

Mesoscopic light transport in nonlinear disordered media

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Nonlinear disordered media uniquely combine multiple scattering and second-harmonic generation. Here, we investigate the statistical properties of the nonlinear light generated within such media. We report super-Rayleigh statistics of the second-harmonic speckle intensity, and demonstrate that it is caused by the mesoscopic correlations arising in extreme scattering conditions. The conductance measured is exceptionally low for an isotropically scattering three-dimensional medium, enabling applications in broadband second-harmonic generation, wavefront shaping in nonlinear disordered media, and photonic computing.

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When a coherent wave interacts with a disordered medium, it generates a complex interference phenomenon that results in the formation of speckle patterns. These speckles, which are randomly distributed diffraction-limited grains, are ubiquitous across waves of various origins, including electromagnetic [1], acoustic [2], and matter waves [3]. Despite their different physical origins, speckles exhibit universal statistical properties that are referred to as Rayleigh statistics. This universality is a consequence of the very general conditions under which Rayleigh statistics emerge. Specifically, the only requirement is that the field arises from the interference of a large number of uncorrelated waves whose phases are uniformly distributed over a 2π range [1,4]. Non-Rayleigh statistics can be achieved by using a spatial light modulator to induce correlations between partial waves and redistribute the intensity among the speckle pattern's grains, while preserving ergodicity [5–7].

Conversely, observing deviations from Rayleigh statistics in multiple-scattering media under conventional laser illumination is significantly more challenging. Deviations occur only when strong scattering introduces mesoscopic correlations between partial waves, breaking ergodicity [8–12]. These correlations are associated with the existence of a finite number g of open transmission channels in disordered media [13,14]. When g (also known as *dimensionless conductance*) is moderate, the central limit theorem does not apply, causing the total intensity in the speckle pattern to fluctuate significantly from one disorder configuration to another. Consequently, the intensity of each speckle grain exhibits super-Rayleigh statistics [9,15]. This peculiar regime has been reported in three-dimensional (3D) systems only in nanowire mats [16] and, more weakly, in isotropic ZnO scattering media [17].

While mesoscopic transport in linear scattering media is well understood, the same cannot be said for nonlinear disordered media, a class of materials that has garnered significant interest recently. These materials are composed of nanodomains with a noncentrosymmetric crystal structure that exhibit a nonzero second-order susceptibility tensor. The nonlinear properties enable each nanodomain to exhibit electro-optic effects and generate second-harmonic (SH) waves when illuminated with a fundamental beam [18]. The interference of the waves generated by the nanodomains results in efficient emission of nonlinear light without stringent conditions on the polarization and wavelength of the fundamental light, contrary to bulk crystals [19]. This phenomenon has been extensively studied in the framework of random quasi-phase-matching [20,21], where the disorder is used to achieve broadband SH generation [22–26]. Researchers investigated the fundamental properties of diffusion and weak localization in this class of media [27–30], as well as the effect of scatterer displacement [31,32]. Notably, nonlinear disordered media are also emerging as a prominent platform for photonics processing. The characterization of the scattering tensor that defines their nonlinear input-output response allows the use of nonlinear disordered media for encryption and as all-optical logic gates [33,34]. In addition, these media enable the implementation of large-scale nonlinear optical operators for photonic machine learning [35]. However, simplifying the analysis by assuming Rayleigh statistics, these applications might overlook the richer physics arising from the interplay of nonlinear light generation and strong scattering effects.

In this paper, we examine the statistical properties of SH light generated within a strongly scattering, nonlinear disordered medium. Specifically, we measure the intensity fluctuations of both the fundamental and SH light under various illumination conditions. The histograms of speckle intensity and total transmission show deviations from Rayleigh statistics, particularly evident for SH light. Since SH light can be generated throughout the medium and cannot be de-

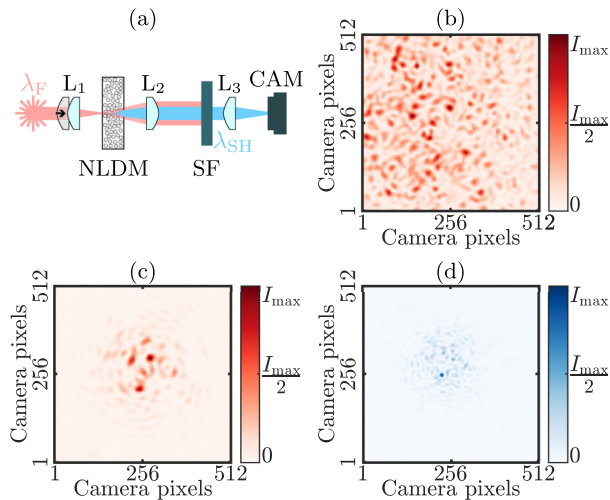


FIG. 1. Experimental setup and typical speckle patterns. (a) $\lambda_F = 976$ nm: pulsed laser; $\lambda_{SH} = 488$ nm; L: lens; NLD: nonlinear disordered medium; SF: spectral filter; CAM: camera. (b), (c) Camera images of typical speckle patterns at λ_F when the sample is illuminated with (b) out-of-focus and (c) in-focus fundamental light. (d) Camera image of a typical speckle pattern from SH light generated within the medium with in-focus fundamental illumination.

