3D stochastic interferometer detects picometer deformations and minute dielectric fluctuations of its optical volume

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Speckle metrology harnesses the interferometric properties of disordered light to achieve remarkable sensitivities. Often relying on time-domain analysis, it is rate-limited by the acquisition of speckle images. In the present work instead, we use a frequency-domain approach which spans 8 to 10 frequency decades up to 100 MHz, and reveals minute changes of speckle decorrelation spectra. We built a 3D stochastic interferometer using a centimeter-sized quartz-powder cavity with arbitrary shape and high Lambertian reflectivity. Filled with a coherent monochromatic photon gas, it creates statistically isotropic and homogeneous 3D interference patterns, whose variations arise from cavity deformations or fluctuations of the dielectric tensor field inside. Speckle decorrelation depends neither on where the perturbation sits nor on where it is measured. With an average 62 m photon transit path and a finesse of 10500, cavity deformations are detected with a power noise floor of $4 \times 10^{-3}$ pm$^2$, i.e., 2.7 pm at 1 kHz. We also demonstrate a 100-fold sensitivity gain compared to conventional light scattering techniques when probing thermal motions of single and multiply scattering colloids.
light interferometry is most familiar to us as a unique tool for measuring variations of optical lengths or wavelengths, i.e. 1D optical features. From Fizeau’s experiment in 1851 on the speed of light\(^1\) or Michelson and Morley’s measurement of the relative motion of the Earth and the Luminiferous Ether in 1887\(^2,3\), to the most recent detection of gravitational waves down to typical strains of 10\(^{-21}\)\(^4\) provided by the LIGO and VIRGO observatories, optical interferometers have consistently redefined what constitutes the limits of metrology and brought major contributions to our understanding of physical laws.

Unlike these techniques which require an extremely precise geometry of the optical field, speckle metrology methods have been devised in the past decades to harness the interferometric response of strongly disordered but coherent light. While it has proven to provide exceptionally sensitive measurements, in particular for wavelength determination and spectral analysis\(^5,6\), it has also been used recently for small angles and displacements metrology\(^7\) or refractive index sensing\(^8\), and recent quantitative comparisons indicate that it can reach sensitivity levels that come close to those of interferometers using well-ordered light\(^9\).

However, and to the best of our knowledge, all speckle metrology methods are based on the time-domain analysis of speckle patterns, i.e. of the images of the speckle field, either through the analysis of spatial correlations of the interference pattern\(^10\) or even the analysis of spatial phase singularities in the field\(^11\). Consequently, time frequencies cannot be probed beyond the image acquisition rate.

In the present work, we propose instead to measure the intensity fluctuation of a single speckle grain to calculate the frequency spectrum of perturbations. By homogeneously filling an ultra-high reflectivity Lambertian cavity of arbitrary shape with a coherent and isotropic photon gas, we produce a fully developed 3D speckle field that is ergodically sensitive to minute fluctuations of its optical volume. Here, we present the design of this 3D stochastic interferometer, together with theoretical predictions of sensitivity confirmed by experimental results. We find that the frequency spectrum is sensitive to volume deformations across 8–10 frequency decades below 100 MHz, with a sensitivity better than 10\(^{-5}\)\(^3\) at 1 kHz. We also show that the interferometer can measure picometric motions of scattering objects placed inside the cavity. We show an increase in sensitivity of about 100-folds in comparison to conventional and multiple light-scattering techniques.

Results

Description of a 3D random light field. Practically, a single-frequency laser light with central wavelength \(\lambda_0 = 660\,\text{nm}\) and coherence length \(\Lambda_{\text{coh}} \approx 95\,\text{m}\) is fiber-coupled into a closed diffuse reflective cavity, with walls made of compressed quartz powder\(^10\) that provide a uniform Lambertian reflectivity with an albedo \(\mathcal{R}\) close to unity. A cylindrical shape was used with centimetric size (Fig. 1a). Thanks to a very small reflection loss coefficient \(\varepsilon = 1 - \mathcal{R} \approx 6 \pm 0.5 \times 10^{-4}\), each photon is subjected to an average number of reflections given by \(GN_0 = 1/\varepsilon \approx 1700\), which represents the path length multiplication factor or geometric gain of the empty cavity (see the “Methods”, section “Construction and optical properties”). Using the classical notion of finesses applied to our cavity and defined from the reflection losses \(\mathcal{R}\) by \(F = -2\pi/\ln(\mathcal{R})\), we obtained \(F \approx 10,500\)\(^11\). In addition, since the Lambertian reflectivity is due to multiple elastic light scattering events by the frozen disorder of the quartz powder structure of the wall, the reflection process is deterministic. However, the extreme complexity and the symmetries of diffuse reflections are such that any incident photon is reflected as an apparently “random” photon, in the sense that its wavevector is distributed according to Lambert’s law, and it has a phase and a polarization state that are uniformly distributed on \([0,2\pi]\) and on the Poincaré sphere, respectively. These statistical properties of reflected photons suggest a strong analogy with the random geometry and polarization of the photon gas emitted by the walls of a blackbody cavity.

