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Radiative heat transfer in nonlinear Kerr media

Chinmay Khandekar,¹ Adi Pick,² Steven G. Johnson,³ and Alejandro W. Rodriguez¹

¹*Department of Electrical Engineering, Princeton University, Princeton, NJ 08540*

²*Department of Physics, Harvard University, Cambridge, MA 02138*

³*Department of Mathematics, Massachusetts Institute of Technology, Cambridge, MA 02139*

We obtain a fluctuation–dissipation theorem describing thermal electromagnetic fluctuation effects in nonlinear media that we exploit in conjunction with a stochastic Langevin framework to study thermal radiation from Kerr ($\chi^{(3)}$) photonic cavities coupled to external environments at and out of equilibrium. We show that that in addition to thermal broadening due to two-photon absorption, the emissivity of such cavities can exhibit asymmetric, non-Lorentzian lineshapes due to self-phase modulation. When the local temperature of the cavity is larger than that of the external bath, we find that the heat transfer into the bath exceeds the radiation from a corresponding linear black body at the same local temperature. We predict that these temperature-tunable thermal processes can be observed in realistic, nanophotonic cavities operating near room temperature.

PACS numbers:

I. INTRODUCTION

The radiative properties of bodies play a fundamental role on the physics of many naturally occurring processes and emerging nanotechnologies^{1,2}. Central to the theoretical understanding of these electromagnetic fluctuation effects is the fluctuation-dissipation theorem of electromagnetic fields, developed decades ago by Rytov and others^{3–5} in order to describe radiative transport in macroscopic media. The same formalism has been recently employed in combination with new theoretical techniques^{6,7} to demonstrate strong modifications of the thermal properties of nanostructured bodies, including designable selective emitters⁸ and greater than black-body heat transport between bodies in the near-field⁹. To date, these studies have focused primarily on linear media, where emission depends only on the linear response functions of the underlying materials. A cubic ($\chi^{(3)}$) nonlinearity, however, can convert light from one frequency to another or alter the dissipation rate¹⁰ and hence the fluctuation statistics. We show that these phenomena lead to a variety of interesting effects in nonlinear radiators, such as lineshape alterations, temperature-dependent emission, and even radiation exceeding the black-body limit in nonequilibrium systems.

In this work, we obtain a nonlinear fluctuation–dissipation theorem (FDT) to describe radiative thermal effects in nonlinear $\chi^{(3)}$ media, extending previous work on nonlinear oscillators¹¹. Since nonlinear optical effects are generally weak in bulk materials, we focus on nanostructured resonant systems with strong effective nonlinear interactions^{10,12}. Such systems are susceptible to universal descriptions based on the coupled-mode theory framework^{13,14}, which we exploit to investigate the ways in which nonlinearities can enable interesting/designable radiative effects. In particular, we show that self-phase modulation (SPM) and two-photon absorption (TPA) effects lead to strong modifications of their emissivity, including thermal broadening and non-Lorentzian, asymmetric lineshapes. These nonlinear effects pave the way for additional material tunability, including designable, temperature-dependent selective emitters and absorbers. We also consider nonequilibrium situations and show that TPA results in selective heat transfer exceeding the black-body limit, a phe-

nomenon that has only been observed in situations involving multiple bodies in the near-field⁹. Finally, we show that recently proposed, wavelength-scale cavities with ultra-large $Q \lesssim 10^8$ and small mode volumes $V \sim (\lambda/n)^3$ can be designed to display these strongly nonlinear effects at infrared wavelengths and near room temperature.

Fluctuation–dissipation relations in nonlinear media have been a subject of much interest in recent decades, starting with the early work of Bernard and Callen¹⁵, Stratonovich¹⁶, and Klimontovich¹⁷. The effects of nonlinearities of both conservative and dissipative nature on the Brownian motion of resonant systems have been studied in the context of Van der Pol oscillators¹⁷, optomechanical systems¹⁸, and mechanical Duffing oscillators^{11,19}. Despite the relatively large body of work on noise in nonlinear systems, the role and consequences of nonlinear damping in mechanical oscillators have only recently begun to be explored^{19,20}, and there remains much to be known about the underlying physical mechanisms in such systems. The effects of nonlinear noise are also non-negligible and of great importance in a variety of applications, e.g. MEMS sensors²¹, frequency stabilization²², frequency mixing²³, and filtering²⁴. While there is increased interest in studying nonlinear effects in micro and nano-mechanical oscillators, studies of nonlinear effects on thermal radiation remain scarce and are largely restricted to driven systems with conservative nonlinearities e.g. resonators based on RF-driven Josephson junctions²⁵ or optomechanical oscillators²⁶. (The situation is different in the quantum regime, where the effects of SPM on the tunneling rate and quantum statistics of photons have been well studied.)^{27,28} Following an approach analogous to the treatment of nonlinear friction in mechanical oscillators¹¹, we extend previous work on Duffing oscillators to the case of nonlinear photonic cavities coupled to external baths/channels, a situation of direct relevance to current-generation experiments on radiative thermal transport in photonic media²⁹. Interestingly, we find that effects arising from the interference of radiation reflecting and emitted from the cavity into the external bath are crucial in order to observe thermal radiation enhancements in realistic situations, such as in cases where the external-bath temperature is at or near room temperature. We believe that these photonic systems not only

offer new opportunities for understanding the role of nonlinear damping on fluctuations, but also greatly extend the functionality and tunability of devices based on thermal radiation. As we argue below, while these effects require very strong optical nonlinearities, the increasing accessibility of ultra-high Q resonators with small modal volumes^{12,30–33}, such as the nanobeam cavity explored below, offers hope that they are within the reach of experiments.

II. LANGEVIN FRAMEWORK

We begin by introducing the Langevin equations of motion of a single-mode nonlinear $\chi^{(3)}$ cavity coupled to an external bath (a single output channel) and an internal reservoir (a lossy channel). As described in Ref. 34, the coupled-mode equations for the field amplitude are given by:

$$\frac{da}{dt} = [i(\omega_0 - \alpha|a|^2) - \gamma]a + \sqrt{2\gamma_e}s_+ + D\xi, \quad (1)$$