scribed by a linear transmission matrix, an open question is whether these observations can be interpreted in terms of an effective number g of open channels for SH light. By analyzing the spatial distribution of SH generation within the medium, we motivate the application of mesoscopic transport theory, which is in excellent agreement with our experimental data, and is consistent with the reduced conductance g due to the shorter transport mean free path at the SH wavelength. This outcome has relevant applications in broadband SH generation, wavefront shaping [36,37], and photonic computing [35,38].

Our experimental setup is shown in Fig. 1(a). A linearly polarized Ti:Sa pulsed laser (wavelength $\lambda_F = 976$ nm, 80 MHz repetition rate, 100 fs pulse duration) is focused by an aspheric lens [numerical aperture (NA) = 0.5] into a nonlinear disordered medium (disordered assembly of LiNbO₃ nanoparticles of thickness $L \approx 10$ μ m; see Supplemental Material Sec. S1 for detailed information [39]). Both the lens and the disordered sample are mounted on motorized translation stages, to accurately control the position of the focal plane, as well as the position of the sample in the plane perpendicular to the beam propagation direction. By moving the focal plane, we can control the size of the beam at the input facet of the disordered medium, a crucial parameter for the analysis of mesoscopic transport. Changing the position of the sample allows the light traveling through the medium to interact with different realizations of disorder. The light scattered and upconverted by the nonlinear disordered medium is collected by an objective (NA = 0.75), and a tube lens (focal length 200 mm) is used to image the output facet of the medium onto a scientific CMOS camera. Finally, we use a linear polarizer to select a single polarization state, and a spectral filter (bandpass filter, central wavelength 488 nm, bandwidth 1 nm) to remove the fundamental light when measuring the SH

signal ($\lambda_{SH} = \lambda_F/2 = 488$ nm). Typical speckle patterns for different illumination conditions are shown in Figs. 1(b)–1(d). Figure 1(b) displays a camera image of the fundamental light when the focal plane is far from the input facet of the medium, i.e., when the illuminating beam size is large. Figures 1(c) and 1(d) report typical speckle patterns for the fundamental light [Fig. 1(c)] and the SH light generated within the nonlinear disordered medium [Fig. 1(d)] when the focal plane of the fundamental illumination coincides with the input facet of the sample.

In the following, we rigorously characterize the statistical properties of the transmitted light after the propagation through the nonlinear disordered medium. In particular, we measure the intensity fluctuations, which contain a strong signature of mesoscopic transport, and are not affected by absorption [40]. For a given realization of disorder, we use the camera to record the intensity distribution at the output facet of the disordered sample. From the measured speckle pattern, we obtain the transmission coefficients T_{ab} , which relate a speckle spot b with an incoming wave a (kept fixed for each statistical dataset). Summing over the output modes, we obtain the total transmission as $T_a = \sum_b T_{ab}$. To collect a statistically meaningful set of measurements, we measure the intensity distributions of 10^4 different realizations of disorder, obtained by moving the sample in the plane perpendicular to the propagation direction of the illuminating light. From T_{ab} and T_a , we then extract the most relevant quantities for our analysis, which are the normalized speckle intensity $s_{ab} = T_{ab}/\langle T_{ab} \rangle$ and the normalized total transmission $s_a = T_a/\langle T_a \rangle$, where the angle brackets denote the average over the ensemble of random configurations (see Supplemental Material Secs. S3 and S4 for details about the data analysis [39]).

The results of the analysis for various illumination conditions are reported in Figs. 2 and 3. We first examine the case of fundamental light with out-of-focus illumination [Figs. 2(a) and 2(c)]. The large size of the input beam ensures that we are in a regime of large number of open channels. In this case, the normalized speckle intensity obeys the Rayleigh law, meaning that the probability distribution $P(s_{ab})$ is a negative exponential [gray solid line in Fig. 2(a)]. Conversely, the probability distribution of the normalized total transmission $P(s_a)$ follows a Gaussian distribution [Fig. 2(c)]. This is a consequence of the central limit theorem, as the numerous scattering paths that are summed to obtain the total transmission are uncorrelated.

