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Impact of fundamental thermodynamic fluctuations on light propagating in photonic waveguides made of amorphous materials

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To improve the detection limit of optical sensors, it is of paramount importance to understand light–matter interaction processes at a fundamental level. At room temperature, the ultimate detection limit is governed amongst others by fundamental thermodynamic fluctuations. Their effect on the properties of light that propagates in amorphous materials is not well understood. Here, we unveil and model for the first time a dominating high-frequency (terahertz-range) noise contribution in the phase of laser light having propagated in optical waveguides, leading to Raman-like sidebands in the optical output spectrum. A salient feature of our approach is to consider a mean relaxation time of the spontaneous random heat flux in the medium, which leads to a spatial correlation of the thermo-refractive noise. The resulting phase noise can be several orders of magnitude larger than what was predicted by earlier models. Our model allows us to explain the origin and specificities of the background that is observed in the Raman optical spectra of silicon nitride waveguides and silica optical fibers. Not only do these findings add a previously unknown dimension to the fundamental knowledge about noise in light–matter interaction, they also need to be taken into account in any optical system in which dynamic fluctuations at the picosecond (*ps*) or *sub-ps* level play a role.

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1. INTRODUCTION

The statistical properties of waves can be strongly modified when they propagate in a random medium subject to temporal fluctuations. The intensity fluctuations of cosmic radio waves induced by the propagation through the ionosphere are an example of such a drastic impact of the random nature of the propagating medium, as observed by Hey *et al.* in 1946 [1]. In a fluid such as the atmosphere, the fluctuations of the dielectric constant are mainly related to the motion of parts of the fluid that have different densities.

In a solid in thermal equilibrium with the surrounding at an average temperature $\langle T \rangle = T_0$, the thermodynamic fluctuations [2] are the most fundamental source of the fluctuations of the dielectric constant. As a result, light propagating in a solid is subject to thermorefractive phase noise, which has mainly been investigated either in long fibers [3–5] or in microsphere resonators [6].

Up to now, the observation of the fundamental thermal noise in optical media has been limited to frequencies lower than 1 MHz [7]. A high frequency cutoff for the associated spectrum seems not to have been experimentally identified in optical structures yet. Our main goal is to investigate the high-frequency part of the optical spectrum of a guided mode subject to fundamental thermal fluctuations.

The conventional modeling of the thermal noise in a waveguide [3,8,9] relies on the classical theory of hydrodynamical fluctuations, as summarized in Fig. 1. In the framework of this theory, a random spontaneous heat flux $\delta\vec{q}$ is incorporated to the Fourier law as follows: $\vec{q} = -\kappa_0 \vec{\nabla} T + \delta\vec{q}$, where $\vec{q}$ is the heat flux in an elementary volume δV, T the local effective temperature of the waveguide, and κ_0 the heat conductivity [10]. The spontaneous heat fluxes result from the constant motion of the elementary charges of the medium. Note that here the temperature T of the waveguide is defined as a local effective temperature, which is different from the ambient temperature T_0. In contrast to T_0, the local effective temperature T is subject to fluctuations δT: $T = T^s + \delta T$, with T^s the local steady-state temperature. The fluctuating part δT is linked to the fluctuation δu of the specific internal energy u via the thermodynamic relationship $\delta u = C_V \delta T$ with C_V the specific heat per unit mass at constant volume.

Determining the resulting noise spectrum of the temperature field requires the complete knowledge of the spatial and temporal

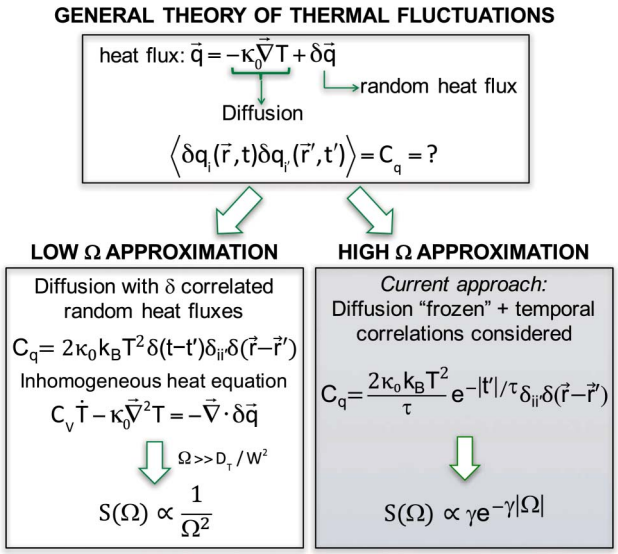

Fig. 1. Positioning of the current model (blue box) against the conventional treatment of the thermo-refractive noise in waveguides. The optical spectrum $S(\Omega)$ is considered at high frequencies. See the main text for the definition of the different symbols.

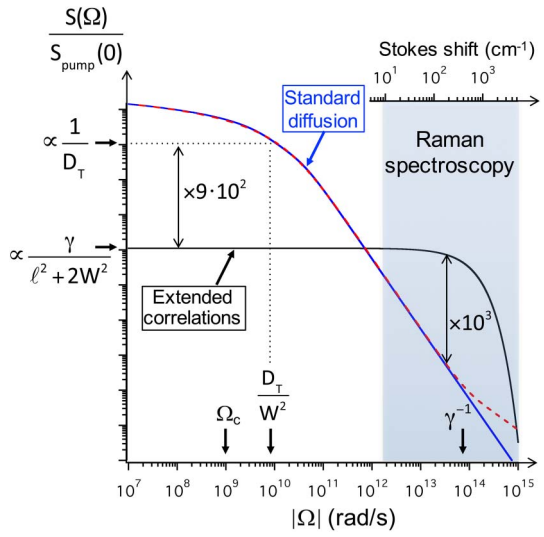

Fig. 2. Theoretical optical spectra with standard thermal diffusion (blue line) and with extended correlation of the heat flux (black line). The parameters are those of a typical silicon nitride waveguide (see main text). Red dashed curve: outcome of Rytov's theory, where quantum effects are included in addition to the standard thermal diffusion. The blue area corresponds to the frequency range relevant for Raman spectroscopy.

