Quantum-Enhanced Continuous-Wave Stimulated Brillouin Scattering Spectroscopy and Imaging

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Brillouin spectroscopy and microscopy is an emerging label-free imaging technique to assess local viscoelastic properties. Quantum enhanced stimulated Brillouin scattering is demonstrated for the first time using low power continuous-wave lasers at 795 nm. A signal to noise ratio enhancement of 3.4 dB is reported by using two-mode intensity-difference squeezed light generated with four-wave mixing process in atomic rubidium vapor. The low optical power and the excitation wavelengths in the water transparency window has the potential to provide a powerful bio-imaging technique for probing mechanical properties of biological samples of cells and tissues prone to phototoxicity and thermal effects. The performance enhancement affordable through the use of quantum light may pave the way for significantly improved sensitivity, data acquisition rate and spatial resolution. The proposed new way of utilizing squeezed light for enhanced stimulated Brillouin scattering can be easily adapted for both spectroscopic and imaging applications in materials science and biology.

Over the past decade, Brillouin scattering spectroscopy and microscopy has witnessed its renaissance providing solutions to fundamental problems and sparking new applications across multiple disciplines. In the innermost part of those revolutionary advancements is the new ways of improving detection either through high-resolution spectrometer or through nonlinear optical excitation. Brillouin scattering is an inelastic scattering of light by electrostrictively or thermally excited acoustic waves. If a narrow line (≤ 10 MHz) light source is used, both the red-shifted (Stokes) and blue-shifted (anti-Stokes) scattered light is detected giving rise to a Brillouin spectrum. By measuring both the frequency shift and the line width of the spectrum, the complex viscoelastic modulus of the sample can be assessed in a single spectroscopic measurement. Traditional applications of Brillouin scattering involved characterization of mechanical properties of solid-state materials, optical fibers and gems and involved spontaneous Brillouin scattering, which is based on detecting the spectrum of scattered photons. Only relatively recently, biological applications of Brillouin spectroscopy became a subject of interest. Recent years brought a deeper understanding of microscopic biomechanics as one of the key governing factors in biological development and diseases such as cancers and progression. Brillouin scattering spectroscopy offers a non-contact, label free method, it is therefore very suited for measurements of biomechanical properties that would be difficult to measure with other methods.

With all the advantages of Brillouin spectroscopy being able to provide unique information in a remote and noninvasive way, there is still a tremendous amount of remaining challenges to improve the accuracy and acquisition speed of such measurements, in order to observe fast dynamic processes and to image large scale objects with microscopic spatial resolution. Stimulated Brillouin scattering (SBS) provides a way of enhancing the signal, which was first observed by Chiao et al. utilizes two beams of light overlapping in space and time in the focal volume. Spontaneous Brillouin scattering becomes SBS when a second counter propagating beam, which is slightly detuned in frequency, causes beating between the light fields and enhances the acoustic wave whenever the frequency detuning between the two beams coincides with the acoustic resonance of one of the acoustic modes supported by the geometry and composition of the sample (see the phase-matching diagram in Fig. ). The efficiency of phonon generation in this case is proportional to the intensity of the stronger one of these two beams, and hence the Brillouin scattering could be orders of magnitude stronger than in the scenario of spontaneous Brillouin scattering. High scattered signal magnitude translates into better signal-to-noise ratio (SNR) of stimulated versus the spontaneous techniques, and consequently faster acquisition times. There are also some technical challenges associated with the use of SBS. The spectral range of Brillouin scattering, which is typically between a few to a few tens of GHz with sub-GHz line-widths due to microseconds long phonon lifetimes, determines the choice of laser suitable for probing the photon-phonon interactions. The spectral width of the laser emission should be well below the Brillouin line-width to avoid spectral broadening of acoustic resonances and maximize inelastic scattering. The frequency stability and noise of the laser are equally important for reliable detection of Brillouin signals and sufficient SNR. Frequency locking to an external reference, such as a cavity or an absorption line, is often required to reduce temporal drifts of the laser.
frequency [6] [17]. Apart from the above considerations, the ultimate choice of the laser is also determined by the sample "transparency window" [18]. In addition to the concern of light source, the alignment of the two counter-propagating beams must be very precise for a significant spatial overlap of their individual focal regions (i.e., Raleigh ranges).