$$s_- = -s_+ + \sqrt{2\gamma_e}a, \quad (2)$$

where $|a|^2$ is the mode energy, $|s_{\pm}|^2$ are the input/output power from/to the external bath (e.g. a waveguide), and ω_0 and $\gamma = \gamma_e + \gamma_d$ are the frequency and linear decay rate of the mode. The linear decay channels include linear absorption from coupling to phonons or other dissipative degrees of freedom (γ_d) as well as decay into the external environment (γ_e). The real and imaginary parts of the nonlinear coefficient α are given by the overlap integral $\alpha = \frac{3}{4}\omega_0 \int \varepsilon_0 \chi^{(3)} |\mathbf{E}|^4 / (\int \varepsilon |\mathbf{E}|^2)^2$ of the linear cavity fields \mathbf{E} and lead to SPM and TPA, respectively.³⁴ In addition to radiation coming from the external bath $\sim s_+$, Eq. (1) includes a stochastic Langevin source $D\xi(t)$ given by the product of a normalized “diffusion coefficient” D , relating amplitude fluctuations to dissipation from the internal (phonon) reservoir, and a time-dependent stochastic process $\xi(t)$ whose form and properties can be derived from very general statistical considerations^{16,35,36}. For linear systems ($\alpha = 0$), the stochastic terms are uncorrelated white-noise sources (assuming a narrow bandwidth $\gamma \ll \omega_0$) satisfying:

$$\langle s_+^*(t)s_+(t') \rangle = k_B T_e \delta(t - t'), \quad (3)$$

$$\langle \xi^*(t)\xi(t') \rangle = k_B T_d \delta(t - t'), \quad (4)$$

$$D(\gamma_d) = \sqrt{2\gamma_d}, \quad (5)$$

where $\langle \dots \rangle$ is a thermodynamic ensemble average, and T_d and T_e are the local temperatures of the internal and external baths, respectively.

The presence of nonlinear dissipation $\sim \text{Im } \alpha |a|^2$ means that D must also depend on a and $\text{Im } \alpha$.³⁵ Note that $\text{Re } \alpha$ does not play any role in nonlinear dissipation. This intuitive result also follows from a microscopic Hamiltonian approach where $\text{Re } \alpha$ appears in the isolated system Hamiltonian as the quartic nonlinearity term while $\text{Im } \alpha$ represents system-heat bath nonlinear coupling²⁸. As a result, the diffusion coefficient D which captures the cavity–bath interaction in Eq. (1) does not depend on $\text{Re } \alpha$. (Interestingly, in the case of a driven quan-

tum oscillator, the real part of $\chi^{(3)}$ affects the tunneling rate between states and hence the fluctuation statistics²⁷.) Such a nonlinear FDT can be obtained from very general statistical considerations such as energy equipartition^{16,35,36}, derived under the assumption that the system is at equilibrium, i.e. $T = T_e = T_d$. As described in Appendix A, one can apply a standard procedure to transform the stochastic ODE [Eq. (1)] into a Fokker–Planck equation for the probability distribution $P(a, a^*)$,³⁷ which in our case is given by:

$$\frac{dP(a, a^*)}{dt} = -\frac{\partial}{\partial a} K_a P - \frac{\partial}{\partial a^*} K_{a^*} P + \frac{1}{2} \frac{\partial^2}{\partial a \partial a^*} K_{aa^*} P \quad (6)$$

with Fokker–Planck coefficients,

$$K_a = [i(\omega_0 - \text{Re } \alpha |a|^2) - (\gamma - \text{Im } \alpha |a|^2)]a + \lambda D \frac{\partial D}{\partial a^*}$$

$$K_{a^*} = [-i(\omega_0 - \text{Re } \alpha |a|^2) - (\gamma - \text{Im } \alpha |a|^2)]a^* + \lambda D \frac{\partial D}{\partial a}$$

$$K_{aa^*} = K_{a^*a} = (2\gamma_e + D^2), \quad K_{aa} = K_{a^*a^*} = 0$$

The precise meaning of these coefficients depends on the integration rule used to describe the stochastic ODE. Here, $\lambda = 0, \frac{1}{2}, 1$ correspond to Ito, Stratonovich and kinetic interpretations of stochastic calculus, respectively. The parameters λ and D must of course ensure that the statistical properties of the system are consistent with the laws of thermodynamics.

Based on the standard working hypothesis of statistical mechanics, the equilibrium state is described by the Maxwell-Boltzmann distribution $e^{-U/k_B T}$, where U is the energy in the cavity field. Since the nonlinearity is considered perturbatively in the derivation of the coupled mode equations,³⁸ nonlinear contributions to the cavity energy $|a|^2$ can be safely neglected.⁵⁰ More generally however, for an undriven oscillator, the nonlinear contribution to the oscillator energy can be ignored if the shift in the frequency is much smaller than the cavity eigenfrequency²⁸, which is the case for the passive, nonlinear cavity considered here. This well-known result can be verified by solving Eq. (6) in the absence of nonlinear dissipation ($\text{Im } \alpha = 0$), in which case the fluctuation statistics remain unchanged and one finds that terms $\sim \text{Re } \alpha$ do not affect the steady-state probability distribution^{35,39}. It follows that the equilibrium state of the system is described by the Gibbs distribution $e^{-|a|^2/k_B T}$ and the stochastic equations must be interpreted according to the kinetic rule $\lambda = 1$, provided that the diffusion coefficient D have the form:

$$D(a, a^*) = \sqrt{2(\gamma_d - \text{Im } \alpha |a|^2)} \quad (7)$$

Hence, the only modification to the diffusion coefficient is the addition of the nonlinear dissipation rate $\text{Im } \alpha |a|^2$. Note that the particular form or choice of multiplicative noise will determine the corresponding stochastic interpretation, and vice versa. For instance, it is also possible to choose the internal noise in Eq. (1) to be of the form $\sqrt{2\gamma_d}\xi_1 + D(a, a^*)\xi_2$, where $\xi_{1,2}$ are independent Gaussian noise sources, provided that $D = \sqrt{-2\text{Im } \alpha} a^*$ and that the stochastic ODE is inter-

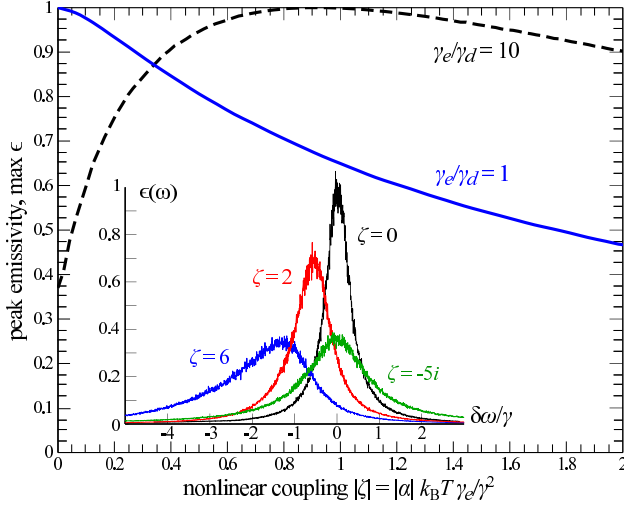


FIG. 1: Peak emissivity ϵ_{\max} of a cavity coupled to an external bath, both at temperature T , as a function of nonlinear coupling $|\zeta| = |\alpha|k_B T \gamma_e / \gamma^2$, for different ratios of the linear dissipation γ_e and external coupling γ_d rates. Inset shows the emissivity $\epsilon(\omega)$ for $\gamma_e = \gamma_d$, corresponding to a cavity with perfect linear emissivity, for multiple values of ζ , illustrating the effects of SPM (red/blue) and TPA (green) on the spectrum.

preted according to the Stratonovich rule $\lambda = \frac{1}{2}$.²⁸ Our choice of interpretation here is chosen purely for convenience.