correlations of the components δq_i $(i = x, y, z)$ of the random heat flux $C_q = \langle \delta q_i(\vec{r}, t)\delta q_{i'}(\vec{r}', t')\rangle$, which is *a priori* unknown. However, some approximations can be carried out. The first one assumes that the characteristic decay time τ of the fluctuations of $\delta\vec{q}$ is much shorter than the time decay of the temperature fluctuations [11], i.e., the thermal relaxation governed by a diffusion process is much slower than the relaxation of the elementary heat flux. As a result, the rate of change of $\delta\vec{q}$ at a given time t is entirely determined by the value of $\vec{q}$ at t. For weak fluctuations, a linear expansion leads to $d\delta q/dt = -\delta q/\tau$ [12]. As a consequence the time correlation of the components of the spontaneous heat flux is given by $\langle \delta q_i(\vec{r}, t)\delta q_{i'}(\vec{r}', t + t')\rangle = \frac{2\kappa_0 k_B T^2}{\tau} \exp(-\frac{|t'|}{\tau})\delta_{ii'}\delta(\vec{r} - \vec{r}')$ [11].

At this stage, the conventional approach that is labeled here as "standard diffusion" model consists of making the mean relaxation time τ of the spontaneous heat flux tend to zero, which implies a "δ" correlated spontaneous heat flux, $\langle \delta q_i(t)\delta q_{i'}(t + t')\rangle = 2k_B\kappa_0 T^2\delta(t')\delta_{ii'}$. Based on the inhomogeneous heat equation (see Fig. 1), such an approximation results in an $1/\Omega^2$ power law of the power spectrum of the thermal fluctuations at high frequency Ω, and concomitantly of the optical spectrum $S(\Omega)$ of a propagating optical mode in the medium as detailed in Supplement 1 and plotted in Fig. 2.

Another way of determining the thermal noise is to make use of the fluctuation-dissipation theorem that was introduced by Callen *et al.* [13]. This approach was implemented by Rytov [14] to develop a spectral theory of the thermal fluctuations that takes into account quantum effects. As shown in Fig. 2, the standard diffusion model is in line with Rytov's theory for frequency $|\Omega|$ smaller than 10^{14} rad/s. Above this frequency, quantum effects begin to appear. Although Rytov's theory goes beyond the standard diffusion model, it is based, as in Ref. [8], on the Fourier law, which has unphysical properties at high frequencies as discussed later, and assumes a local thermodynamic equilibrium, which excludes classical correlation effects of the spontaneous heat flux.

One open question is the consequence of neglecting the mean relaxation time τ. Here, we develop an approach that allows taking into account τ and determining its impact on the noise spectrum at high frequency. In particular, an exponential decay of the optical spectrum of a guided mode is analytically predicted (see Fig. 2), which is in line with the Raman background recently observed in optical waveguide-based sensors [15–17]. In contrast to the conventional approach where the slow dynamic of the thermal diffusion plays a crucial role and is responsible for the $1/\Omega^2$ power low, we will mainly focus on the dynamic of the spontaneous heat flux $\delta\vec{q}$ and consider that the thermal diffusion is "frozen" at high frequency. This means that we are interested in the random heat fluctuations around the instantaneous value of $-\kappa_0\vec{\nabla}T$, which is assumed to be constant during the decay of a random heat flux fluctuation.

The Fourier law on which the above-mentioned standard models of thermal fluctuations are based has the drawback to lack inertial effects. As stated in Ref. [18], it neglects the time needed for acceleration of the heat flow. Such an approximation that leads to infinite speed of propagation is questionable at high frequencies [19]. Our approach aims at avoiding such an unphysical effect [20].

Based on extended irreversible thermodynamics, we will show that a relaxation time associated with the fluctuation of the heat flux implies a correlation length ℓ of the fundamental thermal fluctuations δT. When the correlation length or equivalently the relaxation time tends to zero, the contribution of the heat flux fluctuation to the noise spectrum vanishes for $\Omega \neq 0$ as expected with the conventional approach.

The knowledge of the spatial frequency spectrum of the fundamental thermal fluctuations and of its dynamics allows determining the time varying spectral density of the resulting refractive index fluctuations and as a consequence the optical spectrum of the light field propagating in the medium.

The current paper aims at clarifying the role of the temporal and spatial correlations of the fundamental heat flux on the optical spectrum of the light field, in particular at high frequency. This point is not only of fundamental interest due to the ubiquity of thermal noise but also of paramount importance to set the ultimate limits of optical sensors. We start by summarizing the main outcome of our model and then theoretically determine the optical spectrum of a classical coherent field that is propagating in a waveguide at thermal equilibrium. This determination is carried out in the framework of a perturbation approach that assumes weak fluctuations of the refractive index of the waveguide. The outcome of our model allows us then to discuss the salient features of the impact of thermal noise on the light propagation properties. In the last part, the model is compared with the experimental optical spectrum of a mode guided in a single-mode silicon nitride (SiN) waveguide, which provides an estimate of the correlation length of the index fluctuations, of the mean relaxation time of the spontaneous heat flux, and of the fundamental thermodynamic limit of the optical noise. In particular, we attribute the origin of the background that is observed in Raman spectra of amorphous media to the fundamental thermodynamic fluctuations.

2. MAIN THEORETICAL RESULT

Before providing the different steps of the derivation of the optical spectrum of a guided mode subject to thermal fluctuations, we first reveal the main outcome of our theoretical approach. In particular, we compare in Fig. 2 the "standard diffusion" model (blue line in Fig. 2 and see also Supplement 1) and our current approach, which is called "extended correlations" insofar as it considers a nonvanishing correlation time τ. For such a comparison, the different parameters that are defined below are those of silicon nitride. The optical spectrum (black curve in Fig. 2) resulting from the "extended correlations" model is given by the pivotal Eq. (10) hereafter.

The optical spectrum of the standard model is mainly governed by the thermal diffusivity D_T of the medium and the radial half-width W of the intensity mode profile. In the case of extended correlations, the spatial correlation length ℓ, the half width W, and a relaxation time γ that will be defined later are the three main parameters that allow positioning the extended correlations optical spectrum against the standard diffusion optical spectrum.

Figure 2 highlights that the optical spectrum related to the model based on extended correlations is negligible at frequencies Ω smaller than 10^{11} rad/s but is predominant in a frequency range that matches the one relevant for Raman spectroscopy, namely between 10 and 5000 cm^{-1}. As a consequence, the noise contribution resulting from the extended correlations model sets the main detection limit for waveguide-based Raman sensing, which is the main finding of the current article.

