Stimulated Raman scattering of colored chaotic light

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The quantum theory of forward Stokes generation by means of stimulated Raman scattering is extended to the case of a colored (finite-bandwidth) chaotic pump, which has both amplitude and phase fluctuations. In both transient and steady-state limits the mean Stokes intensity is found to be enhanced over that resulting from a coherent pump. In the limit that the chaotic-pump bandwidth becomes large, the mean Stokes intensity becomes identical with that resulting from a coherent pump of the same total power.

The *amplification*, by stimulated Raman scattering (SRS), of a weak-input Stokes wave in the presence of a strong, incoherent pump wave has received much theoretical attention.[1–8] On the other hand, the *generation* of Stokes light, in the absence of an input Stokes wave, by SRS of an incoherent-pump wave has not been fully studied. The quantum theory of Stokes generation,[9,10] which treats spontaneous scattering and spatial propagation in a unified way, has so far been analyzed only for a coherent pump and for a pump with constant amplitude but fluctuating phase. It was pointed out in Ref. 9 that the average intensity of Stokes generation is predicted to be exactly independent of any pump bandwidth arising solely from phase fluctuations. Although this result gives insight into the nature of the quantum generation of Stokes light, it does not correspond to the usual experimental situation in which a multimode laser (such as Nd:YAG or dye) or an amplified-spontaneous-emission laser (such as nitrogen or excimer) is used as the pump source. The light from pump sources such as these exhibits both amplitude and phase fluctuations, and it is expected that the average Stokes-generation intensity will be quite different from that in the pure phase-fluctuation case.

A well-known model that can account for both amplitude and phase fluctuations in the pump is that of colored chaotic light. This is light with finite bandwidth (not white), whose complex field amplitude $E_L(\tau)$ is a Gaussian random process.[7,8,11] Light of this kind can be thought of as being composed of a large number of statistically independent modes with fluctuating phases and with frequency separations much smaller than any other relevant frequency.[11] Thus in some cases a multimode laser may fall into this category. Consider the case that the spectrum of the chaotic light is a Lorentzian with half-width Γ_L. This leads to a field autocorrelation function of the form

$$\langle E_L^*(\tau)E_L(\tau')\rangle = E_0^2 \exp(-\Gamma_L|\tau-\tau'|) \tag{1a}$$

and an intensity autocorrelation function of the form

$$\langle I_L(\tau)I_L(\tau')\rangle = E_0^4[\exp(-2\Gamma_L|\tau-\tau'|)+1], \tag{1b}$$

where $\langle\ \rangle$ stands for a classical ensemble average over the field fluctuations. Thus the time scale of both amplitude and phase fluctuations is of the order of $1/\Gamma_L$. Note that in the pure phase-fluctuation case Eq. (1a) remains the same while the right-hand side of Eq. (1b) is replaced by a constant.

The purpose of this paper is to extend the quantum theory of forward Stokes generation to the case that the pump wave is described as colored chaotic light. Explicit predictions are made for the average Stokes-generation intensity, and a strong dependence is found on pump bandwidth, in contrast to the above-mentioned case of pure phase fluctuations. In particular, it is found that the highest Stokes intensities result from the smallest pump bandwidths and that in the limit of large pump bandwidths the Stokes intensity becomes identical with that resulting from a coherent pump of the same total power.

The quantum theory of Stokes generation[9,10] is based on the Heisenberg equations of motion for the slowly varying Stokes-field operator $\hat{E}_S^{(-)}(z,\tau)$ and the atomic-transition operator $\hat{Q}^\dagger(z,\tau)$:

$$\frac{\partial}{\partial z}\hat{E}_S^{(-)} = -i\kappa_2 E_L \hat{Q}^\dagger, \tag{2a}$$

$$\frac{\partial}{\partial \tau}\hat{Q}^\dagger = -\Gamma\hat{Q}^\dagger + i\kappa_1 E_L^* \hat{E}_S^{(-)} + \hat{F}^\dagger, \tag{2b}$$

where $\tau = t - z/c$ is the local time in the frame moving at speed c. The medium is assumed to be dispersionless. The Γ and $\hat{F}^\dagger$ terms describe collision-induced damping (dephasing) and fluctuations, respectively. The coefficients κ_1 and κ_2 are related to the mean steady-state gain coefficient $\bar{g}$ by

$$\bar{g} = 2\kappa_1\kappa_2 E_0^2/\Gamma. \tag{3}$$

Equations (2) are valid when the pump-laser wave and the atomic ground states remain undepleted. The approximate dependence of the fields on only one spatial dimension arises because the interaction volume is taken to be a cylinder with cross-sectional area $\mathcal{A}$ and length L, giving a Fresnel number $\mathcal{F} = \mathcal{A}/\lambda_S L$ near unity.