Most studies and applications of SBS were conducted in optical fibers, where signals are intrinsically strong due to large interaction length and high optical damage threshold for most of the glasses used for fiber fabrication. However, as any nonlinear optical technique, coherent Brillouin spectroscopy benefits from the higher excitation intensity, which can induce phototoxicity and/or thermal damage to biological samples of interest. Clearly, there is a tremendous need to improve SBS detection for low-power-light applications, and recent advancements in the most sensitive instrument, LIGO, demonstrate a path to improve the detection limit using squeezed light spectroscopy [19] [20]. Since photon shot-noise is a fundamental limit for optical detection, it is somewhat intuitive to deploy the strategy utilizing quantum light to beat this limitation. In fact, applications and advantage of quantum light generation scheme are strong intensity-difference squeezing (greater than 6 dB) and narrow-band twin beams (~10 MHz) [31] [32], which is extremely beneficial for the intended SBS experiment, where the spectral width of the light source must be well below the Brillouin line width, which is typically a few hundreds of MHz. The SNR for the twin beams, with signal defined as the difference of photon numbers in the twin beams, is better than that for coherent beams by a factor of \( \cosh 2 \gamma \), where \( \gamma \) is the well known squeezing parameter used to characterize the two-mode squeezed state [16]. This improvement in SNR consequently translates to the quantum advantage in the SBS spectroscopy (See Eq. (10) in Ref. [33]).

The schematic of our experimental setup is shown in Fig. 1(a). Two quantum-correlated beams of light, i.e., the "probe" and "conjugate" beams, are produced with the FWM process in the \(^{85}\text{Rb} \) vapor cell. After the cell, the probe beam is overlapped with a counter-propagating laser beam (shown in Fig. 1(a) as "Pump 2") at a homemade sample holder filled with distilled water, to form a phase-matching geometry for the SBS process depicted in Fig. 1(b). The conjugate beam serves as a reference, and two flip mirrors (FM) are used for the introduction of two coherent beams so that the whole setup can be converted to a classical version. The pumps and probe beams are amplitude-modulated by three acousto-optic modulators (AOMs) at 300 KHz (AOM1) and 400 KHz (AOM2&3) respectively. The water SBS signal therefore is expected to appear at 700 KHz where the two-mode squeezing is expected to be the best [25] [34]. Both lasers (the "Pump 1" and "Pump 2") are locked to external cavities so that the relative frequency between them can be scanned. Other experimental details can be found in Ref. [33].

We start with classically characterizing the SBS signal of water. Instead of using the twin beams produced by the FWM process, we use two coherent beams (by flipping up the two flip mirrors shown in Fig. 1(a)) for this classical measurement. Figure 2(a) shows a SBS spectrum of distilled \( \text{H}_{2}\text{O} (T = 25 \degree \text{C}) \) from a lock-in amplifier at one spot on the sample. The time constant of the lock-in amplifier is set at 300 ms. The coherent beam in the pathway of the probe beam (i.e., the coherent probe) is locked while the pump beam of the SBS process ("Pump 2" in Fig. 1(a)) is scanned with scan frequency of 0.02 Hz. By the time the coherent beam and pump beam reach the sample, the average power are 300 \( \mu \)W and 36 mW respectively. From Fig. 2(a) the Brillouin shift and linewidth are measured to be \( \Omega_B / 2\pi = 5.01 \) GHz and \( \Gamma / 2\pi = 292 \) MHz, which are in good agreement with previous experiments [19]. The dip on the left

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FIG. 1. (a) Experimental setup. See text for a detailed description. L: lens, FM: flip mirror, TS: telescope, PBS: polarizing beam splitter, BD: balanced detector, SA: RF spectrum analyser. (b) Phase-matching diagram for the SBS process [10]. The wave-vectors and frequencies for the pump, probe and sound wave are denoted by \( (\vec{k}, \omega) \), \( (\vec{k}', \omega') \) and \( (\vec{q}, \omega_B) \) respectively.