III. THERMAL RADIATION

Equations (1) and (7) can be solved to obtain both the equilibrium and nonequilibrium behavior of the system. Since they do not admit closed-form analytical solutions, we instead solve the stochastic ODE numerically using the Euler–Maruyama method⁴⁰, involving a simple forward-difference discretization which for the kinetic calculus results in additional terms compared to an Ito discretization³⁷. To lowest order in the discretization,

$$\begin{aligned} \Delta a = & [i(\omega_0 - \alpha|a|^2) - \gamma]a\Delta t + D\Delta W_\xi \\ & + \frac{\partial D}{\partial a}\Delta a\Delta W_\xi + \frac{\partial D}{\partial a^*}\Delta a^*\Delta W_\xi + \sqrt{2\gamma_e}\Delta W_{s+}, \end{aligned}$$

where $\Delta a \equiv a(t + \Delta t) - a(t)$ and $\Delta W_f \equiv W_f(t + \Delta t) - W_f(t) = f\Delta t$ is a Wiener process⁴⁰ corresponding to the white-noise stochastic signal $f \in \{\xi, s_+\}$. It follows that to first order in Δt , the discretized ODE is given by:

$$\begin{aligned} \Delta a = & [i(\omega_0 - \alpha|a|^2) + \text{Im}\alpha|\xi|^2 - \gamma]a\Delta t \\ & + 2\sqrt{\gamma_d - \text{Im}\alpha|a|^2}\Delta W_\xi + \sqrt{2\gamma_e}\Delta W_{s+}, \end{aligned} \quad (8)$$

where the additional discretization term $\sim \text{Im}\alpha|\xi|^2$ arises in the kinetic and not the Ito calculus.

Equilibrium—In what follows, we demonstrate numerically that the system described by Eqs. (1) and (7) thermalizes and satisfies all of the properties of an equilibrium ther-

modynamic system, including equipartition and detailed balance, but that nonlinearities lead to strong modifications of the emissivity of the cavity. We consider the equilibrium situation $T \equiv T_d = T_e$, in which case $\langle |s_+|^2 \rangle = \langle |\xi|^2 \rangle = k_B T$. To begin with, we motivate our numerical results by performing a simple mean-field approximation known as statistical linearization⁴¹, which captures basic features but ignores correlation effects stemming from nonlinearities. Specifically, making the substitution $|a(t)|^2 \rightarrow \langle |a(t)|^2 \rangle = k_B T$ in Eq. (1), and solving for the steady-state, linear response of the system, we obtain the emissivity of the cavity $\epsilon(\omega) \equiv 2\gamma_e \langle |a(\omega)|^2 \rangle / k_B T$, defined as the emitted power into the external bath normalized by $k_B T$ in the limit $s_+ \rightarrow 0$. In particular, we find:

$$\epsilon(\omega) = \frac{4\gamma_e(\gamma_d - \text{Im}\alpha k_B T)}{\delta\omega_T^2 + (\gamma - \text{Im}\alpha k_B T)^2} \leq 1, \quad (9)$$

where $\delta\omega_T \equiv \omega - \omega_0 + \text{Re}\alpha k_B T$ and $\epsilon \leq 1$ as expected from Kirchoff's law^{3,5}.

Equation (9) can be integrated to verify the self-consistency condition $\langle |a(t)|^2 \rangle = \int \frac{d\omega}{2\pi} \langle |a(\omega)|^2 \rangle = k_B T$, as required by equipartition. It can also be combined with Eq. (2) to show that detailed balance $\langle |s_-(\omega)|^2 \rangle = \langle |s_+(\omega)|^2 \rangle$ is satisfied, i.e. there is no net transfer of heat from the cavity to the external bath and vice versa. More interestingly, the presence of α leads to a temperature-dependent change in the frequency and bandwidth of the cavity proportional to $\text{Re}\alpha$ and $\text{Im}\alpha$, respectively. These properties are validated by a full solution of the ODE, as illustrated on the inset of Fig. 1, which shows the numerically computed emissivity $\epsilon(\omega)$ for a few values of the dimensionless nonlinear coupling $\zeta \equiv \alpha k_B T \gamma_e / \gamma^2$. Although Eq. (9) yields good agreement with our numerical results for small $|\zeta| \lesssim 0.5$, at larger temperatures correlation effects become relevant and statistical linearization is no longer able to describe (even qualitatively) the spectral features. For instance, in the absence of TPA and for large ζ (such as $\zeta = 6$ in Fig. 1), SPM leads to asymmetrical broadening of the spectrum: broadening is most pronounced along the direction of the frequency shift, as determined by the sign of $\text{Re}\alpha$. This effect is known as “frequency straddling”, has been predicted in the context of Duffing mechanical oscillators,^{37,42,43} and arises due to frequency mixing within the cavity bandwidth, as captured by the perturbative expansion of the emissivity in powers of α in Eq. (11). In particular, at equilibrium one finds that the first-order correction to the emissivity $\sim -(\text{Re}\alpha k_B T)$, and so SPM enhances and reduces thermal contributions from red and blue-detuned frequencies, or vice versa depending on the sign of $\text{Re}\alpha$. Intuitively, the density of states within the cavity favors frequency conversion away from the resonance. Hence, photons on the red side of the resonance will experience larger frequency shifts than those on the blue side for $\text{Re}\alpha > 0$ (red shifting), and vice versa for $\text{Re}\alpha < 0$ (blue shifting). Note that equipartition $\langle |a|^2 \rangle = k_B T$ and detailed balance $\langle |s_+(\omega)|^2 \rangle = \langle |s_-(\omega)|^2 \rangle$ are satisfied even in the presence of strong correlations.

The above SPM and TPA effects pave the way for designing temperature-tunable thermal emissivities. For instance, it is well known that in a linear system, a cavity can become

a perfect emitter/absorber when the emission and dissipation rates are equal, i.e. $\gamma_e = \gamma_d$.⁴⁴ It follows from Eq. (9) that in the nonlinear case there is a modified rate-matching condition whereby $\epsilon = 1$ is achieved only at the critical temperature T_c where $\gamma_e = \gamma_d - \text{Im } \alpha(k_B T_c)$. Hence, a system designed to have $\gamma_e > \gamma_d$ (since $\text{Im } \alpha < 0$ in any passive system¹⁰) at room temperature will become a perfect emitter at $T_c \gtrsim 300\text{K}$. To illustrate this phenomenon, Fig. 1 shows the variation of the peak emissivity of the cavity, ϵ_{max} , by tuning the effective nonlinearity $|\zeta|$ for multiple values of γ_e/γ_d .