The optical field generated at any point \(P\) inside the cavity (Fig. 1b), in the far-field relative to the scattering walls, can therefore be considered as a random field obtained by superposing a very large number of elementary plane waves independently taken from a unique statistical distribution. This unique probability distribution does not depend on the position \(r\) of point \(P\) and jointly aggregates three independent variables: a wavevector \(\mathbf{k}\) isotropically distributed on the sphere \(|\mathbf{k}| = k_0 = 2\pi/\lambda_0\), a uniform phase \(\phi\) on \([0,2\pi]\), and a polarization state represented by a complex vector \(\mathbf{d}\) uniformly distributed on the Poincaré sphere (Fig. 1d). The total field can be described as highly composite random variable constructed as the sum of a random number of independently and identically distributed plane waves \(E_\mathbf{d}(r,t)\). It writes

\[
E(r,t,\psi) = \sum_{\alpha=1}^{N_\psi} a_\alpha e^{i\phi_\alpha} d_\alpha e^{i\mathbf{k}_\alpha r - i\omega_\alpha t},
\]

where \(N_\psi\) is the number of elementary plane waves. \(\psi\) refers to one generic choice of the total random field, i.e. a microscopic configuration of the field. From the Huygens–Fresnel principle, we know that the optical field at \(P\) is determined by the field on the cavity boundary, which comes with a very large but finite number of degrees of freedom. This sets a minimal value for \(N_\psi\) in Eq. (1). A lower estimate of \(N_\psi\) is given by the number of degrees of freedom associated with wavevectors, which typically amounts to \(\Sigma_v/\Lambda^3_0 \approx 10^{10}\) where \(\Sigma_v\) is the surface area of the cavity wall. Equation (1) exactly corresponds to what is known as the 3D random wave model\(^12\), which will be referred here to as the 3D Berry field. It has been the focus of several theoretical numerical investigations but has never been used to formally describe an experimental realization of a 3D speckle field, to the best of our knowledge; nor it has been used in the context of interferometry. In our experiments, the randomness and the high statistical symmetry of the optical field arise from the linear transformation of the input laser field by the extreme but deterministic complexity of multiple scattering by the frozen disorder of the wall structure. This highly complex boundary condition act as feedback that leads to the interferometric properties of the 3D Berry field.

The phase \(\phi_\alpha\) of each field component \(E_\mathbf{d}\) is related to the phase of the laser input transformed by a random number \(n_\alpha(\mathbf{a})\) of reflections alternated with random chords across the cavity with a random travel time \(\tau_\alpha\). This leads to the notion that the field in \(P\) at time \(t\) combines the phases of the laser input taken at a large number of earlier time points \(t - \tau_\alpha\) (Fig. 1b), where the values of \(\tau_\alpha\) are the propagation times or mode lengths between the laser input point and the probe point \(P\). The statistical distribution of these propagation times is assessed from the response of the cavity to a 120 ps laser pulse (Fig. 1c), which leads to the average photon transit or residence time \(\bar{\tau}_\alpha \approx 207\,\text{ns}\), and the average photon path length \(\Lambda \approx 62\,\text{m}\). From the mean chord length theorem\(^13,14\), it is known that the mean chord length \(\Lambda_\alpha\) is given by the ratio \(\Lambda_\alpha = 4V_c/\Sigma_v\) of the cavity volume to the wall area, regardless of the shape. We obtain \(\Lambda_\alpha \approx 3.5\,\text{cm}\), the average number of reflections \(n_\alpha \approx 1700\), and finally the geometric gain mentioned above \(GN_\alpha = n_\alpha\) (see the “Methods” section “Optical setup”). These observations lead to two key remarks on the set of phases \(\{\phi_\alpha\}_\psi\). First, since the average chord length \(\Lambda\) is 8 orders of magnitude larger than the wavelength, the same is true for the
**Fig. 1 3D stochastic interferometer construct.** A Experimental setup: a single-frequency laser is injected by an optical fiber into a high reflectivity Lambertian cavity. Regardless of the cavity shape, the resulting 3D random field is fully coherent and generates a fully developed 3D speckle pattern which is probed at point P by a single mode fiber. The fiber is split, and the intensity is detected by 2 photon counters. The intensity cross-correlation is then calculated by a digital correlator. B Schematic representation of three generic path photons or modes, as a succession of diffuse reflections, between the laser input and a probe point P. C Intensity response function of a compressed quartz powder cavity to a 120 ps laser pulse at 670 nm represented in a $y$-log scale. D Coherent superposition of 3 plane waves at point P, with their respective complex polarization and Poincaré sphere representing the uniformly and randomly distributed polarizations of the plane waves interfering at point P. In particular, the polarization states of the aforementioned 3 plane waves are represented, as well as the 2D polarization state of the wave generated at point P.
standard deviation of the path length distribution, and this strongly supports our assumption that the phases \( \phi_\alpha \) are uniformly distributed on \([0, 2\pi]\). Second, the aforementioned coherence length \( \Lambda_{\text{coll}} \) of the laser exceeds the average \( \Lambda_\alpha \) by a factor of 1.5. This feature was set by design, to ensure that practically any pair of fields \( \{E_{\alpha}, E_{\beta}\} \) would be mutually coherent since 90% of the paths are shorter than \( \Lambda_\alpha \). Therefore, the 3D coherence matrix for any such pair of fields is a rank-two matrix. The coherent sum \( E_{\alpha} + E_{\beta} \), therefore, has a 2D polarization, with a plane and a state of polarization that both depend on the polarization states \( \{d_{\alpha}, d_{\beta}\} \) and the phase difference \( [\phi_{\alpha} - \phi_{\beta}] \). The total field \( E(t, r, \theta) \) is therefore fully polarized (Fig. 1d), but its plane and state of polarization do strongly depend on the position \( r \), with unique geometric and topological properties. In conclusion, in the absence of phase noise or any perturbation of the cavity, a unique microscopic configuration \( \zeta \) is created and maintained, and the optical field \( E(t, r, \theta) \) is a fully developed 3D spherical Gaussian speckle field. This field is associated with a position-dependent wavevector \( k(r, \theta) \) and polarization state \( d(r, \theta) \). Previous theoretical and numerical investigations of the 3D Berry field have revealed a rich set of entangled lines of optical singularities with unique geometric and topological properties. In particular, the field lines associated with the field of wavevectors \( k(r, \theta) \) are expected to exhibit strong curvature radii that can reach \( \lambda^{-1} \), and the optical field is highly structured with very complex geometry at the scale of \( \lambda \). As a consequence, each point of the aperture plane of the probe fiber is projected as a 2D circular Gaussian field, with an exponentially distributed intensity \( I(r, \theta) = (E.E^*)_0(r, \theta) \) and a contrast close to unity (see Supplementary Note 1 “Speckle Statistics”).