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medium possesses a large third-order electric susceptibility \( \chi^{(3)} \), and when appropriately chosen laser light (at 795 nm, near \(^{85}\text{Rb} \) D\(_1\) line) ‘seeds’ the medium, ‘twin beams’, also known as the ‘probe’ and ‘conjugate’ beams, are produced. Major advantages of this FWM-based quantum light generation scheme are strong intensity-difference squeezing (greater than 6 dB) and narrow-band twin beams (~10 MHz) [31] [32], which is extremely beneficial for the intended SBS experiment, where the spectral width of the light source must be well below the Brillouin line width, which is typically a few hundreds of MHz. The SNR for the twin beams, with signal defined as the difference of photon numbers in the twin beams, is better than that for coherent beams by a factor of \( \cosh 2 \gamma \), where \( \gamma \) is the well known squeezing parameter used to characterize the two-mode squeezed state [16]. This improvement in SNR consequently translates to the quantum advantage in the SBS spectroscopy (See Eq. (10) in Ref. [33]).
(at $\sim -5$ GHz) and peak on the right (at $\sim 5$ GHz) of zero are the stimulated Brillouin loss and gain peaks respectively. The center feature is caused by absorptive stimulated Rayleigh scattering. When we change the power of the coherent probe from 150 $\mu$W to 750 $\mu$W while keeping pump power at 36 mW, as shown in Fig. 2(b), we clearly observe the expected linear dependency between the SBS signal (at the peak of gain) and the optical power of the coherent probe $P_c$. Estimation of the SBS signal magnitude can be found in Ref. 33.

Having characterized the classical SBS process in water, in the following we demonstrate the quantum-enhanced SBS spectroscopy of water. To clearly demonstrate quantum-improved performance beyond the classical approach, we conducted the experiment both with the probe beam in a coherent state and in the two-mode squeezed state. The experimental scheme can be easily swapped between the two operations simply by flipping up and down the two flip mirrors depicted in Fig. 1(a). Figure 3 presents our experimental results for the quantum-enhanced SBS spectroscopy of water. In order to acquire the spectra, both lasers are locked so that their frequency difference matches the Brillouin shift of water as a function of the pump beam power below the shot-noise level. The green and black traces correspond to the configuration where the probe beam is in a coherent state while the blue traces correspond to the two beams being in a two-mode squeezed state with -3.40 dB noise reduction below the shot-noise level. The green and black traces correspond to the realization where the two lasers are locked outside of the SBS gain profile of water. All traces are normalized to the shot-noise level.

Loiusin signals even for CW pump powers less than 8 mW. This is extremely beneficial when studying delicate and fragile biological samples where excessive optical power might damage the sample.

We also plot in Fig. 4(a) the SNR of the peak of Brillouin shift of water as a function of the pump beam power (in dBm) for the cases where the probe beam is in a coherent state (red circles) and in a two-mode squeezed state (blue squares). The probe beam power is kept at 750 $\mu$W. The error bars correspond to one standard deviation. From the two fits we see two nice linear dependence of the SBS signal on the pump power with slope of 1.99 and 2.04, which matches our expectation of 2. Also notice that, the average noise suppression (in dB) below the shot-noise level can be calculated from the fitting parameters as $17.59 - 14.23 = 3.36$ dB. This is the quantum advantage of the two-mode squeezed light over coherent light in SNR for the SBS spectroscopy.