The standard model is relevant as long as the frequency is smaller than $1/\tau$, i.e., as long as the temporal correlation of the random heat fluxes can be neglected. From the experimental Raman spectrum of silicon nitride waveguides (see Section 3.F below), the extended correlations model predicts 7×10^8 rad/s $\lesssim 1/\tau \lesssim 2.5 \times 10^9$ rad/s. On another side, above the cutoff frequency $\Omega_c \simeq 10^9$ rad/s of the optical spectrum of the standard model, the instantaneous temperature T, and so $-\kappa_0 \vec{\nabla} T$, can be considered constant. This implies that the above-mentioned approximation of "frozen" diffusion applies. As a result, the extended correlations model is appropriate for frequencies larger than Ω_c.

In the frequency interval defined by $1/\tau$ and Ω_c, as well as in its neighborhood, a more advanced theory is necessary to determine the transition between the optical spectra of the standard diffusion and the extended correlation models. The same holds for frequencies around 10^{15} rad/s where nonclassical effects emerge, as highlighted with Rytov's model. Even if at the crossing points between the different models none of them is accurate, the actual optical spectrum can be approximated by the envelope of the extended correlation model and of Rytov's model.

3. MODEL

The physical system consists of an optical field that propagates in a single-mode waveguide made of a solid of refractive index n as schematically represented in Fig. 3. We assume that the propagation length is identical to the total length L of the medium and that L is much larger than the cross section S of the waveguide. A harmonic field U of single angular frequency ω_0 and amplitude A is coupled to the waveguide. At the input $x_0 = 0$ of the waveguide, this classical stable field is expressed as $U(x_0, r_\perp; t) = A_0(x_0, r_\perp) \exp(-j\omega_0 t)$, with $r_\perp$ the transverse coordinate in the (y, z) plane. It undergoes amplitude and phase fluctuations during the propagation in the waveguide due to thermal noise. At the output $x_L = L$, the field can be written as $U(x_L, r_\perp; t) = A(x_L, r_\perp; t) \exp(-j(\omega_0 t - \varphi(x_L, r_\perp; t)))$, where the amplitude $A(x_L, r_\perp; t)$ and the phase $\varphi(x_L, r_\perp; t)$ are random variables. The amplitude and the phase of the field after a propagation over a distance L are related to the amplitude $A_0(L, r_\perp)$ and the phase $\varphi_0(L, r_\perp)$ of the unperturbed waveguide as follows: $A(L, r_\perp; t) = A_0(L, r_\perp) + \delta A(L, r_\perp; t)$ and $\varphi(L, r_\perp; t) = \varphi_0(L, r_\perp) + \delta\varphi(L, r_\perp; t)$. Knowing the statistical properties of $\delta A(L, r_\perp; t)$ and $\delta\varphi(L, r_\perp; t)$ is sufficient to determine the optical spectrum $S(\Omega)$ of the field at the output of the waveguide, where $\Omega = \omega_0 - \omega$ is the difference between the angular frequency ω and the frequency ω_0 of the initial field, which is labeled as the pump hereafter. It requires solving the equation governing the field propagation and modeling the spatiotemporal fluctuations of the refractive index $\delta n(\vec{r}; t)$ along the waveguide.

A. Spatial Fluctuations of the Thermal Field

We consider a domain D of the medium whose volume is sufficiently small so that the spatial variation of the temperature is negligible and sufficiently large to be treated as a macroscopic

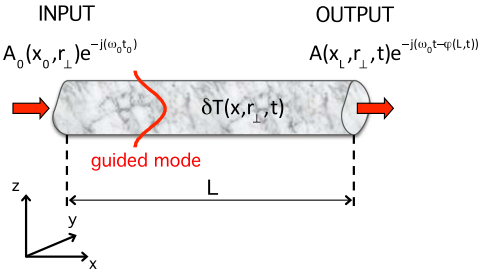

Fig. 3. Schematic representation of the propagating medium with the input and output optical fields. The energy distribution of the guided mode is represented by a Gaussian profile. The gray color pattern represents an arbitrary fluctuation of the temperature field $\delta T(x, r_\perp, t)$.

thermodynamic subsystem. The entire medium of density ρ and volume V is assumed to be incompressible and in thermal equilibrium with the environment at a temperature T_0. If the local domain D is also in equilibrium, as assumed in classical irreversible thermodynamics (CIT) [21], the fundamental thermal fluctuations can be determined from the knowledge of the fluctuations of the local specific entropy s, namely its second differential $\delta^2 s$. The thermal fluctuations are related to the standard deviation of the probability P_D of the macrostate in D. The incessant motion of the elementary charges that constitute the medium implies, however, that a given domain D is subject to a random heat flux. Consequently, the local-equilibrium hypothesis that is used in the CIT is not satisfied.

Such an issue can be overcome by defining a generalized specific entropy $s(u, \vec{q})$ that depends not only on the specific internal energy u but also on the local heat flux $\vec{q}$. This approach is at the heart of the extended irreversible thermodynamics [22]. Assuming that the specific entropy $s(u, \vec{q})$ is an additive quantity and a concave function, and that its rate of production is locally positive, its differential form can be expressed via a generalized Gibbs equation by defining a non-equilibrium temperature θ given by $\theta^{-1}(u, \vec{q}) = (\frac{\partial s}{\partial u})_{\vec{q}}$ [22]

$$ds = \frac{du}{\theta} - \frac{\tau}{\rho\kappa_0\theta^2}\vec{q}.d\vec{q}. \tag{1}$$

As we focus here on small fluctuations $\delta\vec{q}$ of the flux, the non-equilibrium temperature $\theta^{-1}(u, \vec{q})$ can be approximated by the local equilibrium temperature $T^{-1}(u)$. Considering the instantaneous value $q = -\kappa_0\nabla(T^s + \delta T) = -\kappa_0\nabla T$ [11] and using $u = C_V T$, a Taylor expansion of the change in entropy around equilibrium provides an expression for the second differential of the entropy $\delta^2 s$,

$$\delta^2 s(\delta u, \nabla\delta u) = -\frac{1}{C_V T^2}(\delta u)^2 - \frac{\tau\kappa_0}{\rho C_V^2 T^2}\nabla\delta u \cdot \nabla\delta u. \tag{2}$$

