3036 MHz + $\delta$), where $\delta$ is the ‘two-photon detuning’ and typically a few MHz, which can be adjusted by changing the radio frequency that drives the 1.5 GHz AOM. These detunings result in an intensity gain on the seed of 4.5, and the resulting beam is referred to as the ‘probe’ beam. The gain is accompanied by the generation of a ‘conjugate’ beam at frequency $\nu_c$, blue-detuned from the pump by (3036 MHz + $\delta$). It has the same polarization as the probe beam, and propagates at the pump-seed angle on the other side of the pump so that it satisfies the phase-matching condition.

**B. Squeezing measurement**

To measure the squeezing between the twin beams, after the second polarizer the probe and conjugate beams are directed into the two ports of a balanced, amplified photodetector with a transimpedance gain of $10^5$ V/A and 94% quantum efficiency at $\lambda = 795$ nm. The photodetector signals are sent to a radio frequency spectrum analyzer with a resolution bandwidth RBW of 300 kHz and a video bandwidth VBW of 100 Hz (not shown in Fig. 5(a)).

A typical squeezing spectrum is shown in Fig. 5(c). The standard quantum limit (blue curve) of this system is measured by picking off the probe before the cell, splitting it with a 50/50 non-polarizing beam splitter, and directing the resulting beams into the balanced, amplified photodetector. The balanced detection technique subtracts away common-mode noise to better than 25 dB. The balanced photodetector noise level is a measure of the standard quantum limit for the total amount of optical power arriving at the photodetector. The standard quantum limit should be independent of frequency, which is indeed the case within the bandwidth of the detection electronics, which begins to drop down above 3 MHz.

**C. EMCCD camera settings**

Since our pulse duration is 2 µs and the time interval between two consecutive pulses is 24 µs, thus in order to completely transfer all charges from the camera’s image area to the storage area within one pulse cycle, we can in principle choose to set the speed of vertical pixel shift (i.e., the time taken to vertically shift all pixels one row down) to any value as long as it is faster than 4 µs, given our beam size is merely 3 pixels across. However, the
drawback with a fast vertical pixel shift speed is the re-
duction of charge transfer efficiency, which in turn causes
‘vertical smearing’ (i.e., light is still falling on the image
area during the short time taken to transfer the charge
from the image area to the storage area). In our case, we
found a $0.9 \, \mu s$ vertical pixel shift speed in conjunction
with a vertical clock voltage amplitude of 4 (to ensure
that extremely high signals can be fully removed during
the EMCCD clean cycle) worked best for us.

Another important setting of the EMCCD is the read-
out rate. It also ought to be fast enough to be within
one pulse cycle. However, a faster readout rate always
results in a higher readout noise. In our case, we adopt
3 MHz as our readout rate although technically it can be
as fast as 17 MHz, but the price one has to pay is 8-fold
more readout noise.