This $\sim 3.40$ dB quantum advantage can be calculated with a theoretical framework assuming that both of the twin beams are in single modes 35. In brief, we designate $\hat{a}$ and $\hat{b}$ as the mode operators for the probe and conjugate beams respectively, then the input-output relations for the FWM and SBS processes can be expressed as

$$\hat{a}_{\text{FWM}} = (\cosh r)\hat{a} + (\sinh r)\hat{b}^\dagger$$

and

$$\hat{a}_{\text{SBS}} = g\hat{a}\hat{b}_{\text{FWM}} + (\sqrt{g^2 - 1})\hat{n}_B^\dagger$$

respectively, where $r$ and $g$ are the FWM squeezing parameter and the SBS gain parameter (note that the SBS intensity gain $G = g^2$) respectively, and $\hat{n}_B$ is a vacuum noise operator introduced by the SBS gain process. All optical and atomic absorption losses sustained by the twin beams are included (see Ref. 33 for a detailed derivation). The theoretical quantum advantage (i.e., the improvement in SNR) as a function of the SBS gain related parameter $\xi = G - 1$ is shown in Fig. 4(b) as the red curve. Since our $\xi$ is in the range of...
10^{-6} to 10^{-5}$ MHz (within the region highlighted by the gray bar), therefore the measured $\sim 3.40$ dB quantum advantage agrees very well with our theoretical prediction. As a comparison, we also plot the theoretical curve without loss in blue in Fig. 4(b) to show the effect of the SBS gain on the quantum advantage. We see from these two curves that, if there is no loss, the SBS gain would only degrade the quantum advantage as the gain process itself introduces noise, however with loss present, the SBS gain might instead improve the degradation of quantum advantage due to the competition between the gain and loss.

The SBS spectrum of water acquired by a lock-in amplifier shown in Fig. 2(a) can also be attained using the RF spectrum analyser. The results are shown in Fig. 5. This requires manually changing the locking point of the pump laser while keeping the locking point of probe laser fixed so that the frequency difference between the two can be scanned. All data points are obtained by normalizing the water SBS signal at 700 KHz to the shot-noise level, and error bars correspond to one standard deviation. The two laser beam powers are the same as in Fig. 3(a). A quantum advantage of $\sim 3.40$ dB can be clearly seen from these two spectra. It is also worth pointing out that the classical approach is not able to detect the SBS loss dip at $-5$ GHz as small differential absorption of two coherent beams would always be at the shot-noise level.

As the final step, we demonstrate that our scheme can also be utilized for microscopic imaging. We use the SBS signal of water to acquire a 2-Dimensional image of a piece of triangle shaped glass, as shown in the inset of Fig. 6(a). The pump and probe powers are 7.5 mW and 750 $\mu$W respectively (further image acquisition details can be found in Ref. [33]). Pixels in Fig. 6(a) and Fig. 6(b) are registered with the probe beam being in a coherent state and the two-mode squeezed state respectively. Obviously, the image contrast (i.e., the SNR) for the glass triangle in Fig. 6(a) is unappreciable due to the coherent light induced SBS signal of water is overwhelmed by the shot noise (see the red curve in Fig. 3(b)). By using the two-mode squeezed light, however, a clear image contrast of more than 3 dB for the glass triangle is obtained in Fig. 6(b) (see the blue curve in Fig. 3(b)). It is also important to notice that each pixel in Fig. 6 takes $\sim 2$ s to obtain, which is limited by the sweep time of the RF spectrum analyzer, and is an order of magnitude faster than the $\sim 1$ min acquisition time reported in Ref. [10] using a lock-in amplifier.

In conclusion, we demonstrated a quantum-enhanced continuous-wave SBS spectroscopy and imaging system. As a proof-of-principle, we acquired a SBS spectrum of water and a 2-dimensional microscopic image using water as the signal medium with quantum enhanced SNR of $\sim 3.4$ dB. The quantum enhancement is achieved by using the two-mode intensity-difference squeezed light with a spectral width in the range of 10 MHz generated by the FWM process in atomic $^{85}$Rb vapor. It is very important to note that, it is this unique narrow-band feature of our squeezed light that makes the quantum-enhanced SBS spectroscopy and imaging system possible, as for the SBS

FIG. 4. (a) The SNR of the peak of Brillouin shift of water as a function of the pump beam power (in dBm). The red circles and dot-dashed fit line correspond to the probe beam being in a coherent state, while the blue squares and solid fit line correspond to the beam being in a two-mode squeezed state. (b) Theoretical prediction for the quantum advantage as a function of the SBS gain related parameter $\xi = G - 1$. The red curve is plotted with experimental conditions, while the blue curve is plotted with no loss present.

