Supporting Information

Energy equipartition and unidirectional emission in a rotating spaser nanolaser

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Angular representation of the spaser emission

In the presence of a generic polarization $\mathbf{P}(\mathbf{x},t)$ term, which includes both linear and nonlinear effects, the total electromagnetic field evolves according to Maxwell equations, which for the electric field $\mathbf{E}(\mathbf{x},t)$ are expressed as follows:

$$\frac{\partial^2}{\partial t^2}\mathbf{E}(\mathbf{x},t) = -\frac{c^2}{\varepsilon(\mathbf{x})}\nabla^2\mathbf{E}(\mathbf{x},t) - \frac{\partial^2}{\partial t^2}\mathbf{P}(\mathbf{x},t). \tag{S1}$$

The solution of Eq. (S1) can be formulated in terms of angular modulated plane waves $\psi_\alpha(\mathbf{x})$:

$$\psi_\alpha(\mathbf{x}) = \exp\left[-jk(\cos\alpha x + \sin\alpha\,|z|)\right]. \tag{S2}$$

In Eq. (S2), $\psi_\alpha(\mathbf{x})$ denotes an outgoing propagating plane wave, whose wave-vector $\mathbf{k} = k\hat{\boldsymbol{\alpha}} = k(\cos\alpha, \sin\alpha)$ is parallel to the angle α measured from the x axis. The plane waves in Eq. (S2) form an orthonormal basis for the electromagnetic fields with respect to the following scalar product:

$$\langle\psi_\alpha(\mathbf{x}), \psi_\beta(\mathbf{x})\rangle = \int \mathrm{d}\mathbf{x}\,\psi^*_\alpha(\mathbf{x})\psi_\beta(\mathbf{x}) = \delta(\alpha-\beta), \tag{S3}$$

and the total field expansion in terms of $\psi_\alpha(\mathbf{x})$ reads:

$$\mathbf{E}(\mathbf{x},t) = \hat{\mathbf{e}}E(\mathbf{x},t) = \hat{\mathbf{e}}\sum_\alpha A_\alpha(t)\psi_\alpha(\mathbf{x}) + \text{c.c.}, \tag{S4}$$

where c.c. indicates the complex conjugation, $\hat{\mathbf{e}}$ is a unit polarization vector directed along $\mathbf{E}(\mathbf{x},t)$ and:

$$A_\alpha(t) = \langle\psi_\alpha(\mathbf{x}), \mathbf{E}(\mathbf{x},t)\rangle = a_\alpha(t)e^{i\omega_0 t}, \tag{S5}$$

constitutes a set of complex amplitudes, slowly modulated by the envelope function $a(t)$ and rapidly oscillating at ω_0, which denotes the spaser emission frequency. An important relationship between the complex amplitude terms $A(t)$ and the far-field expression of the electric field $\mathbf{E}(x,z,t)$ can be derived by generalizing the approach introduced in [1]. We begin from the Fourier transform of the total field at $z=0$:

$$\tilde{\mathbf{E}}(k_x,0,t) = \frac{1}{2\pi}\int_{-\infty}^{\infty}\mathrm{d}x\,\mathbf{E}(x,0,t)\exp(jk_x x). \tag{S6}$$

When the latter expression is propagated for $z \neq 0$ and transformed back to the spatial coordinates (x,z), it furnishes the expression of the total electric field:

$$\mathbf{E}(x,z,t) = \int \mathrm{d}k_x\,\tilde{\mathbf{E}}(k_x,0,t)\exp[-j(k_x x + k_z|z|)]. \tag{S7}$$

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Eq. (S7) is standardly referred as the angular representation of the electromagnetic field $\mathbf{E}(\mathbf{x},t)$, and is valid both in the near and far field regions [2]. By imposing the far-field condition, $k_x^2 \leq k^2$, equation (S7) can be solved by means of a saddle-point integration, obtaining the fundamental relationship:

$$\mathbf{E}_{r\to\infty}(\alpha,t) = -i2\pi A_\alpha(t)\frac{e^{-ikr}}{r}\hat{\mathbf{e}}, \tag{S8}$$

which connects the evolution of the angular amplitudes $A_\alpha(t)$ with the electric field contribution $\mathbf{E}_{r\to\infty}(\alpha,t)$ in the far-field. As discussed in the main text, the latter can be directly extracted from first principle calculations, hence allowing for a direct measurement of the angular amplitudes evolution.