Nonequilibrium—We now consider nonequilibrium conditions and demonstrate that TPA can lead to thermal radiation exceeding the black-body limit. Assuming local equilibrium conditions, $\langle |s_+|^2 \rangle = k_B T_e$, $\langle |\xi|^2 \rangle = k_B T_d$, the heat transfer between the cavity and external bath is given by:

$$H = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} (\langle |s_-(\omega)|^2 \rangle - \langle |s_+(\omega)|^2 \rangle) \\ = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \Phi(\omega) k_B \Delta T \quad (10)$$

where $\Delta T \equiv T_d - T_e$ and $\Phi(\omega)$ is known as the spectral transfer function⁷, or the heat exchange between the two systems compared to two black bodies. (The transfer function of a black body $\Phi_{\text{BB}}(\omega) = 1$ at all frequencies.)

To begin with, we consider a perturbative expansion of Eq. (1) in powers of α , described in Appendix B, which we find to be accurate to within a few percent up to $|\zeta| \approx 0.5$. In this case we find that statistical linearization does not even qualitatively describe the behavior of the system at small α . To linear order in α , perturbation theory leads to the following expressions for the energy and output-power spectra:

$$\langle |a(\omega)|^2 \rangle = \frac{2\gamma k_B T_{\text{eff}}}{\delta\omega^2 + \gamma^2} - \frac{4\delta\omega\gamma \text{Re } \alpha(k_B T_{\text{eff}})^2}{(\delta\omega^2 + \gamma^2)^2} + \frac{2 \text{Im } \alpha(k_B^2 T_{\text{eff}})}{(\delta\omega^2 + \gamma^2)} \left[T_d + \frac{2\gamma^2(T_d - 2T_{\text{eff}})}{\delta\omega^2 + \gamma^2} \right] \quad (11) \\ \Phi(\omega) = \frac{4\gamma_e\gamma_d}{\delta\omega^2 + \gamma^2} - \frac{8\delta\omega\gamma_e\gamma_d \text{Re } \alpha(k_B T_{\text{eff}})}{(\delta\omega^2 + \gamma^2)^2} - \frac{1}{\Delta T} \frac{4\gamma_e \text{Im } \alpha k_B}{(\delta\omega^2 + \gamma^2)} \left[T_{\text{eff}} T_d + \frac{[2\gamma^2 T_{\text{eff}} + (\delta\omega^2 - \gamma^2) T_e](T_d - 2T_{\text{eff}})}{\delta\omega^2 + \gamma^2} \right], \quad (12)$$

where $\delta\omega \equiv \omega - \omega_0$ and $T_{\text{eff}} = \frac{\gamma_e T_e + \gamma_d T_d}{\gamma}$ is the effective temperature $\langle |a(t)|^2 \rangle / k_B$ of the cavity in the linear regime. At finite α , the effective temperature is given by:

$$T_{\text{eff}}^{\text{NL}} = T_{\text{eff}} - \frac{2 \text{Im } \alpha k_B T_{\text{eff}}}{\gamma} (T_d - T_{\text{eff}}), \quad (13)$$

which reduces to T_{eff} in the absence of nonlinearities and at equilibrium. Furthermore, one can also show that $\Phi \leq \Phi_{\text{BB}}$ and reaches its maximum at the resonance frequency when $\gamma_e = \gamma_d$. For finite $\text{Im } \alpha \neq 0$, we find that $T_{\text{eff}}^{\text{NL}} > T_{\text{eff}}$ irrespective of system parameters and that under certain conditions Φ increases above one. Thus, one arrives at the result that out of equilibrium, the rate at which energy is drawn from the phonon bath can be larger than the rate at which energy radiates from the cavity, causing the effective temperature and overall heat transfer to increase above its linear value, a phenomenon associated with the presence of excess heat.⁴⁵ Note that $T_{\text{eff}}^{\text{NL}}$ is not affected by $\text{Re } \alpha$ to first order since the perturbation is odd in $\delta\omega$ and therefore integrates to zero. One can show that the peak transfer can increase above one whenever $\frac{d\Phi(\omega_0)}{d(-\text{Im } \alpha)} [T_{\text{eff}}(3T_d + 2T_e - 4T_{\text{eff}}) - T_e T_d] > 0$ is satisfied, which occurs for instance when $T_d \gg T_e$, in which case $\frac{d\Phi(\omega_0)}{d(-\text{Im } \alpha)} > 0$. Thus, when a linear system has nearly perfect emissivity, any small amount of TPA can push its radiation

above the black-body limit. For example, the peak emissivity of a system with $\gamma_e = \gamma_d$, $T_e = 0$, and subject to TPA, is given from Eq. (13) by $\eta \equiv \frac{\Phi(\omega_0)}{\Phi_{\text{BB}}} = 1 - \frac{\text{Im } \alpha(k_B T_d)}{2\gamma}$, which increases above one with increasing $-\text{Im } \alpha$.

Figure 2 shows the peak spectral transfer $\eta_{\text{max}} = \Phi_{\text{max}}/\Phi_{\text{BB}}$, along with the normalized, frequency-integrated heat transfer $H(\zeta)/H_{\text{max}}(0)$ as a function of $|\zeta|$, computed by integrating Eq. (1) numerically. Here $H_{\text{max}}(0)$ denotes the maximum possible heat transfer in the linear regime which occurs under the rate matching condition $\gamma_e = \gamma_d$. (The inset shows a realistic structure where such nonlinear radiation effects can potentially be observed.) The largest increase in η occurs when ΔT is largest and so in the figure we consider the case $T_e = 0$, for multiple values of $\text{Re } \alpha / \text{Im } \alpha$ and γ_e/γ_d . As $|\zeta|$ increases from zero, η_{max} increases and in certain regimes becomes greater than one. At larger ζ , the enhancement is spoiled due to thermal broadening causing energy in the cavity to leak out at a faster rate, thereby weakening nonlinearities and causing $\eta_{\text{max}} \rightarrow 0$ as $|\zeta| \rightarrow \infty$. The maximum η is determined by a competition between these two effects, with thermal broadening becoming less detrimental and leading to larger enhancements with decreasing γ_e/γ_d . We find that TPA does not just enhance $\Phi(\omega)$ but also increases the total heat transfer and in particular $\frac{H(\zeta)}{H_{\text{max}}(0)} = \frac{2T_{\text{eff}}^{\text{NL}}}{\Delta T} \rightarrow \frac{2T_d}{\Delta T} = 2$ in