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Equations (2) have known explicit solutions, which are used to find the intensity (in photons/second) of Stokes radiation scattered into the forward solid angle ($\mathcal{A}/L^2$) defined by the interaction region

$$I(\tau) = \frac{\mathcal{A}}{2\pi\hbar\omega_S}\langle \hat{E}_S^{(-)}(z,\tau)\hat{E}_S^{(+)}(z,\tau)\rangle_{QM}, \quad (4)$$

where $\langle\ \rangle_{QM}$ is a quantum expectation value taken in the initial state having all atoms in their ground states and no Stokes photons present. In evaluating Eq. (4) the following expectation values are used:

$$\langle \hat{Q}^\dagger(z,O)\hat{Q}(z',O)\rangle = (\mathcal{A}N)^{-1}\delta(z-z'), \quad (5a)$$

$$\langle \hat{F}^\dagger(z,\tau)\hat{F}(z',\tau')\rangle = 2\Gamma(\mathcal{A}N)^{-1}\delta(z-z')\delta(\tau-\tau'), \quad (5b)$$

where N is the atomic-number density. Equation (5a) describes the initial quantum noise in the coordinate $\hat{Q}$ (i.e., Raman polarization) that is responsible for initiating the Stokes generation. Equation (5b) describes the strength of the collisional fluctuations. The result for the Stokes intensity is[9]

$$I(\tau) = \tfrac{1}{2}\Gamma\bar{g}z|A(\tau)|^2\{e^{-2\Gamma\tau}(I_0{}^2\{[2\Gamma\bar{g}zp(\tau)]^{1/2}\} - I_1{}^2\{2\Gamma\bar{g}zp(\tau)]^{1/2}\}) + 2\Gamma\int_0^\tau d\tau' e^{-2\Gamma\tau'} \times [I_0{}^2(\{2\Gamma\bar{g}z[p(\tau)-p(\tau-\tau')]\}^{1/2}) - I_1{}^2(\{2\Gamma\bar{g}z[p(\tau)-p(\tau-\tau')]\}^{1/2})]\}, \quad (6a)$$

where $A(\tau)$ is the normalized pump-laser field amplitude $A(\tau) = E_L(\tau)/E_0$, so that $\langle|A(\tau)|^2\rangle = 1$, and $p(\tau)$ is

$$p(\tau) = \int_0^\tau |A(\tau')|^2 d\tau'. \quad (6b)$$

$I_n(x)$ is the modified Bessel function of order n. This result shows that the Stokes intensity is independent of fluctuations in the phase of the pump field, as mentioned above. The fluctuations in the amplitude $|A(\tau)|$ of the pump field are accounted for by averaging Eq. (6a) over the corresponding classical ensemble to find the mean Stokes intensity $\langle I(\tau)\rangle$. We evaluate this average by an exact statistical method in two limiting physical cases: transient limit and steady-state limit.

TRANSIENT LIMIT

When the time τ is much less than the collisional dephasing time, i.e., $\Gamma\tau \ll 1$, the scattering is in the transient limit, and the average intensity is given by the first term in Eq. (6a):

$$\langle I_{TR}(\tau)\rangle = \tfrac{1}{2}\Gamma\bar{g}z\langle|A(\tau)|^2(I_0{}^2\{[2\Gamma\bar{g}zp(\tau)]^{1/2}\} - I_1{}^2\{[2\Gamma\bar{g}zp(\tau)]^{1/2}\})\rangle. \quad (7)$$

Note that $\Gamma\bar{g}$ is independent of Γ. The average in this equation is carried out exactly by first replacing the modified Bessel functions by their Laplace transform representations, leaving the pump amplitude appearing only through the form $\langle\exp[\gamma p(\tau)]\rangle$, where γ is a constant. This average can be explicitly evaluated for a chaotic field,[12] leaving an inverse Laplace transform to be carried out. This averaging procedure is detailed in Appendix A.