Expansion of Maxwell-Bloch equations in angular plane-wave modes

By substituting the expansion (S4) into (S1), we obtain:

$$\hat{\mathbf{e}}\sum_\alpha\left[\frac{\omega_0^2}{\varepsilon(\mathbf{x})}A_\alpha(t)+\frac{d^2A_\alpha(t)}{dt^2}t\right]\psi_\alpha(\mathbf{x})+\text{c.c.} = -\frac{\partial^2}{\partial t^2}\mathbf{P}(\mathbf{x},t), \tag{S9}$$

where we have used the dispersion relation $\omega_0 = ck$. Equation (S9) is a general expression for the evolution of the emitted field. In the case of the spaser, we can further specify the polarization term $\mathbf{P}(\mathbf{x},t) = \mathbf{P}_l(\mathbf{x},t)+\mathbf{P}_{nl}(\mathbf{x},t)$ by considering its linear $\mathbf{P}_l$ and nonlinear $\mathbf{P}_{nl}$ contributions. The linear polarization term accounts for the losses $\sigma(\mathbf{x})$ occurring in the metal $\partial\mathbf{P}_l/\partial t = \sigma(\mathbf{x})\mathbf{E}(\mathbf{x},t)$. This originates a dissipative term in the equations of motion:

$$\hat{\mathbf{e}}\sum_\alpha\left[\frac{\omega_0^2}{\varepsilon(\mathbf{x})}A_\alpha+\frac{d^2A_\alpha}{dt^2}+\sigma(\mathbf{x})\frac{dA_\alpha}{dt}\right]\psi_\alpha(\mathbf{x})+\text{c.c.} = -\frac{\partial^2}{\partial t^2}\mathbf{P}_{nl}(\mathbf{x},t). \tag{S10}$$

The nonlinear term P_{nl} appearing in (S10) models the nonlinear process of light-matter interaction occurring in the spaser. We express this term by using a Maxwell-Bloch (MB) model for a two-level atomic system. In the MB system, the dynamics of the resonant medium can be described by employing the density matrix formalism. Detailed calculations are found in [3,4], hence, we here limit to report the main equations for the components ρ_{ij} of the density matrix. If we denote by N_a the density of polarizable atoms in the medium and by q_0 the typical atomic displacement, the MB nonlinear polarization reads:

$$\mathbf{P}_{nl}(\mathbf{x},t) = \hat{\mathbf{e}}q_0N_a\rho_{12}(\mathbf{x},t)+\text{c.c.} \tag{S11}$$

where $\rho_{12}(\mathbf{x},t)$ and $\rho_{21}(t) = \rho_{12}^*(\mathbf{x},t)$ is the density matrix element describing the transition rate between the atomic levels of the two-level atoms and where, following standard approaches, we assumed the polarization field $\mathbf{P}_{nl}$ to be parallel to the electromagnetic field $\mathbf{E}$. T=Such standard approach can be easily extended to the case of The evolution of the density matrix elements follows the standard Bloch equations:

$$\begin{cases}\dfrac{\partial\rho_{12}(\mathbf{x},t)}{\partial t}+\left(\gamma_{12}-i\omega_0\right)\rho_{12}(\mathbf{x},t) = i\dfrac{q_0}{\hbar}E(\mathbf{x},t)\Delta\rho(\mathbf{x},t),\\[2ex] \dfrac{\partial\Delta\rho(\mathbf{x},t)}{\partial t}+\gamma_{11}\left[\Delta\rho(\mathbf{x},t)-\Delta\rho_0\right] = 2i\dfrac{q_0}{\hbar}E(\mathbf{x},t)[\rho_{12}(\mathbf{x},t)-\rho_{12}^*(\mathbf{x},t)],\end{cases} \tag{S12}$$