The spatial correlation of the energy field follows from Einstein's formula $P_D \approx \exp(\rho\delta^2 s(\delta u, \nabla\delta u)/2k_B)$. Using the spatial Fourier transform of the energy fluctuation $\delta u = \int \delta u_k e^{ikx}\mathrm{d}k$, the probability P_D of the macrostate is given by $P_D = \prod_k \exp(-\frac{\rho}{2}(\frac{1+\ell^2k^2}{C_V k_B T^2})|\delta u_k|^2)$, where the characteristic length ℓ is defined as

$$\ell = \left(\frac{\tau\kappa_0}{\rho C_V}\right)^{1/2} = \sqrt{\tau D_T}. \tag{3}$$

It follows that the variance of the Fourier components is $\langle|\delta u_k|^2\rangle = \frac{C_V k_B T^2}{\rho(1+\ell^2k^2)}$ and the spatial correlation can be determined from the Wiener–Khinchin theorem $\langle\delta u(\vec{r}_1)\delta u(\vec{r}_2)\rangle = \int\langle|\delta u_k|^2\rangle e^{i\vec{k}\cdot(\vec{r}_1-\vec{r}_2)}\mathrm{d}\vec{k}$, which results in

$$\langle\delta T(\vec{r}_1)\delta T(\vec{r}_2)\rangle = \frac{k_B T_0^2}{\rho C_V \ell^3}\left\{2\pi^2\frac{\ell}{|\vec{r}_1 - \vec{r}_2|}\ \exp\ \frac{-|\vec{r}_1 - \vec{r}_2|}{\ell}\right\}. \tag{4}$$

The spatial correlation of the fluctuations of the thermal field diverges for a spatial separation $|\vec{r}_1 - \vec{r}_2|$ that goes to zero. This behavior is linked to the minimal volume for which thermodynamic concepts apply, i.e., the minimal volume over which the energy and consequently the effective temperature have been considered spatially constant. The divergence can be circumvented by considering that $\langle\delta T(\vec{r}_1)\delta T(\vec{r}_2)\rangle$ is a sequence of functions parameterized by the characteristic length ℓ and converges toward the Dirac distribution $\frac{k_B T^2}{C_V}\delta(|\vec{r}_1 - \vec{r}_2|)$ when the characteristic length ℓ approaches zero [23,24]. As a result, the spatial correlation $\langle\delta T(\vec{r}_1)\delta T(\vec{r}_2)\rangle$ can be approximated by the following sequence of regular Gaussian functions that are parameterized by ℓ and that also converge towards the same $\frac{k_B T^2}{C_V}\delta(|\vec{r}_1 - \vec{r}_2|)$ function when ℓ tends to zero [24,25],

$$\langle\delta T(\vec{r}_1)\delta T(\vec{r}_2)\rangle = \frac{k_B T_0^2}{\rho C_V \ell^3}\left\{8\pi\sqrt{\pi}\ \exp\ \frac{-|\vec{r}_1 - \vec{r}_2|^2}{\ell^2}\right\}. \tag{5}$$

From a thermodynamic approach that takes into account local non-equilibrium by considering a non-vanishing relaxation time τ of the spontaneous heat flux, we can conclude that the thermal field is spatially inhomogeneous with a spatial correlation ℓ. The spatial correlation ℓ has a square root dependence with τ and is governed by the macroscopic thermal properties of the medium such as the heat conductivity and the heat capacity. The spatial inhomogeneity of the thermal field is at the origin of light scattering.

The motion of the elementary charges that shape the medium constantly induces different statistical realizations of the random thermal field that have always the same value of the correlation length. It results in a dynamic light scattering. In addition to its spatial correlation, the knowledge of the temporal dynamic of the thermal field is necessary to characterize the interaction between the propagating light and the medium.

B. Dynamics of the Thermal Field

As the thermal field fluctuates in space and time, its dynamics can be described by a velocity field as in standard hydrodynamic models. The dynamics is governed by the distribution $f(\vec{v})$ of the velocity field that is associated with the random motion of the elementary constituents of the medium.

In the framework of the generalized entropy [26], the velocity distribution $f(\vec{v})$ can be described by a generalized Boltzmann distribution. More specifically, it can be interpreted as the stationary non-thermalized solution of the Fokker–Planck equation, $f(\vec{v}) \propto \exp(-\frac{v^2}{2\tau_v D_v})$, where τ_v is the relaxation time of the average velocity, and D_v is a diffusion coefficient in the velocities space [27]. Defining the variance as $\sigma_v^2 = \tau_v D_v$, the velocity distribution can be written as $f(\vec{v}) \propto \exp(-\frac{v^2}{2\sigma_v^2})$. Such a velocity distribution combined with the spatial correlation of the thermal field results in the presence of thermo-refractive noise at high frequencies Ω, as discussed below.

C. Refractive Index Fluctuations

The presence of thermal fluctuations δT modifies the dielectric constant of the medium. The fluctuations of the dielectric function, which are at the heart of the Landau–Placzek theory of light scattering from hydrodynamic modes [28], will influence the propagation properties of the guided mode via the wave equation. Considering a single-mode waveguide, we have used a scalar model to describe the propagation of the wave. In addition, the index fluctuations are supposed to be much slower than the oscillations of the exciting field, i.e., the frequency bandwidth of the index fluctuations is regarded to be much smaller than the angular frequency of the exciting field. As a result, the wave equation can be written as the Helmholtz equation in terms of the refractive index $n(r, t) = \langle n\rangle + \delta n_0(r, t)$ and of the vacuum wavenumber k. For a waveguide with a thermo-optic coefficient $\frac{\partial n}{\partial T}$, the variance of the index fluctuations $\langle\delta n_0^2\rangle$ is directly deduced from the

equation for the variance of the temperature fluctuation and from the dielectric state equation. If the optical losses are negligible, it is given by $\langle \delta n_0^2 \rangle = (\frac{\partial n}{\partial T})^2 \langle \delta T^2 \rangle$.