Interferometric response of an unperturbed cavity. Experimentally, the 3D speckle field is probed by introducing a single-mode fiber that equally splits the light toward two avalanche photodiodes and a digital correlator (Fig. 1a) which provides the intensity autocorrelation function \( g^{(2)}(\tau) \) of the time-lag \( \tau \). The normalized intensity autocorrelation function \( g^{(1)}(\tau) = g^{(2)}(\tau) - 1 \) \( \beta \), is derived using the so-called coherence factor \( \beta = g^{(2)}(0) - 1 \) (see the “Methods”, sections “Data acquisition—normalization—representation”). In the unperturbed cavity, simply filled with air under atmospheric pressure at ambient temperature and thermal equilibrium with maximal environmental stability (see the “Methods”, section “Environment control”), we reproducibly find a minute exponential decorrelation of amplitude \( 5 \times 10^{-4} \) with a time-constant \( \tau = 500 \) ps (Fig. 2a), due to the internal dynamics of the laser gain. While this decorrelation is not detectable on the usual linear-0-1 scale, a complete decorrelation is observed for very long time-lags, with a 50% decorrelation for a time-lag \( \tau_{1/2} \) that increases with increasing acquisition time \( T_{\text{acq}} \). Beyond \( T_{\text{acq}} \geq 5 \times 10^4 \) s, a fixed value of \( \tau_{1/2} \approx 3000 \) s is reached (Fig. S5), which reflects the decorrelation probably caused by the fluctuations of the central wavelength of the laser (Supplementary Note 2 “Sources of noise and digital correlation noises”).

The unperturbed cavity thus provides a stable speckle field that decorrelates in a highly reproducible fashion because of known instrumental causes. Therefore, the normalized autocorrelation function of the unperturbed cavity can serve as the baseline of instrumental speckle fluctuations. To assess the interferometric response to various perturbations, we simply measure the excess decorrelation relative to the instrumental baseline, by computing the autocorrelation difference (Supplementary Note 3 “Data handling and Baseline subtraction”). The noise associated with this procedure, i.e. the root-mean square (rms) noise floor of our measurements, is a function of \( \tau \) given by the standard deviation \( \sigma_g^{(1)}(\tau) \) associated with the baseline (Fig. 2b). This noise function which has multiple noise sources (Supplementary Note 2 “Sources of noise and digital correlation noises”) reflects the complex fourth-order statistics of the intensity and will be referred to as “baseline noise”. It decreases for increased laser intensity and longer acquisition times. Since the primary signal, i.e. the decorrelation amplitude, cannot be larger than unity, the detection range has a ceiling fixed by the unity decorrelation. The dynamic range is therefore entirely determined by the minimal rms noise amplitude. The dynamic range reaches up to 6 (resp. 4) decades when the acquisition time is 3 (resp. 1) hours (Fig. 2b).