FIG. 5. SBS spectrum of water acquired using a RF spectrum analyser by manually scanning the locking frequency of the pump laser.

FIG. 6. Quantum enhanced microscopic imaging using water as the signal medium. The imaging object is a triangle-shaped piece of glass shown in the inset of (a) where the white scale bar is 1 mm in horizontal direction. More than 3 dB quantum-enhanced SNR, or image contrast, is clearly visible in (b).
process to occur the spectral width of the light source must be well below the Brillouin line width (≈ 300 MHz in this work). The low optical power (can be < 8 mW) and the excitation wavelengths in the water transparency window used in this work has made our system very applicable for probing mechanical properties of biological samples, which will be the subject of our future study.

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The atomic medium is pumped by a strong (\(\sim 500 \text{ mW}\)) narrow-band continuous-wave (CW) laser (shown in Fig. 7(a) as “Pump 1”) at frequency \(\nu_1 (\lambda = 795 \text{ nm})\) with a typical linewidth \(\Delta \nu_1 < 1 \text{ MHz}\). Applying an additional weak (in the range of a few hundreds \(\mu\text{W}\)) coherent beam (shown in Fig. 7(a) as “Seed”) 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 (\(\delta = 5 \text{ MHz}\) in this work) respectively in Fig. 7(b) (further experimental details can be found in Ref. [25]), 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 \(2\nu_p = \nu_p + \nu_c\) (see the level structure in Fig. 7(b)). The resulting twin beams are strongly quantum-correlated and are also referred to as bright two-mode squeezed light [30]. The twin beams exhibit an intensity-difference squeezing of 6.5 dB measured by balanced photodiodes (further squeezing measurement details can be found in Ref. [25]), which is indicative of strong quantum correlations [30].

After the \(^{85}\text{Rb}\) vapor cell, the pump and the twin beams (shown in Fig. 7(a) as “Probe” and “Conj.”) are separated by a Glan-Laser polarizer, with \(\sim 2 \times 10^9 : 1\) extinction ratio for the pump. The probe beam then passes through a telescope (TS) with an enlarged beam waist (\(\sim 3 \text{ mm}\)) before focused (down to a \(1/e^2\) beam waist of \(\sim 5 \mu\text{m}\) by a plano-convex lens with focal length \(f = 16 \text{ mm}\)) and overlapped with a counter propagating laser beam (shown in Fig. 7(a) as “Pump 2”), a same type of CW laser as “Pump 1”, and having a \(1/e^2\) beam waist of \(\sim 6 \mu\text{m}\) at a homemade sample holder filled with distilled water, to form a phase-matching geometry for the SBS process in water depicted in Fig. 7(c). The sample holder consists of two glass microscope slides separated by 1 mm with water filled between them. Both \(\lambda/2\) and \(\lambda/4\) waveplates are added in the probe beam path in order for the probe beam to be reflected as much as possible by the polarizing beam splitter (PBS) into one port of the balanced detector (BD). Therefore in this configuration, the probe beam is linearly polarized while the pump beam for the SBS process (“Pump 2” in Fig. 7(a)) is circularly polarized. The conjugate beam serves as a reference, and two flip mirrors (FM) are used for the introduction of two coherent beams so that the whole setup can be converted into a classical version. The pumps and probe beams are amplitude-modulated by three AOMs at 300 KHz (AOM1) and 400 KHz (AOM2/3) respectively. The water SBS signal therefore is expected to appear at 700 KHz where the two-mode squeezing is expected to be the best [25] [34].