The conventional treatment of the thermo-refractive noise does not provide any information about the spatial variations of the refractive index. Our approach shows that the spatial variations of the refractive index can be described by a Gaussian autocorrelation function [see Eq. (5)]. At two different positions, r_1 and r_2, the refractive index autocorrelation is $\langle \delta n_0(r_1) \delta n_0(r_2) \rangle = \langle \delta n_0^2 \rangle 8\pi\sqrt{\pi}\ \exp - \frac{|r_1 - r_2|^2}{\ell^2}$. Moreover, the dynamics of the fluctuations of the refractive index field are governed by the statistics of a velocity field that follows a Gaussian probability distribution function, with a standard deviation σ_v. The resulting light scattering problem is therefore similar to the one in turbid media.

D. Analytical Determination of the Optical Spectrum

The thermal fluctuations are expected to be sufficiently weak, which allows us to implement a first-order perturbation approach in order to achieve an analytical expression of the optical spectrum of the guided mode. Within this scope, the unperturbed lossless guided mode U_0^m propagates with a wavenumber $\overline{\beta} = kn_\phi$ and an envelope $A_0^m(x, r_\perp) = A_0^m(r_\perp)$. After the propagation of this mode over an infinitesimal distance dx of the perturbed waveguide, the resulting field can be expressed as $U(x + \mathrm{d}x) = U_0^m \exp(j(\overline{\beta} + k\delta n)dx)$, where we have defined $\delta n = (\partial n/\partial T + n_\phi \alpha_L)$, and α_L is the linear coefficient of thermal expansion. Its projection onto the field of the unperturbed guide mode, provides the fraction $\iint_{-\infty}^{+\infty} U(x + dx)(U_0^m)^* \mathrm{d}S$ of the field that stays within the guided mode after a propagation over dx, where the surface integral is performed in a plane transverse to the propagation. This fraction is assumed to be the same for the perturbed guided mode U^m of wavenumber β. It follows that $U^m(x + dx; t) \simeq \iint_{-\infty}^{+\infty} |A_0^m(r_\perp)|^2 (1 + jk\delta n dx) \mathrm{d}S \times \exp(j\overline{\beta} dx) \times U^m(x; t)$, where we used $\exp(jk\delta n dx) \simeq 1 + jk\delta n dx$. Considering that the envelope of the perturbed mode is constant over the distance dx, which is specific to a regime of light propagation in a disordered waveguide where a dispersion relationship is still valid [29], the variation of the phase of the mode $\Delta\varphi(L; t) = \int_0^L \beta(x; t)\mathrm{d}x - \overline{\beta} L$ at a distance L is given by

$$\Delta\varphi(L; t) = k \int_0^L \left\{ \iint_{-\infty}^{+\infty} |A_0^m(r_\perp)|^2 \delta n(x, \vec{r}_\perp; t) \mathrm{d}S \right\} \mathrm{d}x. \quad (6)$$

The random nature of δn is at the origin of the phase noise $\Delta\varphi(L; t)$ that modifies the shape of the Dirac-like optical spectrum of the guided mode of the unperturbed waveguide. The resulting optical spectrum of the guided field $U^m(L, \vec{r}_\perp; t)$ can be retrieved from its temporal autocorrelation function that is defined at times t_1 and t_2 as $B(L, r_\perp; t_1, t_2) = \langle U^m(L, r_\perp; t_1) U^m(L, r_\perp; t_2)^* \rangle$.

Assuming that the phase noise is sufficiently weak, a first-order Taylor series leads to $U^m(L, r_\perp; t) \approx A_0^m(r_\perp)(1 + j\Delta\varphi(L, t)))\exp(j(\overline{\beta} L - \omega_0 t))$. As a consequence, the autocorrelation at a distance L is a simple function of the phase fluctuations of the field, $B(L, r_\perp; t_1, t_2) = |A_0^m(r_\perp)|^2 (1 + \langle \Delta\varphi(L; t_1) \Delta\varphi(L; t_2) \rangle)$, which can be developed further with Eq. (6).

Using a two-dimensional spectral representation of the random index function, $\delta n(x, r_\perp; t) = \int_{\vec{\kappa}} \exp(j\vec{\kappa} \cdot \vec{r}_\perp) \mathrm{d}\nu(x, \vec{\kappa}; t)$ with $d\nu(x, \vec{\kappa}; t)$, the random amplitude of the stochastic Fourier–Stieltjes integral [30] and the Fourier transform of the square of the field envelope $\mathcal{A}(\vec{\kappa}) = \frac{1}{2\pi} \int_{\vec{r}_\perp} |A_0^m(r_\perp)|^2 \exp(-j\vec{\kappa} \cdot \vec{r}_\perp) \mathrm{d}\vec{r}_\perp$, the correlation function of the phase fluctuations can be simply expressed as a function of the time-varying spectral density Φ_n of the optical index fluctuations,

$$\langle \Delta\varphi(L; t_1) \Delta\varphi(L; t_2)^* \rangle = 2\pi k^2 L \int_{\vec{\kappa}} |\mathcal{A}(\vec{\kappa})|^2 \Phi_n(\vec{\kappa}; t_1, t_2) \mathrm{d}\vec{\kappa}. \quad (7)$$

For simplicity's sake, but without losing the effect of the main physical principles involved, the transverse envelope of the intensity of the guided mode is approximated with a Gaussian profile $\propto \exp(-|r_\perp|^2/W^2)$ of radial half-width W. It follows that $\mathcal{A}(\vec{\kappa}) = \frac{1}{2\pi} \exp(-\frac{\kappa^2 W^2}{4})$.

A crucial step of our model is the determination and physical interpretation of the spectral density Φ_n. The thermal dynamics is the result of the exchange of heat between adjacent domains that are at different temperatures. The average $\langle v \rangle$ of the associated velocity field linked to the thermal field is zero, and the statistics of the velocity field follows a Gaussian probability distribution function, with a standard deviation σ_v. Such a physical representation allows implementing the frozen flow hypothesis that is a standard approximation, for instance, in the analysis of turbulent atmosphere [31,32].

Within the frozen flow hypothesis, the time-varying spectral density can be written as the product of the spatial power spectrum and the power spectrum of the velocity fluctuations (see Supplement 1),

$$\Phi_n(\kappa, |t_1 - t_2|) = \langle \delta n^2 \rangle \ell^3 \exp\left(-\frac{\ell^2 + 2\sigma_v^2 |t_1 - t_2|^2}{4} \kappa^2 \right), \quad (8)$$

where $\langle \delta n^2 \rangle = (\frac{\partial n}{\partial T} + n_\phi \alpha_L)^2 \langle \delta T^2 \rangle$.