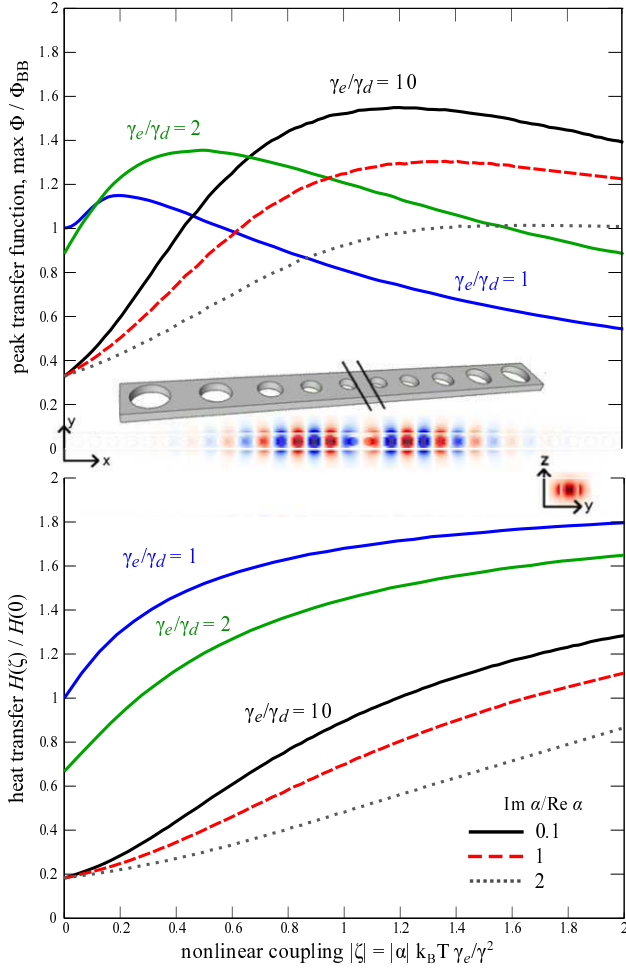


FIG. 2: Peak (on-resonance) spectral transfer function $\Phi_{\text{max}} \equiv \Phi(\omega_0)$ normalized by the black body Φ_{BB} (top), and net heat transfer $H(\zeta)$ normalized by $H_{\text{max}}(0)$ (bottom), as a function of nonlinear coupling $|\zeta| = |\alpha| k_B T \gamma_e / \gamma^2$, for a system consisting of a cavity at temperature T_d coupled to an external bath at $T_e = 0$, for multiple configurations of γ_e/γ_d and $\text{Re } \alpha / \text{Im } \alpha$. Inset shows a cavity design supporting a mode at $\lambda \approx 2.09 \mu\text{m}$ with lifetime $Q \approx 10^8$ and modal volume $V \approx 0.8(\lambda/n)^3$, along with its corresponding H_z and E_y field profiles.

the limit as $|\zeta| \rightarrow \infty$ (not shown), increasing monotonically with increasing ζ . As expected, H is bounded by the largest achievable effective temperature $T_{\text{eff}}^{\text{NL}} \leq T_d$, or alternatively by the maximum rate at which energy can be drawn from the phonon bath. Examination of the reverse scenario ($T_e > T_d$), in which the external bath is held at a higher temperature than the cavity, also leads to similar enhancements. However, because only the internal bath experiences nonlinear dissipation, the system exhibits non-reciprocal behavior with respect to $T_d \rightleftharpoons T_e$, which is evident in Eq. (12). Moreover, we find that $T_{\text{eff}}^{\text{NL}}$ in this reverse scenario always decreases with increasing TPA. Such non-reciprocity in the heat exchange is absent in linear systems and could potentially be useful in technological applications, such as for thermal rectification^{46,47}.

To illustrate the range of thermal tunability offered by TPA,

we consider the on-resonance heat transfer $\Phi_{\text{max}} \equiv \Phi(\omega_0)$ in the highly non-equilibrium regime $T_e = 0$ and for $\text{Re } \alpha = 0$. While SPM offers some degree of tunability, we find that TPA has a significantly larger impact on the radiation rate of the cavity. In this regime, Eq. (12) simplifies and yields $\Phi_{\text{max}} = \frac{4\gamma_e\gamma_d}{\gamma^2} \left[1 + \frac{(-\text{Im } \alpha)}{\gamma} (3 - 4\frac{\gamma_d}{\gamma}) k_B T_d \right]$, from which it follows that at small $\zeta \lesssim 0.5$, where Eq. (12) is applicable, Φ_{max} scales linearly with T_d and depends on the ratio $\frac{\gamma_d}{\gamma}$, increasing above its linear value whenever $3\gamma_e \geq \gamma_d$. Furthermore, one finds that the largest temperature variation, related to the slope $\frac{(-\text{Im } \alpha)}{\gamma} (3 - 4\frac{\gamma_d}{\gamma})$, is obtained in the limit $\gamma_e \gg \gamma_d$ and $\gamma_e, \gamma_d \rightarrow 0$, corresponding to a cavity with negligible linear emissivity, large $\zeta \gg 1$ (for a fixed temperature and $\text{Im } \alpha$), and narrow bandwidth. Interestingly, we find that even for a finite $\zeta \sim 0.4$ and $\gamma_e/\gamma_d \sim 20$, the emissivity of the cavity can increase dramatically from $\Phi_{\text{max}} \approx 0.2$ at $T_d = 300\text{K}$ to $\Phi_{\text{max}} \approx 0.9$ at $T_d = 600\text{K}$.

As mentioned above, it turns out that a nonlinear mechanical oscillator interacting with a medium through nonlinear friction exhibits similar spectral characteristics.^{11,28} However, in contrast to our photonic radiator, enhancements in the spectral peak due to nonlinear friction are only observed when the nonlinear dissipation rate is much larger than the linear loss rate, or equivalently when the source of nonlinear friction is at a very high temperature compared to the internal phonon temperature.¹¹ While realizing these experimental conditions in mechanical oscillators, including the need to have isolated linear and nonlinear heat baths operating at vastly different temperatures, seems difficult, a photonic cavity offers alternative ways of observing thermal radiation above the linear blackbody limit, creating new opportunities for studying nonlinear damping. First, while in the case of a mechanical oscillator one observes large enhancements only when the internal dissipation and hence the bandwidth $\gamma_d \rightarrow 0$, the introduction of an external radiative channel in a photonic system enables large thermal enhancement with finite γ_d and hence larger bandwidths. In particular, as long as the linear cavity losses are dominated by radiation to the external bath, corresponding to the situation $\gamma_d \ll \gamma_e$, the total cavity bandwidth γ_e can be large while still allowing internal losses to be dominated by nonlinear friction. Second, while in the case of mechanical oscillators one observes nonlinear enhancement only when the external (linear dissipation) temperature is small compared to the internal (nonlinear dissipation) temperature, $T_e \ll T_d$, interference effects associated with the presence of the external bath in the photonic system ameliorate this experimentally onerous constraint. In particular, the heat exchanged between the photonic cavity and external bath depends on the sensitive interference between reflected and emitted radiation from the cavity, described by Eq. (2). These interference effects result in amplitude correlations $\sim \text{Im } \alpha \langle s_+^* a a^* \xi \rangle$, corresponding to the last term of Eq. (12), whose contribution to the heat transfer cannot be ignored in situations where $T_e \lesssim T_d$. (In their absence, the spectrum of the cavity resembles that of a mechanical oscillator and one can no longer observe significant enhancements in thermal radiation unless $T_e \ll T_d$.) For illustration, consider a situation in which the cavity and external