where $\Delta\rho(\mathbf{x},t) = \rho_{22}(\mathbf{x},t)-\rho_{11}(\mathbf{x},t)$ defines the difference in population between the two atomic levels, ω_0 is the atomic resonance frequency and $\tau_{ij} = 1/\gamma_{ij}$ are the atomic relaxation times. Contrarily to the electric field case, we here expand the density matrix elements ρ_{12} by using the Feshbach projection technique, which introduces a set of orthogonal modes $\phi_n(\mathbf{x})$, $\xi_m(\mathbf{x},t)$ defined inside and outside the cavity system, respectively. The Feshbach projection technique is introduced and thoroughly described in [5], hence, we here limit to present the main results remanding to Ref. [5] for the calculation of the eigenmodes ϕ_n and ξ_m as well as the mathematical demonstration of their orthogonality, which follows from the following relations:

$$\begin{aligned}\langle\phi_n,\varepsilon\phi_{n'}\rangle &= \int \mathrm{d}x\,\phi_n(\mathbf{x})^*\varepsilon(\mathbf{x})\phi_{n'}(\mathbf{x}) = \delta_{nn'},\\ \langle\xi_m,\varepsilon\xi_{m'}\rangle &= \int \mathrm{d}x\,\xi_m(\mathbf{x})^*\varepsilon(\mathbf{x})\xi_{m'}(\mathbf{x}) = \delta_{mm'},\end{aligned} \tag{S13}$$

This choice of eigenmodes for the expansion of the density matrix element ρ_{12} is particularly advantageous, due to the fact that this component is defined only inside the cavity space and therefore requires only one set of modes to be completely described. With this position, the expansion of ρ_{12} reads as follows:

$$\rho_{12}(\mathbf{x},t) = \sum_n R_n(t)\phi_n(\mathbf{x}), \tag{S14}$$

with $R_n(t)$ being dynamical amplitudes coefficients. By substituting Eqs. (S11) and (S14) into (S10) and (S12), we obtain the full set of equations that describes the spaser dynamics:

$$\begin{cases} \sum_\alpha \left[\left(2+\frac{\sigma}{i\omega_0}\right)\dot{a}_\alpha + \sigma a_\alpha - i\omega_0\left(\frac{1-\varepsilon}{\varepsilon}\right)a_\alpha\right]\psi_\alpha = -i\omega_0 N_a q_0 \sum_n \rho_n\phi_n, \\ \sum_n \rho_n\phi_n = i\frac{q_0}{\hbar\gamma_{12}}\sum_\alpha \Delta\rho\cdot a_\alpha\psi_\alpha \\ \Delta\rho = \Delta\rho_0 + \frac{2iq_0}{\hbar\gamma_{11}}\sum_{\alpha,n}(a^*_\alpha\rho_n\psi^*_\alpha\phi_n - \text{c.c.}) \end{cases} \tag{S15}$$

where we have used the shorthand symbol $\dot{f} = \mathrm{d}f/\mathrm{d}t$, and where we have generalized the standard approach explained in [3,4] to simplify the Maxwell-Bloch equations, which introduces slowly modulated envelope variables $a_\alpha(t)$ [see Eq. (S5)] and $\rho_n(t)$:

$$R_n(t) = \rho_n(t)e^{i\omega_0 t}, \tag{S16}$$

and applies the quasi-static approximation for the variation of the populations and coherence, i.e., $\partial\Delta/\partial t \approx 0$, $\partial\rho_{12}/\partial t \approx 0$, which evolve on scales much longer than the electromagnetic field. By using the expression of the population $\Delta\rho$ found in the last of Eqs. (S15), we can obtain a single equation for the dynamics of the coherence terms $\rho_n(t)$:

$$\rho_n = i\frac{q_0\Delta\rho_0}{\hbar\gamma_{12}}\sum_\alpha C_{n\alpha}a_\alpha - \frac{2q_0^2}{\hbar^2\gamma_{11}\gamma_{12}}\sum_{\alpha\alpha'n'}\left[J_{\alpha\alpha'nn'}a_\alpha a^*_{\alpha'}\rho_{n'} - G_{\alpha\alpha'nn'}a_\alpha a_{\alpha'}\rho^*_{n'}\right], \tag{S17}$$

which is obtained by projecting the second of Eqs. (S15) on the mode ϕ_n. This projection originates the following coupling terms in Eq. (S17):