As a result, the optical spectrum is given by the sum of the initial spectrum, here a Dirac distribution $\delta(\Omega = \omega_0 - \omega)$, and the spectral contribution that comes from the thermal fluctuations. Defining the characteristic time γ as

$$\gamma = \frac{\sqrt{\ell^2 + 2\ W^2}}{\sqrt{2}\sigma_v}, \quad (9)$$

and using $k = \frac{2\pi}{\lambda_0}$ with λ_0 the wavelength of the pump, the optical spectrum can be written as

$$S(\Omega) = A_0^2 \left\{ \delta(\Omega) + 4\pi^2 \langle \delta n^2 \rangle \frac{L\ell}{\lambda_0^2} \frac{\ell^2}{\ell^2 + 2W^2} \gamma e^{-\gamma|\Omega|} \right\}, \quad (10)$$

which is the main theoretical result of our study. Note that the model neglects the intensity depletion at the pump frequency ω_0.

When ℓ, or equivalently, the mean relaxation time τ of the spontaneous heat flux, vanishes, the thermodynamic concepts at play in the current model do not apply anymore. In particular, the inequality $\tau \gg \hbar/(k_B T_0)$ is required in the framework of the theory of thermodynamic fluctuations as discussed in Section 112 of [2]. If such an inequality is not satisfied, a more advanced model based on quantum mechanics is necessary to describe the background.

As stressed by Fig. 2, Eq. (10) provides the main contribution to the optical spectrum in a high frequency range that corresponds to the spectral domain of Raman spectroscopy.

Our model does not describe the fluctuations at low frequencies, as the slow dynamics of the thermal diffusion has been neglected, which contrasts with the standard model for which the slow thermal diffusion is the main focus. A complete description of the optical spectrum at all frequencies Ω would require the consideration of the proper and complete spatial and time

correlations of the random heat flux $\delta q(r_1, t_1)\delta q(r_2, t_2)$ to solve the inhomogeneous heat equation, which is beyond the scope of the current paper.

E. Discussion of the Model Outcomes

The theoretical optical spectrum is subject to an exponential decay whose magnitude is set by the characteristic time γ. The physical origin of the frequency-dependent exponential decay and of γ can be understood from general statistical physics arguments as follows. We consider that the optical field is modulated by the distribution of the non-interacting domains D whose associated local optical index fluctuates at a specific frequency. In the case of a local thermal equilibrium, the probability to find a domain D vibrating at a frequency Ω within the ensemble of distinguishable domains is governed by the classical Boltzmann distribution. Via classical frequency mixing, it follows that the optical spectrum of the field $S(\Omega)$ is proportional to $\exp(-\frac{\hbar|\Omega|}{k_B T_0})$. The exponential decay of the noise is characterized by a universal parameter $\gamma_0 = \hbar/k_B T_0 \simeq 0.25 \times 10^{-13}$ s that depends only on the reduced Planck constant $\hbar$ and the Boltzmann constant k_B. Defining the cutoff frequency of the optical spectrum by the universal decay constant $1/\gamma_0$, the bandwidth of the field fluctuations is independent of the physical properties of the propagating medium for a system in thermal equilibrium. We will see in the following that the actual characteristic time γ is slightly smaller than γ_0, which is in line with the departure from the local thermal equilibrium approximation.

In contrast to the cutoff frequency γ_0 that is material independent, the amplitude of the optical spectrum $S(0)$ defined at $\Omega = 0$ without the initial pump contribution $\delta(\Omega)$, namely the background, depends on the material properties via the variance $\langle \delta n^2 \rangle$ of the index fluctuations, on the correlation length ℓ, on the propagation length L, and on the width W of the envelope of the guided mode. The intrinsic physical properties, such as the heat capacity, the density, and the thermo-optic coefficient, come into play via $\langle \delta n^2 \rangle$. Increasing the heat capacity and the density or decreasing the thermo-optic coefficient minimizes the contribution of the optical spectrum induced by the thermal noise. Note that the background amplitude scales as $1/\lambda_0^2$, which implies an increase of its impact at shorter wavelength.

As regards the structural parameters of the waveguide, the value of the modal width W compared to the correlation length ℓ defines two regimes. In the case of $W \gg \ell$, the amplitude of the spectrum $S(0)$ varies as $1/W^2$ and, by taking into account the $1/\ell^3$ dependence of $\langle \delta n^2 \rangle$, it is independent of the correlation length ℓ. Note that we have assumed that γ is independent of ℓ, which implies via Eq. (9) that the standard deviation σ_v of the velocities' field scales with ℓ. It follows that low-index-contrast waveguides can be more advantageous in terms of optical noise compared to high-index-contrast waveguides due to a larger mode envelope W.

In the other case, $W \ll \ell$, it appears that the optical spectrum is proportional to $1/\ell^2$. The correlation length of the disorder consequently plays a crucial role with respect to the background noise. The fabrication method of the medium determines the nature of the disorder and is therefore critical for minimizing the impact of the thermal noise.

The existing theory that describes the low-frequency thermal noise in optical fibers predicts a $1/\Omega^2$ variation of the optical spectrum in the high-frequency limit [8,9], which is incompatible with the exponential decay of the experimental optical spectrum observed in Fig. 4, as discussed in more details in the next section.