In the limit of infinite bandwidth $\Gamma_L \gg \Gamma$, the averaging procedure can be carried out analytically, leading to

$$\langle I_{TR}(\tau)\rangle = \tfrac{1}{2}\Gamma\bar{g}z\{I_0{}^2[(2\Gamma\tau\bar{g}z)^{1/2}] - I_1{}^2[(2\Gamma\tau\bar{g}z)^{1/2}]\}. \quad (8)$$

This can be easily seen from Eq. (7) to be identical with the case of a coherent laser, in which the amplitude is nonfluctuating $[|A(\tau)| = 1, p(\tau) = \tau]$. In the infinite-bandwidth case the quantity $|A(\tau)|^2$ is fluctuating so fast that it can be effectively replaced by its mean value, unity.

In the monochromatic limit $\Gamma_L \ll \Gamma$, this procedure can also be carried out analytically, leading to

$$\langle I_{TR}(\tau)\rangle = \tfrac{1}{2}\Gamma\bar{g}zI_0(\Gamma\tau\bar{g}z)\exp(\Gamma\tau\bar{g}z). \quad (9)$$

In the general case of arbitrary pump-laser bandwidth Γ_L, the averaging procedure leads to

$$\langle I_{TR}(\tau)\rangle = \tfrac{1}{2}\Gamma\bar{g}z\int_C ds s^{-1}f(s)\exp(s\Gamma\tau\bar{g}z) \times [I_0(s\Gamma\tau\bar{g}z) - I_1(s\Gamma\tau\bar{g}z)], \quad (10a)$$

where, defining $\delta = (1 - 2/s\Gamma_L\tau)^{1/2}$, $f(s)$ is given by $f(s) =$

$$\frac{8\delta\{(\delta+1)\exp[\Gamma_L\tau(\delta-1)] + (\delta-1)\exp[-\Gamma_L\tau(\delta+1)]\}}{\{(\delta+1)^2\exp[\Gamma_L\tau(\delta-1)] - (\delta-1)^2\exp[-\Gamma_L\tau(\delta+1)]\}^2}. \quad (10b)$$

This has the form of an inverse Laplace transform, in which the contour integration path is parallel to the imaginary axis and is to the right of the singular points of the integrand, which form a series coming from the left side to zero. We carried out the integral in Eq. (10a) numerically by using the method of Krylov and Skoblya,[13] which is necessary because of the rapidly oscillating exponential factor. In this method the integrand is approximated with polynomials (order 29), for which the integral can be done explicitly.

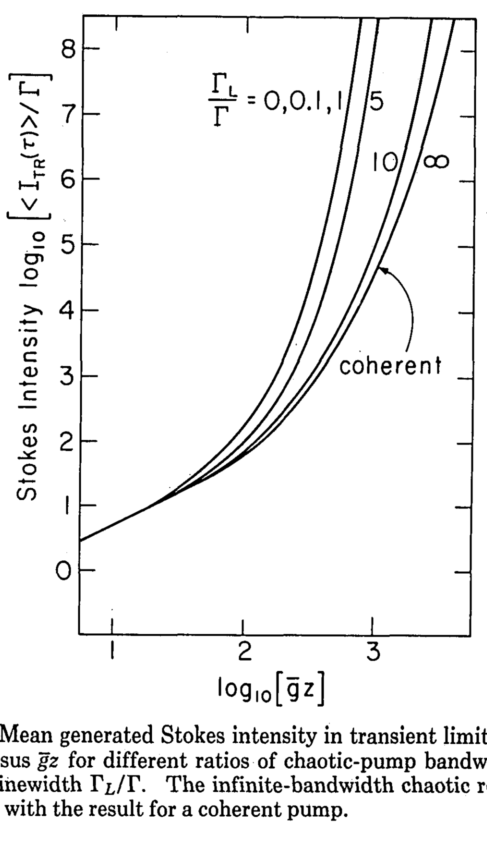

Fig. 1. Mean generated Stokes intensity in transient limit ($\Gamma\tau$ = 0.01) versus $\bar{g}z$ for different ratios of chaotic-pump bandwidth to Raman linewidth Γ_L/Γ. The infinite-bandwidth chaotic result is identical with the result for a coherent pump.