In addition to the components shown in Fig. 7(a), there are also frequency-locking optics and electronics for the probe and pump beams of the SBS process so that they can be locked and separated by the phonon frequency \(\Omega_B/2\pi\) (or so-called “Brillouin frequency shift”) of water, which is in the range of \(\sim 5 \text{ GHz}\) [10]. In this work, we use fringes from a room temperature Fabry-Perot cavity as the locking error signal and absorption lines from a
room temperature natural abundant Rb cell as the locking reference for each laser beam. We change the frequency difference between the two beams by fixing the probe frequency (at one photon detuning $\Delta \sim 1$ GHz shown in Fig. 7(b)) so that the FWM process can yield the best two-mode squeezing, while scanning the locking frequency of the pump with a minimal step of 30 MHz (determined by the resolution of the scanning voltage).

Microscopic Imaging Acquisition

The triangle-shaped glass used for imaging shown in the inset of Fig. 6(a) was made by cutting off a corner of a microscope slide whose thickness is 1 mm, and the lengths of the triangle’s two sides are 7.5 mm and 6.5 mm respectively. Since our homemade sample holder consists of two glass microscope slides separated by 1 mm with water filled between them, the glass triangle therefore can be introduced into the sample holder so that there is no water content within the area of the triangle.

To acquire the images in Fig. 6 in the main text, we use two translational stages with differential micrometer screws to automatically move the sample holder’s position with a spatial scan step size of 100 $\mu$m in both directions. The images are obtained by scanning each pixel under the experimental conditions shown in Fig. 3(b). Namely, the pump and probe powers are 7.5 mW and 750 $\mu$W respectively, and the two lasers are locked so that their frequency difference matches the 5 GHz Brillouin shift of water.

THEORETICAL FRAMEWORK

We use a single-mode quantum-mechanical model to simulate the experiment [35]. We denote the optical field operators for the probe and conjugate modes as $\hat{a}_{0,f}$ and $\hat{b}_{0,f}$ with subscripts 0 and $f$ labeling the operators at the initial and final stages of transformation, respectively. The input-output relation for the FWM process, $\hat{a}_{\text{FWM}} = (\cosh r)\hat{a} + (\sinh r)\hat{b}$, where $r$ is the squeezing operator, is well known. For the input-output relation for the SBS process, we can write it as $\hat{a}_{\text{SBS}} = g\hat{a}_{\text{FWM}} + \hat{f}$, where $g$ is the SBS gain parameter, and $\hat{f}$ is the noise operator introduced by the SBS gain process. The field operator $\hat{a}_{\text{SBS}}$ must satisfy the commutation relation $[\hat{a}_{\text{SBS}}, \hat{a}_{\text{SBS}}^\dagger] = 1$, from which the noise operator $\hat{f}$ can be derived as $\sqrt{g^2 - 1}\hat{\nu}_B$, where $\hat{\nu}_B$ is a vacuum noise operator introduced by the SBS process. All optical and atomic absorption losses sustained by the twin beams are modeled by three beams splitters with transmission $\eta_P$, $\eta_B$ and $\eta_c$ [29]. They represent the atomic and optical loss in the probe pathway between the FWM cell and the SBS sample holder ($\eta_P$), between the SBS sample holder and the balanced detector ($\eta_B$), and the optical loss in the conjugate pathway ($\eta_c$), respectively. We treat all pump beams classically. The field operators for the probe and conjugate modes as $\hat{a}_1$, $\hat{b}_1$, $\hat{a}_f$, $\hat{b}_f$ are defined by

$$\hat{V}_0 = \begin{pmatrix} \hat{a}_0 \\ \hat{b}_0 \\ \hat{\nu}_0 \end{pmatrix} \quad \text{and} \quad \hat{V}_f = \begin{pmatrix} \hat{a}_f \\ \hat{b}_f \\ \hat{\nu}_f \end{pmatrix}. \quad (1)$$