Reducing the size of the illuminating beam by aligning the focal plane with the input facet of the medium (referred to as the *in-focus* condition) increases the probability of two paths crossing during light propagation inside a disordered medium. When two paths cross, there is a small but nonzero probability that they will become correlated, giving rise to long-range correlations between distant speckle grains [10,41]. This, in turn, lowers the dimensionless conductance g , which quantifies the number of open channels, i.e., the number of channels that carry the majority of the light [13,14,42]. The introduced correlations change the statistical properties of the scattered light, enhancing in particular the intensity fluctuations [43]. Mesoscopic transport theory predicts modified probability distributions, referred to as $P_g(s_{ab})$ and $P_g(s_a)$, containing the dimensionless conductance g as a single parameter (see Ref. [15] and Supplemental Material Sec. S6 [39]).

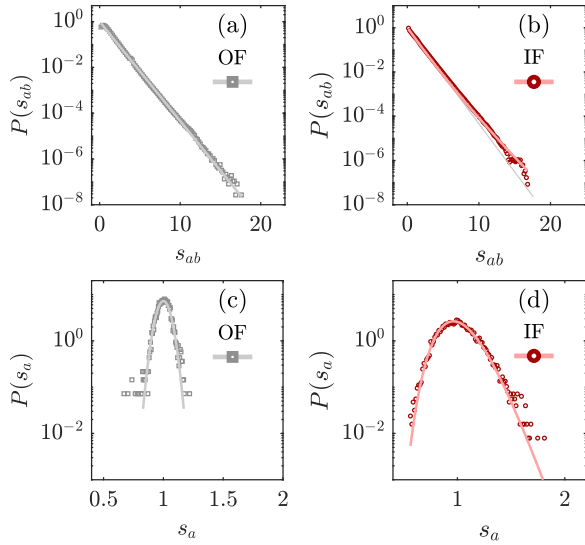


FIG. 2. Intensity fluctuations statistics of fundamental light. (a), (b) Histograms of normalized speckle intensity for (a) out-of-focus (OF) and (b) in-focus (IF) illumination. Solid lines represent a negative exponential $P(s_{ab}) = -\exp(s_{ab})$ (gray), and the mesoscopic transport model for normalized speckle intensity $P_g(s_{ab})$ (red). (c), (d) Histograms of normalized total transmission for (c) out-of-focus (OF) and (d) in-focus (IF) illumination. Solid lines indicate a Gaussian fit (gray), and the mesoscopic transport model for normalized total transmission $P_g(s_a)$. The fitted conductance value in (b) and (d) is $g = 13.8$.

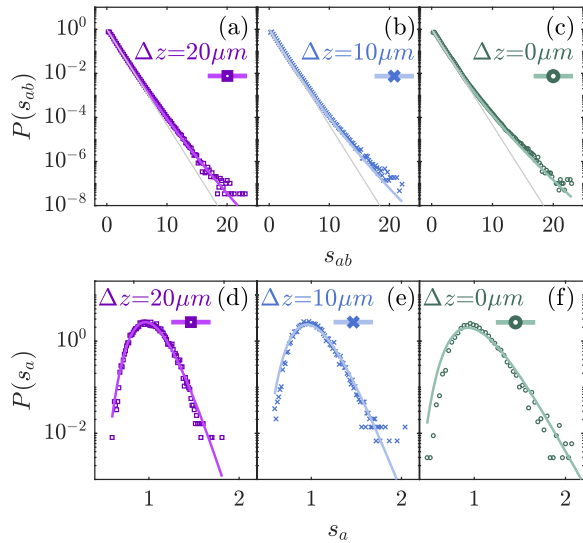


FIG. 3. Intensity fluctuations statistics of SH light generated within the nonlinear disordered medium for varying distance Δz between the focal plane of the input lens and the entry facet of the nonlinear disordered medium ($\Delta z = \{20 \mu\text{m}, 10 \mu\text{m}, 0\}$). (a)–(c) Histograms of normalized speckle intensity. Solid lines represent a negative exponential $P(s_{ab}) = -\exp(s_{ab})$ (gray), and the mesoscopic transport model $P_g(s_{ab})$ [(a) $\Delta z = 20 \mu\text{m}$, violet, (b) $\Delta z = 10 \mu\text{m}$, blue, (c) $\Delta z = 0$, green]. (d)–(f) Histograms of normalized total transmission. Solid lines are the mesoscopic probability density functions $P_g(s_a)$ [(d) $\Delta z = 20 \mu\text{m}$, violet, (e) $\Delta z = 10 \mu\text{m}$, blue, (f) $\Delta z = 0$, green]. The fitted conductance values are (a), (d) $g = 13.1$, (b), (e) $g = 10.2$, and (c), (f) $g = 7.1$.