In Fig. 1 the mean Stokes intensity $\langle I_{TR}(\tau)\rangle$, from Eqs. (8)–(10), is plotted versus $\bar{g}z$. We chose $\Gamma\tau = 0.01$ to guarantee that the scattering be in the transient limit. (This means that the abscissa of the graph may also be interpretted as $200 \times \kappa_1\kappa_2 E_0{}^2\tau$.) The four curves are labeled by different ratios of pump-laser bandwidth to Raman linewidth Γ_L/Γ. The transition from linear, spontaneous scattering at small $\bar{g}z$ to nonlinear, stimulated scattering at higher $\bar{g}z$ is seen to depend on the pump bandwidth. The curve labeled 0, 0.1, 1 is obtained either from Eq. (9) ($\Gamma_L = 0$) or from Eqs. (10) ($\Gamma_L/\Gamma = 0.1, 1$). The curves labeled 5 and 10 are from Eqs. (10), whereas the curve labeled ∞ and coherent is from Eq. (8), the infinite-bandwidth limit, which is identical with the coherent-laser result.

STEADY-STATE LIMIT

When the time τ is much larger than the collisional dephasing time, $\Gamma\tau \gg 1$, the scattering is in the steady-state limit, and the average intensity is given by the second term in Eq. (6a):

$$\langle I_{SS}\rangle = 2\Gamma\left(\tfrac{1}{2}\Gamma\bar{g}z|A(\tau)|^2 \int_0^\tau d\tau' e^{-2\Gamma\tau'} \times \left[I_0{}^2(\{2\Gamma\bar{g}z[p(\tau) - p(\tau-\tau')]\}^{1/2}) - I_1{}^2(\{2\Gamma\bar{g}z[p(\tau) - p(\tau-\tau')]\}^{1/2})\right]\right), \quad (11)$$

where the limit $\tau \to \infty$ is understood. The formal resemblance of this equation to $I_{TR}(\tau)$ in Eq. (7) and the stationarity of $A(\tau)$ enable one to show that

$$\langle I_{SS}\rangle = 2\Gamma \int_0^\infty d\tau' e^{-2\Gamma\tau'} \langle I_{TR}(\tau')\rangle. \quad (12)$$

So the above results for the averaged transient intensity can be used to obtain the steady-state result.

In the limit of infinite bandwidth $\Gamma_L \gg \Gamma$, Eq. (8) is inserted into Eq. (12) and the integral performed[14] to give

$$\langle I_{SS}\rangle = \tfrac{1}{2}\Gamma\bar{g}z[I_0(\tfrac{1}{2}\bar{g}z) - I_1(\tfrac{1}{2}\bar{g}z)]\exp(\tfrac{1}{2}\bar{g}z), \quad (13)$$

which is identical with the case of a coherent laser,[9] as it must be.

In the monchromatic limit $\Gamma_L \ll \Gamma$, Eq. (9) is inserted into Eq. (12) and the integral performed[14] to give

$$\langle I_{SS}\rangle = \tfrac{1}{2}\Gamma\bar{g}z/(1 - \bar{g}z)^{1/2}, \quad (14)$$

which shows a singularity as $\bar{g}z$ approaches unity. This result in the generator is closely related to the analogous limit in the Raman amplifier, where an input Stokes wave with intensity I_{S0} is found[2,7,8] to be amplified to a level

$$\langle I_{SS}\rangle_{AMP} = I_{S0}/(1 - \bar{g}z). \quad (15)$$

This result can be simply obtained by averaging the coherent result for the amplifier, $I_{SS}{}^{COH} = I_{S0}\exp(gz)$, over pump intensity (or gain coefficient g) with an exponential probability distribution

$$\langle I_{SS}\rangle_{AMP} = \int_0^\infty I_{SS}{}^{COH}\frac{\exp(-g/\bar{g})}{\bar{g}}\,dg. \quad (16)$$

This leads readily to Eq. (15). In fact, our result [Eq. (14)] can be obtained in similar fashion by replacing $\bar{g}$ by g in Eq. (13), which is then the coherent result, and averaging Eq. (13) over pump intensity (gain) as in Eq. (16).

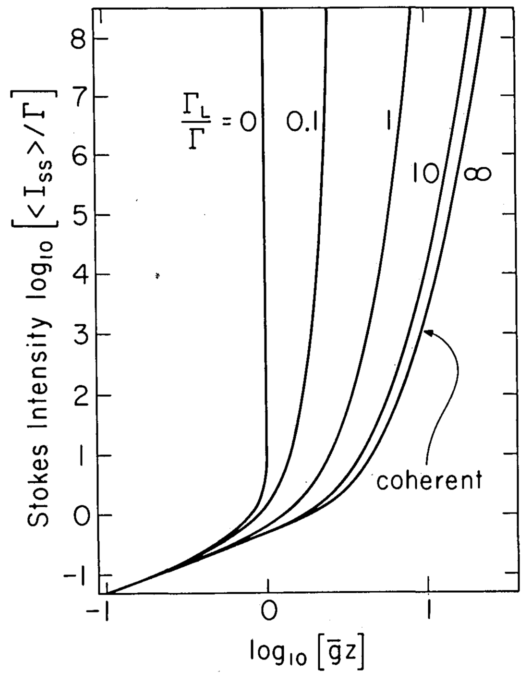

Fig. 2. Mean generated Stokes intensity in steady-state limit ($\Gamma\tau \to \infty$) versus $\bar{g}z$ for different values of Γ_L/Γ.