The experiment can then be described by the transformation of field operators

$$\hat{V}_f = \hat{T}_2 \cdot \left( B \cdot \left( \hat{F} \cdot \hat{V}_0 + \hat{\nu}_B \right) + \hat{L}_B \right) + \hat{L}_2, \quad (2)$$

where

$$F = \begin{pmatrix} \cosh r & 0 & 0 & \sinh r \\ 0 & \cosh r & \sinh r & 0 \\ 0 & \sinh r & \cosh r & 0 \\ \sinh r & 0 & 0 & \cosh r \end{pmatrix}, \quad (3)$$

$$B = \begin{pmatrix} g & 0 & 0 & 0 \\ 0 & g & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}, \quad (4)$$

and

$$\hat{L}_B = \begin{pmatrix} \sqrt{g^2 - 1} \hat{\nu}_B \\ 0 \\ 0 \end{pmatrix}. \quad (5)$$

Matrix $F$ describes the FWM process, while matrix $B$ together with vector $L_B$ describe the SBS process. Matrices $T_1$ and $T_2$ describe the transmission of the beam splitters, and vectors $\hat{L}_1$ and $\hat{L}_2$ contain the field operators $\hat{\nu}_B$ and $\hat{\nu}_c$ for the vacuum noise coupled in by optical losses:

$$T_1 = \begin{pmatrix} \sqrt{\eta_P} & 0 & 0 & 0 \\ 0 & \sqrt{\eta_P} & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}, \quad (6)$$

$$T_2 = \begin{pmatrix} \sqrt{\eta_B} & 0 & 0 & 0 \\ 0 & \sqrt{\eta_B} & 0 & 0 \\ 0 & 0 & \sqrt{\eta_c} & 0 \\ 0 & 0 & 0 & \sqrt{\eta_c} \end{pmatrix}, \quad (7)$$

$$\hat{L}_1 = \begin{pmatrix} i\sqrt{1 - \eta_P} \hat{\nu}_B \hat{\nu}_P \\ -i\sqrt{1 - \eta_P} \hat{\nu}_B \hat{\nu}_P \\ 0 \\ 0 \end{pmatrix}. \quad (8)$$
\[
L_2 = \begin{pmatrix}
i \sqrt{1 - \eta_p \mu_p} \\
-i \sqrt{1 - \eta_b \mu_b} \\
i \sqrt{1 - \eta_c \mu_c}
\end{pmatrix}.
\]

When a coherent state \(|\beta\rangle\), \(\beta = |\beta| e^{i\phi}\), where \(\phi\) is the input phase, sends 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 last four zeros are inputs for the vacuum/noise operators \(\hat{\nu}, \hat{\mu}, \hat{\mu}_c\) respectively. Although not trivial, it is fairly straightforward to calculate the number operators \(\hat{n}_a = \hat{a}_f \hat{a}_f^\dagger\) and \(\hat{n}_b = \hat{b}_f \hat{b}_f^\dagger\) for the probe and conjugate modes after detection, and the expectation values of quantities such as the noise suppression below the shot noise level, i.e. the quantum advantage:

Quantum Advantage [dB] = \(-10 \times \log_{10} \left[ \frac{\Delta^2(\hat{n}_a - \hat{n}_b)}{\Delta^2 SNL} \right] \),

(10)

where \(\Delta^2 SNL\) is the shot noise level, which is defined as the variance of the intensity difference of two coherent beams having the same intensities as the measured probe and conjugate beams, therefore in our case \(\Delta^2 \hat{\eta}_{SNL} = \langle \hat{n}_a \rangle + \langle \hat{n}_b \rangle\). With the measured FWM gain and optical losses, theoretical curves shown in Fig. 4(b) in the main text can be thus readily plotted.

**SBS INTENSITY GAIN PARAMETER \(\xi\) AND SIGNAL MAGNITUDE ESTIMATION**

The SBS signal after interaction length \(L\) can be written as

\[
I_{out} = I_{probe} \cdot e^{g_0 I_{pump} L},
\]

(11)

where \(g_0\) is the maximal SBS gain [10]. In our experiment, \(g_0 = 0.048\) m/GW [16], \(L \approx 2 z_R \approx 70\) \(\mu m\), where \(z_R\) is the Rayleigh range as the signal comes almost entirely from the region where the intensity is the largest, and \(I_{pump} \sim 1.2\) GW/m\(^2\) given the experimentally achievable maximal pump power of 36 mW and \(1/e^2\) beam waist of 6 \(\mu m\) at the focal point. This yields \(g_0 I_{pump} L = 4.2 \times 10^{-6}\). Therefore

\[
I_{out} = I_{probe} \cdot e^{4.2 \times 10^{-6}} \cong (1 + 4.2 \times 10^{-6}) \cdot I_{probe} = G_{SBS} \cdot I_{probe}.
\]