channel are held at temperatures $T_d = 600\text{K}$ and $T_e = 300\text{K}$, respectively. In this case, we find that the maximum transfer increases from $\Phi_{\max} \approx 0.6$ at $\zeta = 0$ to $\Phi_{\max} \approx 1.2$ at $\zeta = 1$ almost entirely due to interference between the reflected and emitted radiation from the cavity, in the absence of which Φ_{\max} actually decreases with increasing ζ .

IV. CONCLUSION

We conclude by proposing a realistic system in which one could potentially observe these effects. In order to reach the strongly nonlinear regime, it is desirable to have $|\zeta| = \frac{|\alpha|k_B T \gamma_e}{\gamma^2} \sim 1$. Given a choice of operating temperature, the goal is therefore to design a cavity with a large Purcell factor $\alpha/\gamma \sim Q/V$. If the goal is to observe large enhancements from TPA, it is also desirable to operate with materials and wavelengths where the nonlinear FOM $\sim \frac{n_2}{\lambda \beta_{\text{TPA}}} \lesssim 1$, corresponding to large TPA.¹⁰ All of these conditions can be achieved in a number of material systems and geometries. For illustration, we consider the Ge nanobeam cavity shown on the inset of Fig. 2 and based on the family of nanobeam cavities described in Ref. 31, which supports a mode at $\lambda = 2.09\mu\text{m}$. At this wavelength, Ge has an index of refraction $n \approx 4$ and Kerr coefficient $\chi^{(3)} \approx (1.2 - 11i) \times 10^{-17}(\text{m/V})^{210,48}$, corresponding to a FOM ≈ 0.008 . This yields a mode with $\alpha \approx 0.001(\chi^{(3)}/\varepsilon_0 \lambda^3)$, $Q \approx 10^8$, and modal volume $V \approx 0.8(\lambda/n)^3$, leading to $|\zeta| \approx 1$ for operation at $T = 1000\text{K}$. (Large Purcell factors such as these were recently predicted in a similar, albeit silicon platform³¹.) We note that there are other possible cavity designs, wavelength and material choices, including GaP and ZnSe, and that it is also possible to operate with larger bandwidths at the expense of larger temperatures and/or smaller mode volumes. Because these thermal effects scale linearly with Purcell factor, we believe that nanophotonic cavities with ultra-small modal volumes and bandwidths are the most promising candidates for experimental realization. This is in contrast to the situation encountered in traditional nonlinear devices involving incident (non-thermal) light, where the threshold power for observing strong nonlinear effects $\sim V/Q^2$ and therefore favors designs that sacrifice modal volume in favor of smaller bandwidths.¹²

Finally, we note that the predictions above offer only a glimpse of the potentially interesting radiative phenomena that can arise in passive nonlinear media at and out of equilibrium. In future work, it may be interesting to consider the impact of other nonlinear phenomena on thermal radiation, including free-carrier absorption and third harmonic generation, as well as applications of the Kerr effect to thermal rectification^{46,47}. In a related context of optomechanics, the coupling of photonic and mechanical resonances leads to novel nonlinear effects that are also manifested in the radiation spectrum of photonic cavities, often studied in the presence of incident, non-thermal radiation pressure.^{18,49} We believe that electronic nonlinearities such as the Kerr effect in semiconductors offer an alternative approach to exploring similar ideas related to nonlinear fluctuations.

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Appendix A: Fokker-Planck Equation

In this appendix, we review the procedure for deriving the FP equation [Eq. (6)] from the corresponding nonlinear Langevin equation [Eq. (2)], which can be written in the following simplified form:

$$\begin{aligned} \dot{a} &= f(a, a^*) + D(a, a^*)\xi + \sqrt{2\gamma_e} s_+ \\ f(a, a^*) &= i(\omega_0 - \alpha|a|^2)a - \gamma_a, \end{aligned}$$

where D is the diffusion coefficient, and ξ and s_+ are delta-correlated white-noise sources obtained by taking the derivative of standard Wiener processes⁴⁰, $\xi = \dot{W}_\xi$ and $s_+ = \dot{W}_s$. For a finite discretization time Δt , the coupled-mode equations can be written as follows³⁷:

$$\begin{aligned} \frac{a(t) - a(t - \Delta t)}{\Delta t} &= f(\lambda a(t) + (1 - \lambda)a(t - \Delta t)) \\ &+ D(\lambda a(t) + (1 - \lambda)a(t - \Delta t)) \frac{(W_1(t) - W_1(t - \Delta t))}{\Delta t} \\ &+ \sqrt{2\gamma_e} \frac{(W_1(t) - W_1(t - \Delta t))}{\Delta t} \quad (\text{A1}) \end{aligned}$$

where the choice of $0 \leq \lambda \leq 1$ determines the corresponding Stochastic interpretation rule. Taylor expanding each term and defining $\Delta a \equiv a(t) - a(t - \Delta t)$ and $\Delta W_i = W_i(t) - W_i(t - \Delta t)$, with ΔW_i denoting a standard Brownian increment with zero mean and variance $\langle \Delta W_i^* \Delta W_i \rangle = k_B T \Delta t$, one finds the following expression to $O(\Delta t)$,

$$\begin{aligned} \Delta a &= f(a, a^*)\Delta t + D\Delta W_1 + \lambda D \frac{\partial D}{\partial a} \Delta W_1 \Delta W_1 + \\ &\lambda D \frac{\partial D}{\partial a^*} \Delta W_1^* \Delta W_1 + \sqrt{2\gamma_e} \Delta W_2 \quad (\text{A2}) \end{aligned}$$

Transforming the Langevin equation into a FP PDE involves a standard procedure³⁷ and leads to an equation of the form $\frac{\partial P}{\partial t} = -\sum_\alpha \frac{\partial}{\partial a_\alpha} K_\alpha P + \sum_{\alpha, \beta} \frac{\partial^2}{\partial a_\alpha \partial a_\beta} K_{\alpha, \beta} P$, where the FP coefficients are given by:

$$\begin{aligned} K_\alpha &= \lim_{\Delta t \rightarrow 0} \frac{\langle a_\alpha(t) - a_\alpha(t - \Delta t) \rangle}{\Delta t} \\ K_{\alpha, \beta} &= \lim_{\Delta t \rightarrow 0} \frac{\langle (a_\alpha(t) - a_\alpha(t - \Delta t))(a_\beta(t) - a_\beta(t - \Delta t)) \rangle}{\Delta t} \end{aligned}$$

Carrying out the above limiting procedures, one obtains the FP equation given in Eq. (6).