Interferometric response to picometric deformations. To demonstrate how our 3D interferometer responds to minute deformations, the cavity was split into two halves, with two 1 mm-thick piezo actuators inserted in between at a 120° angular distance to harmonically modulate the separation distance \( \Delta l \) and
hence the cavity shape and volume at 1 kHz (Fig. 3a and see the “Methods”, section “Setup for piezo experiments”). As expected, the speckle intensity decorrelation exceeds the instrumental baseline by a difference that is modulated at the same frequency (Fig. 3b), with a depth of modulation that grows linearly with the modulation amplitude of the piezo voltage and thickness (Fig. 3c). In this set of experiments with a 3-h acquisition time, the response coefficient for the decorrelation amplitude is $3.7 \times 10^{-5}$ pm$^{-1}$, the experimental noise is reached for a 2.7 pm piezo amplitude at a 1 kHz detection frequency, and the signal-to-noise ratio is $\approx 40$ for a 1 Å modulation amplitude. Using the experimental measurement of the aforementioned “baseline noise amplitude” (Fig. 2b), we can express the power spectrum of the system expressed in pm$^2$ as a function of frequency, for a 3-h acquisition time.

The above results somehow contradict the observed non-ergodicity of the unperturbed cavity. By analogy with the sensitivity of a two-arm interferometer that is proportional to $|\sin \Delta \phi|$ where $\Delta \phi$ is the phase difference between the arms, the interferometric response of the Berry field is different for each microscopic configuration $C$. It is indeed determined by the specific contribution of the phase difference $\Delta \phi_{C_{1},C_{2}}$ for each pair of elementary fields contained in $C$. This is not what we observe, since the perturbed cavity indeed responds in a reproducible way. The fundamental reason is that any physical perturbation of the cavity shape can be considered to be microscopically irreversible. The
speckle field thus undergoes a self-ergodization process caused by the perturbations of the cavity.

We can then estimate the sensitivity of our interferometer, by analogy with the Lorentzian finesse of a Fabry–Pérot, which is given by the ratio of the free spectral range over the full-width at half-maximum linewidth of the spectral line shape: $\frac{\Delta \nu_{FSR}}{\Delta \nu_{FWHM}}$. Since our interferometer has an equivalent free spectral range $\Delta \nu_{FSR} = \frac{\lambda}{\Lambda}$ and a finesse $\nu = 2\pi G_w$, we can derive that our capacity to resolve a small change in wavelength goes as $\frac{\lambda^2}{2\pi G_w \Lambda_w}$. Independently of the Fabry–Pérot theory, the interferometric sensitivity of the cavity can also be directly derived from Eq. (1), leading to the same result. Practically, if we measure an intensity trace with $n_{ph}$ photons, with a frequency-dependent signal-to-noise ratio SNR($\tau$) given by the noise power spectrum in Fig. 2b, we can write our detection limit as

$$[\delta \lambda]_{\text{min}} = \frac{\lambda^3}{2\pi G_w \Lambda_w} \text{SNR}(\tau)/\sqrt{n_{ph}} = 1.1 \times 10^{-15} \times \text{SNR}(\tau)/\sqrt{n_{ph}}$$

(2)

which, for a typical number of photons $n_{ph} = 10^5$ photons and SNR $\geq 10$ gives $[\delta \lambda]_{\text{min}} = 3.5 \times 10^{-17}$ m. To account for the observed picometer sensitivity, we consider that a rms variation of the path length distribution and a rms variation of the wavelength have conjugated effects on the phase, and hence have statistically similar effects: $[\delta \lambda]/\Lambda = [\delta \lambda]/\lambda$, and it comes:

$$[\delta \lambda]_{\text{min}} \approx 1.6 \times 10^{-12} \text{ m}$$

(3)

This simple theory leads to a 4-fold smaller sensitivity compared to experiments, and this underestimation is likely due to two fundamental difficulties. First, it is physically impossible to probe the actual 3D isotropic Berry field described by the theory, simply because the optical fiber truncates the field by casting a shadow that necessarily destroys its spherical symmetry. Second, the fiber does not even probe that truncated 3D field, but only its 2D isotropic projection on the fiber input plane. The fiber does not collect the theoretical 3D speckle intensity, but it probes the projected intensity of the truncated field, spatially integrated over the fiber aperture. Given the geometric complexity of the field lines of $k(\tau)$ and the field correlations at the scale of $\lambda$, the susceptibility to optical fluctuations is expectedly larger for the actual 2D “projected” speckle for the 3D Berry speckle intensity.

**Interferometric response to minute dielectric fluctuations.** To demonstrate how 3D stochastic interferometry probes optical fluctuations generated inside the cavity volume with unperturbed walls, we first introduced a dense and strongly scattering jammed emulsion made of calibrated oil-in-water droplets with a monodispersed radius of 459 ± 15 nm. It has been extensively characterized for its optical and mechanical properties. Given the very high volume-fraction used here (0.785), droplets are elastically jammed with small shape deformations, and they undergo sub-nanometer thermal motions that saturate at a root-mean-square (rms) amplitude of 900 pm which corresponds to the low-frequency plateau modulus. As a result, we find that the speckle intensity decorrelation is proportional to the time-lag $\tau$ for 10 ns $< \tau < 10$ μs, and saturates at a plateau decorrelation of the value of 6% (Fig. 4a, purple line) with a cross-over time of $\tau = 70$ μs (see the “Methods”, section “Details on the experiments with intra-cavity samples”). Using the known values of the rms displacement of droplets as a function of $\tau$, we find that the decorrelation scales linearly with the mean-square displacement (Fig. 4a inset). Our results indicate that jammed droplets exhibit high-frequency Brownian fluctuations and demonstrate that we can measure rms motion amplitude up to 100 MHz with high sensitivity: at the shortest time scale, a 25 pm rms motion amplitude is recovered.