$$\begin{cases} C_{n\alpha} = \int \mathrm{d}\mathbf{x}\,\varepsilon(\mathbf{x})\psi_\alpha\phi^*_n = \varepsilon_g\int \mathrm{d}\mathbf{x}\,\psi_\alpha\phi^*_n, \\ J_{\alpha\alpha'nn'} = \int \mathrm{d}\mathbf{x}\,\varepsilon(\mathbf{x})\psi_\alpha\psi^*_{\alpha'}\phi^*_n\phi_{n'}, \\ G_{\alpha\alpha'nn'} = \int \mathrm{d}\mathbf{x}\,\varepsilon(\mathbf{x})\psi_\alpha\psi_{\alpha'}\phi^*_n\phi^*_{n'}, \end{cases} \tag{S18}$$

with ε_g the dielectric constant of the gain medium, assumed uniform in the spatial region where the density matrix elements ρ_{ij} are defined. The resulting equations can be cast into dimensionless form by rescaling the dynamical variables:

$$t \Longrightarrow t\left(\frac{2-i\frac{\sigma}{\omega_0}}{\omega_0}\right) \qquad a_\alpha \longrightarrow a_\alpha\left(\frac{\hbar\gamma_{12}}{\sqrt{2}q_0}\right) \qquad \rho_n \longrightarrow \rho_n\left(\frac{\hbar\gamma_{12}}{\sqrt{2}N_a q_0^2}\right), \tag{S19}$$

which yields the following set of equations:

$$\begin{cases} \dot{a}_\alpha + \sum_{\alpha'} K_{\alpha\alpha'}a_{\alpha'} = -i\sum_n D_{n\alpha}\cdot\rho_n, \\ \rho_n = ig\sum_\alpha C_{n\alpha}\cdot a_\alpha - \tau\sum_{\alpha\alpha'n'}\left[J_{\alpha\alpha'nn'}a_\alpha a^*_{\alpha'}\rho_{n'} - G_{\alpha\alpha'nn'}a_\alpha a_{\alpha'}\rho^*_{n'}\right], \end{cases} \tag{S20}$$

where $g = N_a\Delta\rho_0 q_0^2/\hbar\gamma_{12}$ is the gain factor, $\kappa = \sigma/\omega_0$ are the rescaled mode losses, $\tau = \gamma_{12}/\gamma_{11}$ is a dimensionless number that represents the degree of nonlinear interactions in the system response and:

$$K_{\alpha\alpha'} = K^*_{\alpha'\alpha} = \int \mathrm{d}\mathbf{x}\left[\frac{\sigma(\mathbf{x})}{\omega_0} + i\frac{1-\varepsilon(\mathbf{x})}{\varepsilon(\mathbf{x})}\right]\psi_{\alpha'}(\mathbf{x})\psi_\alpha(\mathbf{x})^*, \qquad D_{n\alpha} = \int \mathrm{d}\mathbf{x}\,\psi^*_\alpha(\mathbf{x})\phi_n(\mathbf{x}) \tag{S21}$$

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are linear coupling coefficients. The matrix elements $K_{\alpha\alpha'}$ can be further specified by expanding the permittivity $\varepsilon(\mathbf{x})$ and conductivity $\sigma(\mathbf{x})$ of the spaser as follows:

$$\varepsilon(\mathbf{x}) = \begin{cases} \varepsilon_g, & |\mathbf{x}| \leq a, \\ 1, & |\mathbf{x}| > a, \end{cases} \quad , \qquad \sigma(\mathbf{x}) = \begin{cases} \sigma, & |\mathbf{x}| \leq a, \\ 0, & |\mathbf{x}| > a, \end{cases} \quad , \tag{S22}$$

being a the radius characterizing the spaser nanoparticle. Due to the nanoscale volume of the spaser, we have $a \ll \lambda$ and we can neglect the contributions for $|\mathbf{x}| < a$, obtaining $K_{\alpha\alpha'} = (i+\kappa)\delta_{\alpha\alpha'}$, with $\kappa = \sigma/\omega_0$. The inclusion of off-diagonal contributions provides only perturbative corrections, which are inessential to understand the main dynamical emission properties of the spaser. The nonlinear coupling integrals $J_{\alpha\alpha' nn'}$ and $G_{\alpha\alpha' nn'}$, conversely, possess a leading order contribution from the term $J_{\alpha\alpha nn}$:

$$J_{\alpha\alpha nn} = \int \mathrm{d}\mathbf{x}\, \varepsilon(\mathbf{x})\, |\phi_n|^2 = 1, \tag{S23}$$