In Fig. 2 the mean steady-state Stokes intensity $\langle I_{SS}\rangle$, from Eqs. (12)–(14), is plotted versus $\bar{g}z$ for different ratios Γ_L/Γ. The curve labeled 0 is from Eq. (14); that labeled ∞ and coherent is from Eq. (13). The others are from the numerical evaluation of Eq. (12) with Eqs. (10).

In conclusion, the gain for Stokes-wave *generation* by stimulated Raman scattering of a chaotic pump-laser wave is found to be a strong function of pump bandwidth.[16] This is in contrast to the case of a phase-diffusion pump laser, in which the gain is totally independent of bandwidth.[9] This indicates that the bandwidth dependence of the gain is sensitive to the detailed nature of the pump fluctuations. In both transient and steady-state limits the case of a chaotic pump with infinite bandwidth is identical with the case of a coherent pump of the same power. The gain with a finite-bandwidth pump is always found to be larger than that with a coherent pump. This gain enhancement by pump incoherence is of similar origin to that found in N-photon ionization, which depends on the pump intensity as I^N. When averaged over intensities as in Eq. (16), the ionization goes as $\langle I^N\rangle = N!\langle I\rangle^N$.[11,15] In the steady-state Raman case the gain diverges when $\Gamma_L/\Gamma \to 0$. This divergence is prevented in practice by gain saturation, which is usually due to pump depletion.[2]

Finally, a comparison is made between our results and two experimental results.[5,17] In those studies the threshold gain for Stokes generation was measured versus pump-laser bandwidth. If we define threshold gain $\bar{g}_{TH}$ to be that at $\Gamma^{-1}\langle I_{SS}\rangle = 10^{15}$, our results shown in Fig. 2 lead to the dependence $\bar{g}_{TH} \propto (\Gamma_L/\Gamma)^{0.6}$ near the point $\Gamma_L = \Gamma$. The experiments in Refs. 17 and 5 led to $\bar{g}_{TH} \propto (\Gamma_L/\Gamma)^{0.2}$ and $\bar{g}_{TH} \cong$ constant, respectively. Neither is in agreement with the present results for a chaotic pump, probably because the lasers used were not well described by the chaotic model. Laser gain saturation and/or large frequency separations between modes are probable reasons for this. Further experiments are needed to clarify this situation.

APPENDIX A

In this appendix we describe the averaging procedure leading to the key results [Eqs. (8)–(10)]. We first note that Eq. (7) can be written in the alternative form[14]

$$\langle I_{TR}(\tau)\rangle = \frac{\alpha z}{2\pi}\left\langle |A(\tau)|^2 \int_0^{\pi/2} d\theta \frac{I_1\{2\cos\theta[\alpha z p(\tau)]^{1/2}\}}{\cos\theta[\alpha z p(\tau)]^{1/2}}\right\rangle, \tag{A1}$$

where $\alpha = 2\Gamma\bar{g}$ is defined for brevity. Next we express the modified Bessel function by its inverse Laplace transform

$$(z/\kappa)^{1/2} I_1[2(\kappa z)^{1/2}] = \frac{1}{2\pi i}\int_C d\zeta \zeta^{-2} e^{\zeta z} e^{\kappa/\zeta}, \tag{A2}$$

and we insert a derivative $d/d\tau$ to remove the factor $|A(\tau)|^2$ to give

$$\langle I_{TR}(\tau)\rangle = \frac{\alpha}{2\pi}\frac{1}{2\pi i}\int_0^{\pi/2} d\theta \int_C d\zeta \frac{e^{\zeta z}}{\zeta\alpha\cos^2\theta} \times \frac{d}{d\tau}\langle \exp[\zeta^{-1}\alpha p(\tau)\cos^2\theta]\rangle. \tag{A3}$$