(12)

Since we define in the main text that \(\xi = G_{SBS} - 1\), thus \(\xi = 4.2 \times 10^{-6}\), which is within the range indicated by the gray bar in Fig. 4(b) in the main text.

Since \(I_{out} = (1 + 4.2 \times 10^{-6}) \cdot I_{probe} = I_{probe} + 4.2 \times 10^{-6}\), \(I_{probe} = I_{common\ mode} + I_{signal}\), by using a balanced detector, the common mode intensity \(I_{probe}\) is rejected, therefore we see that the maximum SBS signal going into the detector is \(4.2 \times 10^{-6}\) of the total probe power (750 \(\mu W\)) going into the detector. The balanced detector has an electronic gain of \(~10^5\) V/W, therefore this SBS signal is \(\sim 4.2 \times 10^{-6} \times 750 \times 10^{-5} = 315\ \mu V\). As shown by the highest point in Fig. 2(b) in the main text, this estimation agrees with the measurement very well.

**DERIVATION OF THE RELATIONSHIP BETWEEN SNR AND THE PUMP POWER**

In this section, we derive the relationship between the SNR (in dB) and the pump power (in dBm) of the SBS process. From Eq. (11), we see that when \(g_0 I_{pump} L < 1\),

\[
I_{out} = I_{probe} \cdot e^{g_0 I_{pump} L} \cong I_{probe} \cdot (g_0 I_{pump} L)
\]

(13)

with constant \(\alpha = g_0 L\). This relationship is demonstrated in Fig. 2(b) in the main text by keeping \(I_{pump}\) unchanged. If we write noise on the SBS signal as

\[
\Delta I_{out} = \beta \cdot \sqrt{I_{probe}},
\]

(14)

where \(\beta\) is the noise factor, then for coherent excitation \(\beta_{coh} = 1\), while for squeezed excitation \(\beta_{sqz} < 1\). The SNR of this SBS signal registered by a RF spectrum analyser can thus be written as

\[
\text{SNR [dB]} = 10 \times \log_{10} \left( \frac{I_{out}^{1/2}}{\Delta I_{out}} \right)^2
\]

\[
= 10 \times \log_{10} \left( \frac{\alpha \cdot I_{probe} \cdot 1_{pump}^{1/2} \cdot \beta \cdot \sqrt{I_{probe}}}{\beta \cdot \sqrt{I_{pump}}} \right)^2
\]

\[
= 20 \times \log_{10}(I_{pump}) + 10 \times \log_{10} \left( \frac{1}{\beta} \cdot \alpha^2 I_{probe} \right).
\]

(15)

Therefore for a fixed probe power (i.e., \(\alpha^2 I_{probe}\) is a constant), the SNR has a linear dependence on \(10 \times \log_{10}(I_{pump})\) (i.e., pump power in the unit of dBm) with a slope of 2. This dependence is demonstrated in Fig. 4(a) in the main text. It is also worth noticing that information about the noise factor \(\beta\) is contained in the second term of Eq. (15). In our case, the quantum advantage of using squeezed light for the SBS spectroscopy over coherent light would be simply the difference between the two second terms,

\[
\text{Quantum Advantage [dB]} = 10 \times \log_{10} \left( \frac{1}{\beta_{sqz}^2} \cdot \alpha^2 I_{probe} \right)
\]

\[
-10 \times \log_{10} \left( \frac{1}{\beta_{coh}^2} \cdot \alpha^2 I_{probe} \right) = -20 \times \log_{10}(\beta_{sqz}),
\]

(16)

which in our case is 3.36 dB calculated from the two fits in Fig. 4(a) in the main text.