In comparison, Fig. 4a shows in blue a measurement on the very same sample and under the same acquisition conditions in conventional diffusing wave spectroscopy (DWS) in a transmission configuration. While the DWS measurement also recovers the saturation plateau of longer time delays, it only provides a readable signal down to about 10$^{-6}$ s for a 314 pm displacement. The noticeably less noisy signal from the cavity spans 8 frequency decades and is limited here by the acquisition
rate of our digital correlator \((r_{\text{sampling}} = 12.5 \text{ ns})\). Our capacity to detect motions approximately one order of magnitude smaller than for DWS at frequencies two orders of magnitude higher for these samples clearly shows the amplification of multiple scattered light fluctuations. Our data also indicates that SNR = 2000 for the detection of 0.8 nm rms motions in the 0.1–100 kHz frequency range. This suggests that 8 pm motions could be detected with SNR = 20 in that frequency range, with a dynamic range that can reach almost 4 decades. Keeping in mind that the decorrelation signal is proportional to the mean chord length of the sample, which amounts to 6.7 mm in these emulsion experiments, one could access the fluctuation dynamics of sub-millimeter-sized scattering samples in miniature cavities with Ångstrom level sensitivity.

To further demonstrate the interferometric response to intracavity optical fluctuations, we introduced three different suspensions of Mie scatterers (20 µm diameter PMMA spheres, anisotropy coefficient \(g = 0.957\)), with volume fractions such that the most (resp. least) concentrated sample is in the multiple (resp. single) scattering regime (Fig. 4b). The speckle intensity decorrelation exhibits the expected monotonic increase, from the instrumental noise floor to unity decorrelation. The response combines a diffusive regime with a linear decorrelation with \(r\), followed by a quadratic scaling dominated by sedimentation-induced motions (Fig. 4b, purple line). Given the diffusion coefficient and the sedimentation velocity, the 1 nm rms displacement expected for \(r = 10 \mu\text{s}\) is detected with SNR = 30. In comparison, measurement is done on the same sample with a commercial dynamic light scattering (DLS) instrument yields a measurement much noisier and more limited in dynamic range (Fig. 4b, yellow line). Indeed, the signal is readable for times larger than \(2 \times 10^{-4}\) s and is about 150 times smaller than the cavity signal. The cavity extends the frequency range by two orders of magnitude and the dynamic range of decorrelation. By 1.5–2 decades. We also observe with our cavity that the decorrelation amplitude unexpectedly scales with the transport mean free path length (inset to Fig. 4b). This observation points to the need for a theory to better explain how the cavity amplifies the fluctuation amplitude of scattered light intensity compared to classical methods of DLS and DWS.

**Discussion**

In the present work, we have introduced the concept of 3D stochastic interferometry enabled by the feedback of a diffuse reflective cavity on a coherent and monochromatic photon gas. It detects minute variations of the "optical volume", as qualitatively defined by the geometry of the cavity, and the dielectric tensor field inside. Thanks to the feedback on the 3D Berry field, the decorrelation function essentially detects all geometric and dielectric fluctuations through a 3D random paths integration process that is statistically isotropic and homogeneous, and fundamentally non-local. Indeed, the detection fiber statistically probes all geometric or dielectric perturbations regardless of where they occur. No information can be recovered on their spatial structure. More theoretical and experimental work is needed to quantitatively define this concept of "optical volume", and especially to characterize quantitatively the statistical response of the random optical field to the variations of the dielectric tensor field. Beyond the preliminary theory sketched here, several other theoretical problems arise, such as the photon statistics of the intensity probed by the fiber, the expectedly extreme degree of classical entanglement of the Berry field, the consequences of its complex sub-wavelength structure and super-oscillations on the interferometric properties, or a quantitative theory for the amplification of light scattering.

Practically, despite the simplicity of the design which can also be used for incoherent applications, our 3D interferometer provides a fluctuation spectrum over almost 10 frequency decades, with a dynamic range of 4–6 decades for the decorrelation signal and a picometric sensitivity. The resulting finesse \(F = 10,500\) competes favorably when compared to the best classical room-temperature Fabry–Perot interferometers designed with similar laser sources. Alternative designs can be envisioned in the frequency- and time-domain, with cations in the fields of light scattering and for ultrasensitive dielectric spectroscopy. In particular, thanks to the amplification of the fluctuation spectrum by the reflectivity gain \(G_\text{v}\), the cavity can be used to measure dynamic light scattering from highly diluted or miniature samples and quasi-non-scattering objects such as proteins, thus providing the spectrum of internal modes of motion without labels or markers. Cavity deformations could also be coupled to various external force fields (gravity, seismic motion or acoustic vibration, electromagnetic forces) for 3D picrometer metrology and ultrasensitive detection of these force fields using quite a simple design.