Appendix B: Perturbation Theory

In this appendix, we derive perturbative expressions for the energy spectrum $\langle |a(\omega)|^2 \rangle$ and transfer function $\Phi(\omega)$ of the nonlinear cavity. For convenience, we define $\alpha = \alpha_1 - \alpha_2 i$, with $\alpha_2 = -\text{Im } \alpha > 0$ as required by any passive nonlinear system. We begin by defining a perturbed cavity field $a(t) = a_0(t) + \delta a(t)$, where a_0 is the linear cavity field and δa is a correction of linear order in α . Plugging in the perturbed field into the coupled-mode equations and ignoring terms $O(\alpha^2)$ and higher, one obtains the coupled equations:

$$\dot{a}_0 = (i\omega_0 - \gamma)a_0 + \sqrt{2\gamma_d}\xi + \sqrt{2\gamma_e}s_+ \quad (\text{B1})$$

$$\dot{\delta a} = (i\omega_0 - \gamma)\delta a - (i\alpha + \alpha_2)|a_0|^2 a_0 + \frac{\alpha_2}{\sqrt{2\gamma_d}}|a_0|^2 \xi \quad (\text{B2})$$

Fourier transforming both equations, their solution to first order in α can be written as:

$$a_0(\omega) = D(\omega)^{-1} \left[\sqrt{2\gamma_d}\xi(\omega) + \sqrt{2\gamma_e}s_+(\omega) \right] \quad (\text{B3})$$

$$\delta a(\omega) = \frac{D(\omega)^{-1}}{\sqrt{2\gamma_d}} \mathcal{F} \left\{ \alpha_2 |a_0|^2 \xi - (i\alpha_1 + \alpha_2) \sqrt{2\gamma_d} |a_0|^2 a_0 \right\} \quad (\text{B4})$$

where $D(\omega) \equiv i(\omega - \omega_0) + \gamma$ and $\mathcal{F} \equiv \int dt e^{-i\omega t}$ denotes the Fourier transform operator.

1. Energy spectrum

We first compute the energy spectrum of the perturbed cavity which, to first order in δa , is given by:

$$\langle |a(\omega)|^2 \rangle = \langle |a_0(\omega)|^2 \rangle + 2 \text{Re} \{ \langle a_0^*(\omega) \delta a(\omega) \rangle \}, \quad (\text{B5})$$

As discussed below, the second term can be obtained by exploiting the following linear, two-point correlation functions:

$$\langle a_0^*(\omega) \xi(\omega') \rangle = \frac{\sqrt{2\gamma_d} k_B T_d \delta(\omega - \omega')}{D^*(\omega)} \quad (\text{B6})$$

$$\langle a_0(\omega) \xi(\omega') \rangle = \frac{\sqrt{2\gamma_d} k_B T_d \delta(\omega + \omega')}{D(\omega)} \quad (\text{B7})$$

$$\langle a_0^*(\omega) s_+(\omega') \rangle = \frac{\sqrt{2\gamma_e} k_B T_e \delta(\omega - \omega')}{D^*(\omega)} \quad (\text{B8})$$

$$\langle a_0(\omega) s_+(\omega') \rangle = \frac{\sqrt{2\gamma_e} k_B T_e \delta(\omega + \omega')}{D(\omega)} \quad (\text{B9})$$

$$\langle a_0^*(\omega) a_0(\omega') \rangle = \frac{\gamma k_B T_{\text{eff}} \delta(\omega - \omega')}{D(\omega) D^*(\omega)}, \quad (\text{B10})$$

where $T_{\text{eff}} = \frac{\gamma_e T_e + \gamma_d T_d}{\gamma}$ denotes the linear effective temperature of the cavity. In deriving Eqs. (B6)–(B10), we employed the fact that s_+ and ξ are uncorrelated white-noise sources described by Eqs. (3) and (5). Equation (B10) is precisely the zeroth-order term of the energy spectrum (in the absence of

nonlinearities), while the first-order correction is given by the more complicated expression:

$$\langle a_0^*(\omega) \delta a(\omega') \rangle = \left\langle \frac{[\sqrt{2\gamma_d}\xi^*(\omega) + \sqrt{2\gamma_e}s_+^*(\omega)] \times \frac{1}{\sqrt{2\gamma_d}}G(\omega')}{D^*(\omega)D(\omega')} \right\rangle \quad (\text{B11})$$

where the function

$$G(\omega) \equiv \mathcal{F} \left[\alpha_2 |a_0|^2 (\xi - \sqrt{2\gamma_d} a_0) - i\alpha_1 \sqrt{2\gamma_d} |a_0|^2 a_0 \right] \\ = \int_{-\infty}^{\infty} dx \int_{-\infty}^{\infty} dt e^{-i(\omega-x)t} \int_{-\infty}^{\infty} d\omega_1 e^{-i\omega_1 t} \int_{-\infty}^{\infty} d\omega_2 e^{i\omega_2 t} \\ a_0^*(\omega_1) a_0(\omega_2) \left[\alpha_2 (\xi(x) - \sqrt{2\gamma_d} a_0(x)) - i\alpha_1 \sqrt{2\gamma_d} a_0(x) \right],$$

encapsulates the spectral response of the perturbed cavity field, here simplified by exploiting the relation $\mathcal{F}\{|a_0|^2\} = \mathcal{F}\{a_0\} \star \mathcal{F}\{a_0^*\}$. Focusing first on the α_2 terms of the numerator of Eq. (B11), one obtains:

$$\alpha_2 \int_{-\infty}^{\infty} dx \int_{-\infty}^{\infty} dt \int_{-\infty}^{\infty} d\omega_1 \int_{-\infty}^{\infty} d\omega_2 e^{-i(\omega' + \omega_1 - \omega_2 - x)t} \\ a_0^*(\omega_1) a_0(\omega_2) [\xi(x) - \sqrt{2\gamma_d} a_0(x)] [\xi^*(\omega) + \sqrt{\frac{\gamma_e}{\gamma_d}} s_+^*(\omega)],$$