The normalized speckle intensity and total transmission histograms for fundamental light with in-focus illumination is shown in Figs. 2(b) and 2(d), respectively. Considering the speckle intensity, the slight deviation from Rayleigh distribution at large s_{ab} values is consistent with previously reported observations in isotropic scattering materials [17]. Conversely, the total transmission histogram shows a clear deviation from the Gaussian distribution predicted by the uncorrelated wave model. Fitting the measured data with the mesoscopic transport model of Ref. [15] demonstrates good agreement [Figs. 2(b) and 2(d), solid red lines], resulting in a conductance value $g = 13.8$ (see Supplemental Material Secs. S2, S6, and S7 [39]).

Having established how the statistical properties of fundamental light depend on the illumination conditions, we now turn to the central focus of our work: the intensity fluctuations of SH light generated within the nonlinear disordered medium. Specifically, we studied how these fluctuations vary with the input lens position (L_1 in Fig. 1), which controls the fundamental spot size at the entrance of the nonlinear medium. The results are shown in Fig. 3. We present histograms for three illumination conditions, based on the distance Δz of the focal plane to the input facet of the medium: $\Delta z = 20 \mu\text{m}$, $\Delta z = 10 \mu\text{m}$, $\Delta z = 0$. As predicted by mesoscopic transport theory [41], the deviation of the speckle intensity histograms [Fig. 3(a)] from a negative exponential (gray solid line) becomes more evident as the focal plane of the illumination approaches the input facet of the sample. Similarly, the histograms of the normalized total transmission [Fig. 2(b)] demonstrate increased variance for shorter Δz . This is particularly evident for large s_a values, where the data points closely follow a negative exponential of the form $P(s_a) \propto e^{-gs_a}$. The fitted probability distributions (violet, blue, and green solid lines in Fig. 3) show excellent agreement for both the speckle intensity and the total transmission histograms, using the same conductance values for the expressions of $P_g(s_{ab})$ and $P_g(s_a)$ ($g = 13.1$ for $\Delta z = 20 \mu\text{m}$, $g = 10.2$ for $\Delta z = 10 \mu\text{m}$, and $g = 7.1$ for $\Delta z = 0$; see Supplemental Material Secs. S6 and S7 [39]).

The reduced conductance measured with the SH light is explained by the shorter transport mean free path ℓ_t associated to the SH wavelength ($\ell_t \approx 147 \text{ nm}$ at 450 nm , compared to $\ell_t \approx 674 \text{ nm}$ at 950 nm ; see Supplemental Material Sec. S8 for the experimental characterization of ℓ_t [39]). The transport mean free path is relevant for the characterization of the mesoscopic effects, because it limits the minimum conductance to a value $g_{\min}(\lambda) \approx (2\pi\ell_t/\lambda)^2$ [41,44]. Considering the measured ℓ_t , we obtain $g_{\min}(950 \text{ nm}) \approx 10.5$ and $g_{\min}(450 \text{ nm}) \approx 2.2$ [39]. Notably, by accounting for the beamwidth dependence of g with a finite SH illumination width of 800 nm , consistent with our experimental parameters, we estimate a conductance of $g = 7$ (see Supplemental Material Sec. S8 [39]).

The question remains on how the statistics can be so accurately described with mesoscopic transport theory, given that the generation of the SH light throughout the medium prevents the definition of a linear transmission matrix [33]. The explanation could be that, due to the tight focusing, the SH generation efficiency is maximized at the entrance of the medium. Indeed, theoretical calculations show that the SH signal is mostly generated at $z < 500 \text{ nm}$ [see Fig. 4(a),