**Methods**

**Construction and optical properties of quartz powder cavities.** Quartz cavities were prepared from a synthetic amorphous silica powder (ZANDOSIL 30, Heraeus) in a process first described by the group Ed Frü (2005,33,37). Using transparent quartz molds (1 cm thick), the powder was mechanically compressed into a cylindrical shape, which was then baked and machined to obtain the desired cavity shape. The typical internal radius of such cylindrical cavities was 2.5 cm while the height was 6 cm and the thickness of the walls 3 cm (Fig. S8). To characterize the reflectivity of the cavity (albedo) and monitor its degradation induced by air humidity, it was measured using a ring-down method described by Fry et al. (2006)37. The response of the empty cavity to a laser pulse directly yields the reflection coefficient of the walls. Practically, a picosecond laser was used (PC-670M and PDL-800B, PicoQuant) with 120 ps and 2 nJ pulses at 670 nm, together with a time-correlated single photon counter (TCSPC Picoharp 300 system, PicoQuant) with a 4 ps time bin width, a dead time smaller than 95 ns, and 2 16-bits channels. The albedo of the quartz cavity was found to be \(\rho = 0.99994 \pm 1 \times 10^{-4}\), in agreement with the 0.99919 value found by Cone et al. at 532 nm.37 The Lambertian reflectivity profile of the walls was confirmed through the measurement of the angular intensity profile using a goniometer equipped with a power meter (Fig. S1). The expected cosine law was recovered.

**Optical setup.** The laser used for all interferometry measurements was a single frequency CW laser (Cobalt Flamenco 04-01 series), with a central wavelength \(\lambda_0 = 659.6 \pm 0.3\) nm and an optical bandwidth, \(\Delta \nu \leq 1\) MHz, i.e. a wavelength band-width \(\Delta \lambda \leq 2.5\) fm. This corresponds to a coherence time \(t_c = \pi/\Delta \lambda \approx 0.3\) µs and a coherence length \(L_\text{coh} = c/\Delta \nu \approx 95\) m. Typically, over 8 h and when the temperature fluctuations do not exceed 2 °C, the central wavelength fluctuations saturate with a maximal standard deviation \(\sigma_\lambda \approx 1\) pm. The laser intensity is subjected to fluctuations with the time that typically does not exceed 2% over 8 h if temperature variations are \(\lesssim 3^\circ\text{C}\), and the relative rms intensity noise is <0.1% in the 20 Hz–20 MHz frequency range. The laser is injected into the cavity with multimode optical fiber (NA = 0.22, FG050LA, Thorlabs). The spectrally sensitive intensity was detected inside the cavity by inserting a single-mode fiber at 670 ± 50 nm with insertion loss <3.9 dB (TW630RSF1, Thorlabs). The fiber was split into two branches connected to two-photon counting avalanche photodiodes (SPCM-AQRH, Excelitas) with the following characteristics: diameter 180 µm, 5% efficiency at 650 nm, 10 ns output pulse width, 22 ns dead time, and 97% after-pulsing probability. The photodiodes were coupled to a two-channel correlator (LS Instruments) operated in the 16 MHz multi-tau correlation scheme with the following features: lag times spanning from 12.5 ns to 3436 s, 322 channels, 54,976 s lag time range, and a maximum count rate of 2 x 10^7 counts per second over 52 ns integration time intervals.

**Environmental control.** All experiments were performed in an isolated acrylic box, covered with foam panels for thermal and acoustic isolation, and enclosed in a Faraday cage for precautionary measures against electromagnetic noise. The setup was placed on an active isolation optic breadboard (DVIA–T, Dael Systems) on top of a compressed-air passive isolation optical table. Hygrometry and temperature inside the box were continuously monitored, as well as the temperature of the laser which was passively cooled. All measurements were made after waiting a precautionary 30 min warm-up time of the cavity to ensure a steady-state behavior was reached. The temperature inside the cavity reached maximum fluctuations of about ±0.1°C and did not drift from the timescale of minutes to days.
Setup for piezo experiments. Two piezoelectric ceramics with perovskite structures (ABO3),... usage of piezo actuators. These actuators had a thickness of 1 mm and a surface of 0.8 m². They were inserted in between the two halves of the cavity and their thickness was harmonically modulated at 1 kHz with a range of voltage amplitudes using a function generator. Using piezo-response force microscopy, the piezoelectric coefficient was measured at about σ = 300 ± 15 Pa/N, and the ferroelectric domains were visualized with the same technique as well as polarization/strain graphs (Fig. S3).