which yields the normalization condition (S13). All the remaining terms $J_{\alpha\neq\alpha' n\neq n'}$, due to the presence of oscillatory terms $\psi_{(\alpha-\alpha')}$ in the integral, are then $J_{\alpha\neq\alpha' n\neq n'} < J_{\alpha\alpha nn}$, . The same consideration applies for $G_{\alpha\alpha' nn'} < J_{\alpha\alpha nn}$, due to the presence of the oscillatory functions $\psi_{\alpha+\alpha'}$. We can therefore write:

$$J_{\alpha\alpha' nn'} = J_{\alpha\alpha nn}\delta_{\alpha\alpha'}\delta_{nn'} + \varepsilon J_{\alpha\neq\alpha', n\neq n'}, \qquad G_{\alpha\alpha' nn'} = \varepsilon G_{\alpha\alpha' nn'}, \tag{S24}$$

with $\varepsilon < 1$. With the aid of Eqs. (S24), we can solve Eqs. (S20) iteratively by expanding $\rho_n = \rho_n^0 + \varepsilon\rho_n^1 + O(\varepsilon^2)$. At order $\mathscr{O}(\varepsilon)$, we obtain a single set of equations for the variables $a_\alpha(t)$:

$$\dot{\mathbf{A}} + i\mathbf{A} + \kappa\mathbf{A} = g\frac{\chi^{(1)} \cdot \mathbf{A}}{1+\tau I} + \varepsilon g\tau \frac{\chi^{(3)} \vdots \mathbf{A}\mathbf{A}\mathbf{A}^*}{(1+\tau I)^2} \tag{S25}$$

where $I = \sum_\alpha |a_\alpha|^2$ the total energy emitted by the spaser, $\chi^{(3)}$ rank-2 and rank-4 tensors, respectively, with components $\chi^{(1)}_{\alpha\alpha'}$, $\chi^{(3)}_{\alpha\alpha'\alpha''\alpha'''}$ defined as follows:

$$\chi^{(1)}_{\alpha\alpha'} = \sum_n D_{n\alpha} C_{n\alpha'} = \varepsilon_g \sum_n C_{n\alpha} C_{n\alpha'}, \tag{S26}$$

$$\chi^{(3)}_{\alpha\alpha'\alpha''\alpha'''} = -\sum_{n,n'} \left[C_{n\alpha'''} G_{\alpha\alpha' nn'} + C^*_{n\alpha} J_{\alpha'''\neq\alpha'', n\neq n'} \right]. \tag{S27}$$

Equation (S26) can be simplified by rescaling $\mathbf{A} \to \mathbf{A}e^{-it}$, obtaining:

$$\dot{\mathbf{A}} + \kappa\mathbf{A} = g\frac{\chi^{(1)} \cdot \mathbf{A}}{1+\tau I} + \varepsilon g\tau \frac{\chi^{(3)} \vdots \mathbf{A}\mathbf{A}\mathbf{A}^*}{(1+\tau I)^2}, \tag{S28}$$

which represents Eq. (6) of the main text. In Eq. (S28), the symbols $\cdot$ and $\vdots$ represent tensorial products, which follow the standard convention used in nonlinear optics (see, e.g., [6] for additional details).

Change of basis in the equations of motion

For the analysis of Eq. (S28), is convenient to move to a angular basis where the Hermitian tensor $\chi^{(1)}$ appears diagonal. By indicating the eigenvalues and eigenvectors of $\chi^{(1)}$ as ξ_n and φ_n, respectively:

$$\chi^{(1)}\varphi_n = \xi_n\varphi_n \tag{S29}$$

and by applying the following coordinates change:

$$\mathbf{B} = T^{(1)} \cdot \mathbf{A}, \tag{S30}$$

being $T^{(1)}$ a rank-2 tensor containing the eigenvectors of the coupling matrix $\chi^{(1)}$, we obtain:

$$\dot{\mathbf{B}} + \kappa\mathbf{B} = g\frac{\xi^{(1)} \cdot \mathbf{B}}{1+\tau I} + \varepsilon g\tau \frac{\xi^{(3)} \vdots \mathbf{B}\mathbf{B}\mathbf{B}^*}{(1+\tau I)^2}, \tag{S31}$$