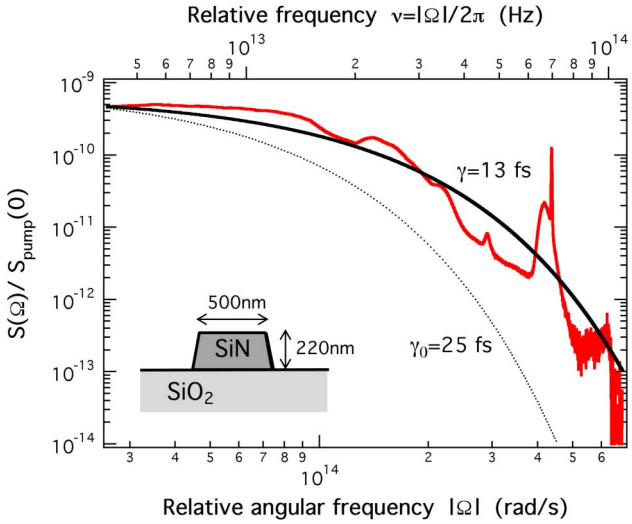

Fig. 4. Optical spectrum of a single-frequency pump laser beam after propagating through a 1 cm long silicon nitride waveguide (SiN). It is normalized according to the maximum of the laser optical spectrum $S_{\text{pump}}(0)$ at the input of the waveguide and plotted versus the frequency $\Omega < 0$ relative to the frequency of the laser beam at the input of the waveguide. The spectral resolution of the spectrometer is $\frac{d\omega_{\text{spectro}}}{2\pi} = 9 \times 10^{10}$ Hz. The inset shows a schematic of the cross section of the SiN waveguide on top of a 2 μm thick silicon oxide layer. Red curve: experimental data. Black curve: outcome of the model fit [see Eq. (10)]. Dotted curve: model fit with a characteristic time γ_0 fixed to be equal to $\hbar/(k_B T_0)$ and $T_0 = 298$ K.

The breakdown of the existing theory at high frequencies comes from the use of a Langevin source to solve the heat equation, which discards any spatial and temporal correlations of the fast microscopic dynamics of the random heat current. In particular, this theory does not take into account the microscopic structural disorder intrinsic to amorphous materials. It contrasts with our model that leads to Eq. (10). Our model describes the random nature of the heat flux by introducing the correlation volume ℓ^3, and it assumes that the local temporal fluctuation is caused by a local translation of the optical index value with a locally random velocity, which is the so-called "frozen-in" condition [30]. As an important consequence, the current model enables us to quantify the microscopic correlations of the random heat flux, in particular the mean relaxation time τ from the experimental characterization of the optical spectrum at terahertz frequencies, as described below.

F. Experimental Background of a Typical Raman Spectrum

In Fig. 4, the model is compared with the high-frequency part of the optical spectrum of an initially monochromatic light beam that has been collected at the output of a single-mode SiN channel waveguide. The waveguide has a length of $L = 1$ cm and a rectangular cross section of height $h = 0.22$ μm and width $w = 0.5$ μm, which leads to $W = 82$ nm. The classic coherent field is provided by a Ti:Sapphire continuous-wave (CW) laser, the spectral linewidth of which is smaller than 500 kHz and is negligible compare to the spectral resolution of the spectrometer. The field is coupled into the waveguide from free space via microscope objectives as explained in Ref. [15] in order to avoid any background

contribution coming from the propagation in optical fibers (see Supplement 1 for the determination of the noise floor and dynamic range of the Raman characterization setup). From the point of view of a material characterization, the measured spectrum corresponds to the Stokes part of the Raman spectrum of the SiN waveguide. The Raman spectrum is generally associated with well-defined spectral features that are characteristic of the local vibrations of atomic bonds. Such kind of spectral peaks are visible in Fig. 4, for instance at a frequency of $\nu = 70$ THz relative to the pump frequency. They appear on top of a broad background that exponentially decays with the frequency.

Fitting the theoretical optical spectra $S(\Omega)$ to the experimental Stokes spectrum by adjusting the characteristic time γ and the amplitude S^0 at zero frequency, the global shape of the background is well reproduced. The values of the fitted parameters of the model are $(S^0/S_{\text{pump}}, \gamma) = (6.5 \pm 0.5 \times 10^{-10}, 13 \pm 3$ fs). From these parameters, the knowledge of the spectral resolution of the spectrometer $\frac{d\omega_{\text{spectro}}}{2\pi} = 9 \times 10^{10}$ Hz and the SiN specific heat $C_V = 0.17$ Jg^{-1} K^{-1} [33], thermo optic coefficient $\frac{\partial n}{\partial T} = 5 \times 10^{-5}$ K^{-1} [34], linear coefficient of thermal expansion $\alpha_L = 3.9 \times 10^{-6}$ K^{-1} [35], effective index $n_\phi = 1.8$, and density $\rho = 2.5$ g/cm^3, the correlation length can be estimated to be $\ell = 200 \pm 50$ nm. This value corresponds to a silicon nitride material that was deposited via dense plasma-enhanced chemical vapor deposition (PECVD) and is one of the largest values that we have currently achieved. A 50% uncertainty on the experimental value of S^0, which mainly comes from the determination of the coupling loss at the input of the waveguide, results in a confidence interval defined between 145 and 300 nm for the correlation length.

Based on the reported heat conductivity of PECVD silicon nitride that spans between 16 and 33 Wm^{-1} K^{-1} [33], the current experimental value of ℓ corresponds to a mean relaxation time of the random heat flux τ that is between 0.4 ns and 1.4 ns. Such a mean relaxation time that is related to the second sound in solids is, in general, difficult to determine [19,36]. Importantly, the model elaborated in this article provides a new method to determine τ. We have remarked that the fabrication method and the parameters of the deposition process have a strong impact on the retrieved value of τ. The value reported here corresponds to a fabrication process that leads to the minimal background that we observed.

The experimental characteristic time γ is 1.9 times lower than the value expected from a statistical approach based on a Boltzmann distribution. Our model suggests that the dynamics of the heat transfer at the nanoscale is responsible for this unexpectedly low characteristic time. The heat transfer takes place between the different domains of typical volume ℓ^3 and at slightly different temperatures. Based on the current value of the correlation length, the standard deviation $\sigma_T = \sqrt{\langle (\delta T)^2 \rangle}$ of the temperature fluctuations among the different domains amounts to $\sigma_T \simeq 20$ mK.

From the value of γ, the standard deviation of the velocity field linked to the thermal field is $\sigma_v = 1.3 \times 10^6$ m/s. Such a value implies that relativistic effects can still be neglected, which reinforce the choice of a Boltzmann distribution for the velocity field.

The exponential shape of the experimental background in Fig. 4 is not limited to integrated SiN waveguides. As unveiled in Supplement 1, single-mode silica optical fibers exhibit a similar background. However, the level of this background is much lower, mainly due to a weaker confinement in optical fibers compared to single-mode SiN waveguides.