Details on the experiments with intra-cavity samples. For experiments conducted with samples inside the cavity (Fig. 4), the setup can be characterized by the geometric parameters shown in Fig. S2:

1. the volumes (in cm³) are Vc = 118, Vfl = 0.785 and Vw = 19.6 for the cavity, the jammed emulsion and suspended microsphere samples, respectively.
2. the surface areas (in cm²) are, respectively, Σc = 154, Σfl = 4.71 and Σw = 55.
3. the resulting mean chord (in cm) are, respectively, Lc = 3.53, Lfl = 0.67 and Lw = 1.43 given by the mean-chord length formula L = 4V/Σ.

The first series of samples (Fig. 4a) consists of a jammed emulsion of monodisperse polymethyl siloxane (PDMS) oil-in-water prepared and characterized by Kim et al. (kindly provided by Prof. Thomas Mason, UCLA).

The free-space path length variable Λ is given by the effective Albedo Λ = Vc/Vw = 1.001 at 660 nm, and their volume fraction is 0.738. The resulting transport mean free path is Λfl = 0.15 mm. Given the size of emulsion samples, the mean chord length Lc = 6.7 mm exceeds Λfl by a factor of 27. Light is thus multiply scattered by the jammed emulsion, and the photons follow a Brownian excursion through the sample with typically 700 scattering events. In addition, because of the path invariance in scattering media13,14, the average length of Brownian paths before exiting the sample volume equals Lc regardless of Λfl. Given the estimated absorption length l0 = 10 mm, the absorption probability along the Brownian path is given by Λfl/λ0 = 1/4 000 and can safely be neglected. However, the effect on the albedo is obtained by multiplying this very small absorption probability by the probability of a random cavity chord to intersect the sample volume, which is given by the view factor of the sample from the cavity. This factor can be estimated by the surface ratio (s = 1/3), and the estimated contribution to the loss typically amounts to 10⁻⁵. Practically, we found a larger reflection loss, probably due to the sample container absorption. The effective albedo drops from 99.94% to 99.8% and the cavity gain are reduced from 1700 to about 500, thus reducing the overall amplifying effect of the cavity.

The second series of samples (Fig. 4g) consists of R = 10 μm radius polymethylmethacrylate (PMMA) particles in water (Bangs Laboratories), with a refractive index 1.498 ± 0.001 at 660 nm, and their volume fraction is 0.738. The resulting transport mean free path is Λfl = 0.15 mm, and the absorption probability along the Brownian path is given by Λfl/λ0 = 1/4 000 and can safely be neglected. However, the effect on the albedo is obtained by multiplying this very small absorption probability by the probability of a random cavity chord to intersect the sample volume, which is given by the view factor of the sample from the cavity. This factor can be estimated by the surface ratio (s = 1/3), and the estimated contribution to the loss typically amounts to 10⁻⁵. Practically, we found a larger reflection loss, probably due to the sample container absorption. The effective albedo drops from 99.94% to 99.8% and the cavity gain are reduced from 1700 to about 500, thus reducing the overall amplifying effect of the cavity.

Cavity geometry—statistics—photon budget Statistics of reflections and intracavity radiation. Thanks to the high diffuse reflectance of cavity walls, the optical intensity P0 injected into the cavity ends up being multiplied and uniformly reflected by the cavity walls. The reflection efficiency can be considered at the quantum level by a photon survival probability that corresponds to the Albedo Λ of A. At steady-state, uniform reflection losses exactly balance the intensity P0, and the walls receive a uniform irradiance Σ/P0 over a total surface area Σ, such that P0 = (1−Λ)/Σ. Consequently, the wall radiance originates from the irradiance as Λ = Σ/P0. The number ν of reflections follows a geometric distribution with probability p[ν] = Λν/(1−Λ) for 0 ≤ ν and its mean gives the cavity gain Gc as

\[ \bar{n} = G_c = \frac{1}{\Lambda} \left( 1 - \frac{\Lambda}{\bar{n}} \right) \]  

The total intensity reflected by the wall surface is, therefore, GcP0, and the wall radiance is

\[ \mathcal{A}_w = G_c P_0 / \Sigma_3 \]  

Practically, with the relatively large gain we obtained, namely Gc = 1700, and using a typical input intensity of P0 = 100 mW at 0.15 mm, 3.3 x 10⁻³ Ps⁻¹ injected into a cylindrical cavity with a height h = 6 cm and a diameter δ = 5 cm, Σc = 134 cm² and the radiance is Λ = 5 x 10⁻¹² Ps⁻¹ m⁻² sr⁻¹. The losses of energy at the cavity walls are small: Gc = 6 x 10⁻³, that optical energy is very slightly imbalanced in their vicinity. This is not true at the point of laser injection, where the local radiance exceeds the average wall radiance by a factor (1−Λ)/Σc/Σw, where Σw is the effective cross-section area of the laser input. This heterogeneity of the wall radiance can be mitigated by installing a simple baffle screen. As a result, if we virtually introduce a small planar two-sided object with two opposite surfaces ± δ, at a random point inside the cavity, we can safely assume that both surfaces will be receiving the same irradiance Λw, but with opposite orientations. We can also safely assume that the same irradiance will impact the entrance surface of any light detection device introduced inside the cavity, such as the surface of a small camera, or the input cross-section of a fiber introduced to collect light as described in the "Methods", section "Optical setup". However, the light collector by such objects is removed from the intracavity power density and this leads to a reduction of the effective Albedo. This is why the effective Albedo must be calibrated for each cavity configuration, and this precaution obviously includes the situations where a possibly absorbing sample is introduced into the cavity. Under this provision, the cavity gain Gc and the cavity radiance are correctly given by the effective Albedo Λw determined from the pulse response by

\[ \mathcal{A}_w = G_c P_0 / \Sigma_3 \]  