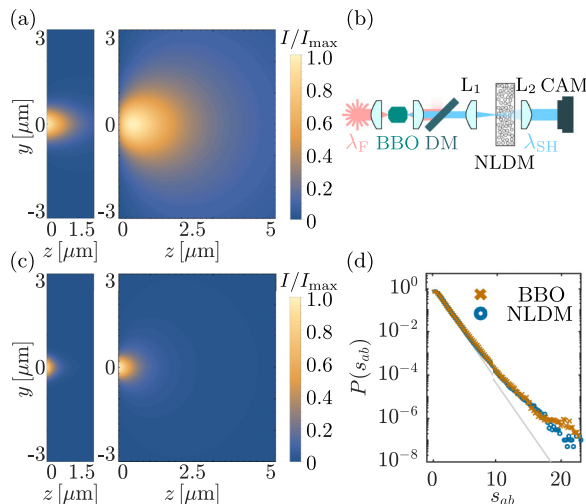


FIG. 4. Comparison of SH light generated within the nonlinear disordered medium, or externally, via a BBO crystal. (a), (c) Calculated mean intensity profiles within the disordered medium of the sources (left) and the resulting diffuse halos (right) for SH light generated (a) within the nonlinear disordered medium or (c) with a BBO crystal [39]. (b) Experimental setup. Light from a pulsed laser is upconverted with a BBO crystal ($\lambda_F = 976$ nm, $\lambda_{SH} = 488$ nm). A dichroic mirror (DM) filters out the fundamental light, while the SH is focused by the lens L_1 onto the nonlinear disordered medium (NLDM). The scattered light is collected by an objective L_2 and measured with a camera (CAM). (d) Histograms of normalized speckle intensity $P(s_{ab})$ for SH light generated with a BBO crystal (cross, yellow) or by the disordered medium (circles, blue).

on the left; theoretical and simulation details in Supplemental Material Sec. S9 [39]. This means that, given that the nanoparticles' sizes are distributed between 100 and 400 nm [39], the SH light is generated within the very first layers, and then undergoes linear scattering for the rest of the thickness $L \approx 10$ μm of the material [Fig. 4(a), on the right]. Therefore, under tightly focused illumination, the transport of SH light could be treated similarly to that in a linear scattering medium, despite its generation occurring within the disordered medium. To prove it, we compare the statistical properties of SH light generated within the nonlinear medium, and by an external 200- μm -thick BaB_2O_4 crystal [BBO, experimental setup depicted in Fig. 4(b)]. Although the light generated within the nonlinear disordered medium [Fig. 4(a), right] penetrates slightly deeper than the light generated by the BBO [Fig. 4(c), right], the sources of both concentrate at a depth $z \ll L \approx 10$ μm [Figs. 4(a) and 4(c), left]. In addition, the measured histograms of s_{ab} for SH light from both the nonlinear medium and the BBO [shown in Fig. 4(d) in yellow and blue, respectively] closely match. The excellent agreement between the two histograms demonstrates that the conductance values measured for SH light generated in the medium are primarily determined by the sample's scattering properties. Remarkably, they can be accurately described by mesoscopic transport theory, despite both mesoscopic correlations and SH light build up within the same ultrathin region.

In summary, we have experimentally measured the intensity fluctuations of fundamental and SH light generated by a nonlinear disordered medium. We observed distinct devia-

tions from the predictions of the uncorrelated wave model in the histograms of SH speckle intensity and total transmission. The measured deviations are well described by mesoscopic transport model, providing clear evidence of mesoscopic transport in a nonlinear disordered medium. Moreover, the measured conductance of $g = 7.1$ is exceptionally low for an isotropically scattering 3D medium. We motivated the description of the nonlinear input-output response in terms of the effective number g of open channels by observing that the SH light is mainly generated within the initial layers of the nonlinear disordered medium. We validated this approach by comparing the speckle intensity of SH light generated within the nonlinear medium and externally with a BBO crystal, obtaining similar fluctuation statistics. Nevertheless, it is crucial to emphasize that our nonlinear disordered medium differs fundamentally from a linear scattering material. Random quasi-phase-matching, in fact, remains a critical feature of the system. Although most of the SH generation occurs within the first layers of the medium, the extreme scattering conditions cause multiple scattering events to take place inside this ultrathin region. Therefore, combining random quasi-phase-matching with the low conductance values measured in this work, we achieve efficient broadband SH generation in an extremely thin medium, regardless of temperature, polarization, and wavelength. Indeed, our results show SH speckles with intensities more than 20 times stronger than the average, occurring with a probability two orders of magnitude higher than that of uncorrelated light. The increased probability of generating higher SH intensity condensed in few speckles finds useful applications in broadband SH generation from nonlinear disordered media. Additionally, achieving a low number of open transmission modes has also significant implications for nonlinear wavefront shaping, as the long-range correlations enhance the achievable control [36,37]. The deviations from the uncorrelated wave model indicate that, when the fundamental light is strongly focused to achieve a higher SH signal, the wave propagation through the complex nonlinear medium cannot be described by a fully random nonlinear input-output response [33]. It is therefore essential to consider this effect for photonic computing applications that aim to exploit randomness to achieve large-scale nonlinear operations [35].

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A.N. and A.M. contributed equally to this work.

DATA AVAILABILITY

The data that support the findings of this article are openly available [45].

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