The ensemble average of the expression under the integrals involves four-point correlation functions and is given by:

$$\langle \dots \rangle = \langle a_0^*(\omega_1) a_0(\omega_2) \xi(x) \xi^*(\omega) \rangle \\ + \sqrt{\frac{\gamma_e}{\gamma_d}} \langle a_0^*(\omega_1) a_0(\omega_2) \xi(x) s_+^*(\omega) \rangle \\ - \sqrt{2\gamma_d} \langle a_0^*(\omega_1) a_0(\omega_2) a_0(x) \xi^*(\omega) \rangle \\ - \sqrt{2\gamma_e} \langle a_0^*(\omega_1) a_0(\omega_2) a_0(x) s_+^*(\omega) \rangle$$

Because the noise sources follow Gaussian distributions, four-point correlation functions can be written in terms of products of two-point correlation functions via Wick's theorem.³⁵ Summing the resulting two-point correlation functions, described by Eqs. (B6)–(B10), one obtains the following three terms:

$$T_{\text{eff}} \left[T_d - \frac{2\gamma_d T_d + 2\gamma_e T_e}{D(\omega)} \right] \frac{\delta(\omega - \omega')}{D^*(\omega) D(\omega')}, \\ \frac{2\gamma_d T_d + 2\gamma_e T_e}{D(\omega)} [T_d - T_{\text{eff}}] \frac{\delta(\omega - \omega')}{D^*(\omega) D(\omega')}, \\ \frac{2\gamma_d T_d + 2\gamma_e T_e}{D^*(-\omega)} \left[T_d - T_{\text{eff}} \frac{\gamma^2 + i\omega_0 \gamma}{\gamma^2 + \omega_0^2} \right] \frac{\delta(\omega - \omega')}{D^*(\omega) D(\omega')}.$$

It follows that the α_2 term in Eq. (B11) is given by:

$$\frac{\alpha_2 k_B^2}{|D(\omega)|^2} \left[T_{\text{eff}} T_d - \frac{2\gamma T_{\text{eff}}^2}{D(\omega)} + \frac{2\gamma T_{\text{eff}} T_d}{D(\omega)} - \frac{2\gamma T_{\text{eff}}^2}{D(\omega)} \right. \\ \left. + \frac{\gamma T_{\text{eff}} T_d}{D^*(-\omega)} - \frac{2\gamma T_{\text{eff}}^2}{D(\omega)} \frac{\gamma^2 + i\omega_0 \gamma}{\gamma^2 + \omega_0^2} \right] \quad (\text{B12})$$

Note that the last two terms can be neglected since the quantities $\propto D^*(-\omega)$ involve off-resonant, counter-rotating fields and furthermore, our coupled-mode theory is only valid in the regime $\gamma \ll \omega_0$. Performing a similar calculation for the α_1 term yields:

$$\frac{-\alpha_1 k_B^2}{|D(\omega)|^2} \left[\frac{2\gamma T_{\text{eff}}^2}{D(\omega)} + \frac{2\gamma T_{\text{eff}}^2}{D(\omega)} + \frac{2\gamma T_{\text{eff}}^2}{D^*(-\omega)} \frac{\gamma^2 + i\omega_0\gamma}{\gamma^2 + \omega_0^2} \right] \quad (\text{B13})$$

Putting together the above two expressions for both real and imaginary α and neglecting counter-rotating terms, one obtains the energy spectrum in Eq. (11).

2. Spectral transfer function

The spectral transfer function is defined as the relative power transfer from the cavity into the output channel divided by their temperature difference,

$$\Phi(\omega) = \frac{\langle |s_-(\omega)|^2 \rangle - \langle |s_+(\omega)|^2 \rangle}{k_B \Delta T}.$$

To first order in α , the outgoing power is given by:

$$\langle |s_-(\omega)|^2 \rangle = \langle |s_+(\omega)|^2 \rangle + 2\gamma_e S_{aa}(\omega) - 2\sqrt{2\gamma_e} \text{Re}\{\langle s_+^*(\omega) [a_0(\omega) + \delta a(\omega)] \rangle\}, \quad (\text{B14})$$

where the first and second terms are the incident power and energy spectra of the cavity, obtained above, and so it only remains to calculate the third term, or the interference between the incoming and outgoing radiation. Following the same procedure as before, the zeroth- and first-order correction terms

are given by $\langle s_+^*(\omega) a_0(\omega) \rangle = \frac{\sqrt{2\gamma_e} T_d}{D^*(\omega)}$ and

$$\begin{aligned} \langle s_+^*(\omega) \delta a(\omega) \rangle &= \frac{1}{\sqrt{2\gamma_d}} \int_{-\infty}^{\infty} dx \int_{-\infty}^{\infty} dt \int_{-\infty}^{\infty} d\omega_1 \\ &\int_{-\infty}^{\infty} d\omega_2 e^{-i(\omega' + \omega_1 - \omega_2 - x)t} \left[\alpha_2 a^*(\omega_1) a(\omega_2) (\xi(x) \right. \\ &\left. - \sqrt{2\gamma_d} a(x) s_+^*(\omega) - i\alpha_1 \sqrt{2\gamma_d} a^*(\omega_1) a(\omega_2) a(x) s_+^*(\omega) \right], \end{aligned}$$

As before, these can be broken down into contributions from α_2 and α_1 , which yields:

$$\begin{aligned} \frac{\alpha_2 \sqrt{2\gamma_e} k_B^2}{|D(\omega)|^2} \left\{ -T_{\text{eff}} T_e + T_e (T_d - T_{\text{eff}}) \right. \\ \left. + \frac{T_e D^*(\omega)}{D^*(-\omega)} \left[T_d - T_{\text{eff}} \left(\frac{\gamma^2 + i\omega_0\gamma}{\gamma^2 + \omega_0^2} \right) \right] \right\}, \quad (\text{B15}) \end{aligned}$$

and

$$-\alpha_1 k_B^2 \left[\frac{2\sqrt{2\gamma_e} T_{\text{eff}} T_e}{D(\omega)^2} + \frac{\sqrt{2\gamma_e} T_{\text{eff}} T_e}{D(\omega) D^*(-\omega)} \right], \quad (\text{B16})$$

respectively. As before, the counter-rotating terms $\sim D^*(\omega)$ can be neglected, leading to the following expression:

$$\begin{aligned} \langle s_+^*(\omega) \delta a(\omega) \rangle &= -4\alpha_2 \gamma_e T_e [T_d - 2T_{\text{eff}}] \frac{\gamma^2 (\omega - \omega_0)^2}{|D(\omega)|^4} \\ &+ 8\alpha_1 \gamma_e T_e T_{\text{eff}} \frac{\gamma (\omega - \omega_0)}{|D(\omega)|^4}. \quad (\text{B17}) \end{aligned}$$

Finally, after collecting like terms one obtains the spectral transfer function in Eq. (12).

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