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with:

$$\xi^{(1)}_{\alpha\alpha'} = \xi_\alpha \delta_{\alpha\alpha'}, \tag{S32}$$

and $\xi^{(3)}$ a new transformed rank-4 tensor with components $\xi^{(3)}_{\alpha\alpha'\alpha''\alpha'''}$:

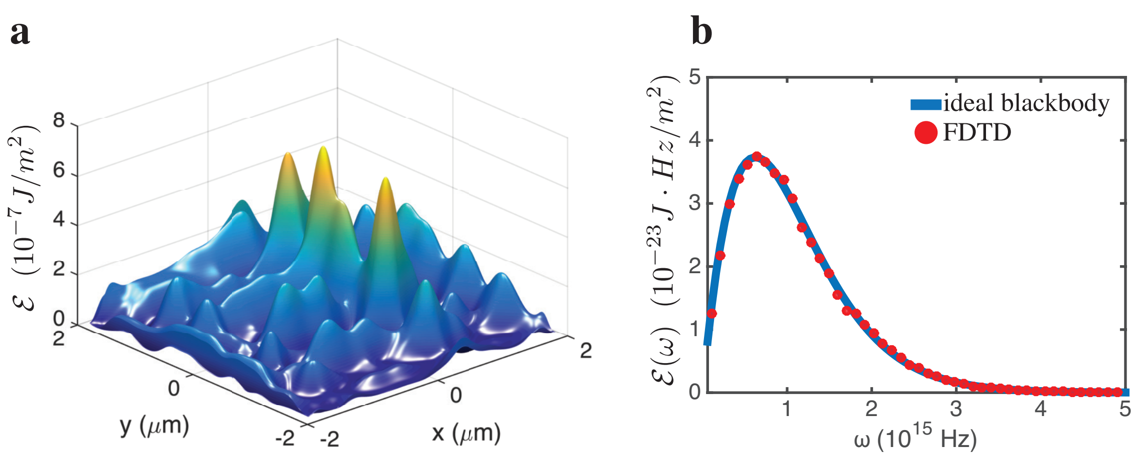
$$\xi^{(3)}_{\alpha\alpha'\alpha''\alpha'''} = \sum_{\beta\beta'\beta''\beta'''} \chi^{(3)}_{\beta\beta'\beta''\beta'''} T^{(1)}_{\alpha\beta} T^{(1)}_{\alpha'\beta'} T^{(1)}_{\alpha''\beta''} T^{(1)}_{\alpha'''\beta'''} \tag{S33}$$

Numerical implementation of quantum noise in MB-FDTD simulations

In our FDTD simulations, we introduced quantum noise by considering the field fluctuations generated by an external blackbody source, generalizing to the 2D and 3D the 1D approach introduced in [7]. This is based on the observation that the Uniaxial Perfectly Matched Layer (UPML) acts as a blackbody material, absorbing all energy that impinges on it. At thermal equilibrium, the UPML emits thermal radiation inside the FDTD computational grid, acting as a source of quantum noise for all components of the electromagnetic field. The power density spectrum $P(\omega)$ of the radiated energy corresponds to that of a blackbody radiation, whose expression in two and three dimensions reads as follows:

$$P_{2D}(\omega,T) = \frac{\hbar\omega^2}{\pi c^2 (e^{\frac{\hbar\omega}{k_B T}} - 1)}, \qquad P_{3D}(\omega,T) = \frac{\hbar\omega^3}{\pi^2 c^3 (e^{\frac{\hbar\omega}{k_B T}} - 1)}. \tag{S34}$$

In Eqs. (S34), k_B and $\hbar$ are the Boltzmann's and Planck's constants, respectively, while T is the temperature of equilibrium of the blackbody source. In Supplementary Figure 1-a we show a typical realization of quantum noise in an empty $4\mu m \times 4\mu$ box for an example temperature $T = 3000K$, while in Supplementary Figure 1-b we illustrate the average power density of the system as calculated by averaging over 500 different FDTD simulations.



Supplementary Figure 1 Simulation of quantum noise in the MB-FDTD algorithm. Panel (a) shows an example of the spatial distribution of electromagnetic energy corresponding to quantum noise at $T = 3000K$. Panel (b) shows the average power spectral density of the fluctuating field (red circles) as computed from 200 FDTD simulations and compared to Eqs. (S34) (solid blue line).

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