Statistics of intracavity chord length and photon path length. The pulse response function (Fig. 1) indicates the residence time of optical pulse energy inside the cavity, or the typical time a photon survives before it is absorbed by the walls. A simplistic picture can be considered, in which a random photon path is made of a succession of a random number n of reflections, and n = nsrc + 1 free-space chords through the cavity. These chords can be represented by the random variable Λ, and the well-known mean-chord-length property applies13,14, for any finite volume V enclosed by a surface Σ and filled with any perfectly scattering medium, be it homogeneous and isotropic or not, and regardless of the geometry of the cavity or the medium, a particle entering the volume with a uniform and isotropic incidence will follow a random trajectory inside the volume with a mean given by the very simple geometric invariant 4V/Λ. For our cavities, this property implies that the mean of free-space chords with cavity volume V is

\[ \bar{\Lambda}_c = 4V / \Sigma \]  

The free-space path length variable Λ is therefore the product of the two independent random variables n and Λ, which averages to n Λ = Λc Λw = 4V / Σw.

Besides the free-space contribution to the photon path, the diffusive random walk inside the walls needed for the reflection process corresponds to an additional random variable τr that reflects the random reflection time. Its mean has been measured for visible light as a typical delay of τr ~ 5 ps. As a consequence, the total reflection time averages as 4V/nτr = 4V/τr, the total photon path length can be represented as a composite random variable

\[ \Lambda = n_s r + n_c r_c \]  

with the variance σ² and the average Λ given by

\[ \bar{\Lambda} = G_c [\bar{\Lambda}_c + c t_c] \]  

Practically, in the conditions of our setup, with cylindrical cavities with h = 6 cm and δ = 5 cm, it comes:

\[ \bar{\Lambda}_c = h / \left[ \frac{1}{2} + \frac{c}{\bar{n}} \right] \]  

and Λc = 3.5 cm, while cτr = 1.5 mm. The mean total photon path, therefore, corresponds to a length of Λ = 62 m and a mean residence time τ = Λ/Λw ~ 200 ns. Importantly, the coherence length of the laser exceeds the average photon path length by a factor Λc/Λw ~ 1.5. The photon path is longer than the coherence length. This leads us to safely assume that most pairs of modes are mutually coherent, and the optical field is practically coherent across the cavity, thus enabling multiphoton interferences.

Photon density. The light inside the cavity can be considered as an isotropic and monochromatic photon gas, with a homogeneous density P0/ρ0. This density can be computed from the photon energy, Λc/Λw, the photon injection rate P0/ρ0 = P0 Λc/Λw and the mean residence time τ described above. It comes.
\[ p_{ph} = P_{ph} \times V_c, \] or \( p_{ph} = 4n \times \kappa_{ph} / C_c. \) With \( P_{ph} = 100 \text{ mW} \) as considered above, the photon injection rate is \( P_{ph} \approx 3.3 \times 10^{12} \text{ s}^{-1} \) and the photon density is, while the photonic radius is \( \xi_{ph} = 5 \times 10^{-5} \text{ m} \times 10^{-12} \text{ s}^{-1}. \)

Capture of the speckle intensity by a monomode fiber. Using the photon gas equation, we can estimate the rate at which photons can be collected from the cavity with the single-mode fibers used in our experiments. Knowing the surface of the collected optical fiber, we can use the concept of irradiance to calculate the number of detected photons. Here, the irradiance is \( I_{ph} = 2 \times 10^{-5} \text{ W/m}^2 \) so that the flux through an optical fiber with a core of cross-section \( n_{d} = 19 \mu\text{m}^2 \) (diameter 5 mm) would be about \( 1.2 \times 10^{-5} \text{ W} \) or \( 10^{-5} \text{ W} \). 

Data normalization. \( g_{i}(\tau)\) is assessed from the experimental data (see Fig. S4). Therefore, decorrelation measurements only make sense when compared to the ‘reference’ decorrelation function measured with the unperturbed cavity. As a consequence, the decorrelation \( g_{i}^{\text{ref}}(\tau)\) obtained in a given experiment should not be compared to the function \( g_{i}(\tau)^2\), but to the decorrelation \( g_{i}^{\text{ref}}(\tau)\) obtained from reference experiments conducted with the unperturbed cavity. The relevant signal is therefore the excess of decorrelation given by the difference:

\[ \delta(\tau) = 1 - [g_{i}(\tau)^2]_{\text{measure}} - 1 - [g_{i}^{\text{ref}}(\tau)^2]_{\text{measure}}. \]
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