The average appearing in this expression can be evaluated exactly for chaotic light by using the known result[12]

$$\left\langle \exp\left[\gamma\int_0^\tau |A(\tau')|^2 d\tau'\right]\right\rangle = 2\{(1 + r/q)\exp[\Gamma_L\tau(q-1)] + (1 - r/q)\exp[-\Gamma_L\tau(q+1)]\}^{-1}, \tag{A4}$$

where $r = 1 - \gamma/\Gamma_L$ and $q = (1 - 2\gamma/\Gamma_L)^{1/2}$. For simplicity we specialize to the monochromatic limit $\Gamma_L \to 0$, where Eq. (A4) becomes

$$\left\langle \exp\left(\gamma\int_0^\tau |A(\tau')|^2 d\tau'\right)\right\rangle = (1 - \gamma\tau)^{-1}. \tag{A5}$$

Using this formula in Eq. (A3) and changing variables to $s = \zeta/\alpha\tau\cos^2\theta$, we get

$$\langle I_{TR}(\tau)\rangle = \frac{\alpha}{2\pi}\frac{1}{2\pi i}\int_0^{\pi/2} d\theta \int_C ds \frac{\exp(sz\alpha\tau\cos^2\theta)}{\alpha\tau\cos^2\theta(s-1)^2}. \tag{A6}$$

Now, from Eq. (A1) it is evident that $\langle I_{TR}(\tau)\rangle = 0$ at $z = 0$. Thus we can rewrite Eq. (A6) as

$$\langle I_{TR}(\tau)\rangle = \frac{\alpha}{2\pi}\frac{1}{2\pi i}\int_0^{\pi/2} d\theta \int_C ds \frac{[\exp(sz\alpha\tau\cos^2\theta) - 1]}{\alpha\tau\cos^2\theta(s-1)^2}, \tag{A7}$$

which can in turn be rewritten as

$$\langle I_{TR}(\tau)\rangle = \frac{\alpha}{2\pi}\frac{1}{2\pi i}\int_0^{\pi/2} d\theta \times \int_C ds \frac{zs}{(s-1)^2}\int_0^1 dx \exp(xsz\alpha\tau\cos^2\theta). \tag{A8}$$

Integrating first over θ, then over x,[18] we find that

$$\langle I_{TR}(\tau)\rangle = \frac{\alpha z}{4}\frac{1}{2\pi i}\int_C ds \frac{s}{(s-1)^2} \times \exp(\tfrac{1}{2}sz\alpha\tau)[I_0(\tfrac{1}{2}sz\alpha\tau) - I_1(\tfrac{1}{2}sz\alpha\tau)], \tag{A9}$$

which can be evaluated simply by finding the residue at $s = 1$:

$$\langle I_{TR}(\tau)\rangle = \tfrac{1}{4}\alpha z I_0(\tfrac{1}{2}z\alpha\tau)\exp(\tfrac{1}{2}z\alpha\tau). \tag{A10}$$

This verifies Eq. (9) in the text.

Equations (10) are verified by carrying out the same procedure, using Eq. (A4) instead of Eq. (A5) for the average. This results in replacing $(s - 1)^{-2}$ in Eq. (A6) by $f(s)/s^2$, giving

$$\langle I_{TR}(\tau)\rangle = \frac{\alpha}{2\pi}\frac{1}{2\pi i}\int_0^{\pi/2} d\theta \int_C ds \frac{\exp(sz\alpha\tau\cos^2\theta)}{\alpha\tau\cos^2\theta}\frac{f(s)}{s^2}, \tag{A11}$$

where $f(s)$ is given by Eq. (10b). This reduces back to Eq. (A6) in the limit $\Gamma_L \to 0$. The same steps leading to Eq. (A9) then lead to Eq. (10a).

The infinite-bandwidth limit is obtained by taking $\Gamma_L \to \infty$ in Eq. (A11). In that limit $f(s)$ becomes $\exp(1/s)$. The Laplace-transform relation Eq. (A2) can then be used to find

$$\langle I_{TR}(\tau)\rangle = \frac{\alpha z}{2\pi}\int_0^{\pi/2} d\theta \frac{I_1[2\cos\theta(\alpha z\tau)^{1/2}]}{\cos\theta(\alpha z\tau)^{1/2}}. \tag{A12}$$

This result is seen to be exactly the result for a coherent pump by comparing it with Eq. (A1) while noting that $|A(\tau)| = 1$ and $p(\tau) = \tau$ for a coherent pump. Integrating Eq. (A12) leads directly to Eq. (8).

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