4. DISCUSSION

A. Microscopic Approach

The purpose of this section is to discuss approaches that treat the light–matter interaction at the level of the fundamental vibrational modes of the medium, also called phonons. In particular, we investigate which phonon-based model is able to reproduce the exponential shape of the experimental background.

When the vibrational modes are strongly localized and isolated, the system can be viewed as an ensemble of independent molecules. The Stokes spectrum can be determined from the correlation function of the fluctuations of the local optical dielectric tensor. Expressing the displacements of the vibrational modes in terms of normal coordinates and using the correlation properties of harmonic oscillators, the temporal correlations are determined from the Bose–Einstein occupation number $n(\Omega, T_0) = 1/(e^{\hbar\Omega/k_B T_0} - 1)$. As the density of states of molecules is discrete, the Stokes spectrum is promotional to the initial thermal population of the normal mode, $S^{\text{Stokes}}(\Omega) \propto 1/\Omega(1 + n(\Omega, T_0))$. The result of such an extreme approximation does not, however, account for the shape of the experimental background.

In amorphous solids, the phonon density of states $g_b(\Omega)$ has to be considered for each of the vibrational bands b of the normal modes, as discussed in Ref. [37]. If the product of the optical coupling tensor A_m and the correlation length Λ_m is assumed to be the same for each of the normal-mode vibrations m, and if Λ_m is at least ten times smaller than the wavelength ($\Lambda_m < \lambda/10$), the contribution of each band b to the Stokes spectrum is proportional to $A_m \Lambda_m^3/\Omega(1 + n(\Omega, T_0))g_b(\Omega)$ [37]. From an isotropic three-dimensional model, the density of states of the vibrational mode is expected to vary as Ω^2. The resulting shape of the Stokes spectrum is again not in line with the experimental spectrum, which invalidates the hypothesis of this approach. It follows that a microscopic approach needs to be much more elaborated to treat systems that are in an out-of-thermal-equilibrium state.

B. Prospect of the Macroscopic Model

Equation (10), which is the main result of our study, predicts a quadratic temperature variation of the noise contribution of the optical spectrum. We have tested this prediction against experiments by varying the temperature of the waveguide. A critical experimental aspect is to keep the light coupling into the nanophotonic waveguide stable enough during a temperature variation. To minimize light coupling issues, we have used a commercial Raman microscope that allows collecting the back-propagating light from the waveguide. Starting from room temperature, we have observed a clear quadratic increase of the background intensity as shown in Fig. 5. Such an observation strongly supports the thermodynamic origin of the spectral noise and the relevance of the current model.

The experimental optical spectrum at a fixed frequency has also been determined for different lengths of the SiN waveguide (see Fig. 6). In each case, the signal attenuation resulting from the propagation losses has been taken into account. All the data points are normalized to the value obtained for a 1 cm long waveguide. Figure 6 shows that the macroscopic model is in line with the

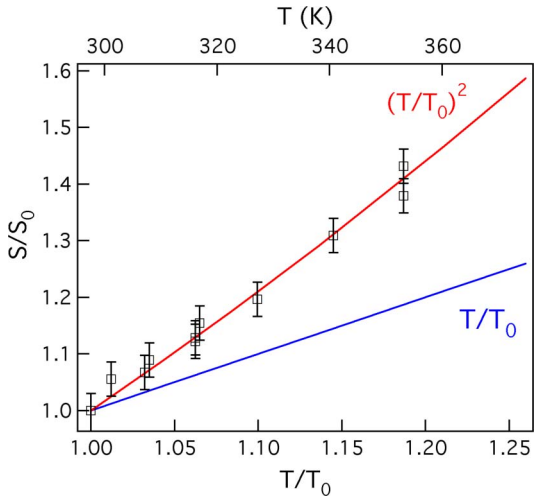

Fig. 5. Variation of the intensity S of the experimental spectral noise versus the temperature T normalized to the spectral noise S_0 at $T_0 = 297.6$ K. Squares with error bars: experimental data. Blue line: linear variation. Red line: quadratic variation. The error bars correspond to the alignment error of the light coupling into the waveguide.

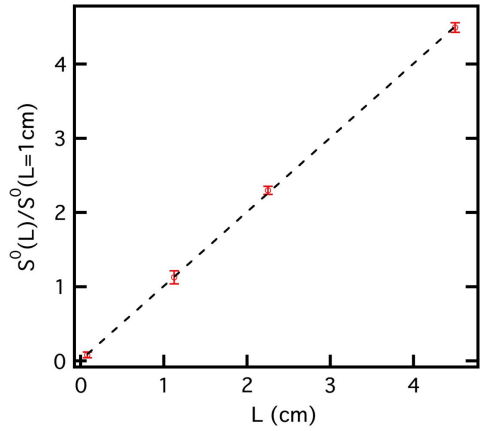

Fig. 6. Variation of the intensity S^0 of the experimental Stokes spectrum at $\Omega = 4 \times 10^{14}$ Hz versus the waveguide length. Dots with error bars: experimental data normalized to the value S^0 at a length of 1 cm. Dashed line: linear fit.

observed linear dependence of the optical spectrum with the propagation length.

5. CONCLUSION

We have modeled and characterized the impact of the thermodynamic fluctuations on the optical spectrum of a classical coherent field that propagates in silicon nitride optical waveguides. Our study suggests that the fundamental thermal fluctuations are at the origin of the background of the Raman signal that is collected through a guiding structure. The model relies on a macroscopic approach that considers the dielectric map of the waveguide subject to weak spatial and temporal perturbations. It reproduces satisfactorily the shape of the experimental Stokes spectrum and provides an estimate of the correlation length of the thermal fluctuations.

Importantly, our approach goes beyond the current treatment of the thermo-refractive noise in optical waveguides by considering the impact of random heat fluxes that are not "δ" correlated. It allows us to investigate the high-frequency regime of the thermo-refractive noise.

The benefit of the current approach is to highlight few macroscopic parameters that quantify the intensity value of the background, such as the specific heat and the correlation length of the thermal fluctuations. In addition, if the entire background originates from the proposed physical model, we have found a simple way to quantify the correlation length and the relaxation time of the thermal fluctuations in an amorphous solid that has been subject to specific conditions of fabrication. It may be of interest for the development of low-thermal-noise materials and should allow prediction of the limitations of different photonic structures such as interferometer-based integrated photonic sensors.

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See Supplement 1 for supporting content.

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