Linewidth of a traveling-wave Raman laser excited by nonmonochromatic radiation

G. A. Pasmanik and M. S. Sandler

Scientific Research Radiophysics Institute
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A theory of the linewidth of a Raman laser excited by nonmonochromatic pump radiation with a spectral width much smaller than the linewidth of spontaneous scattering is developed. It is shown that the linewidth is determined by the additive contributions of two components, one of which is connected with the nonmonochromatic nature of the pump and the other with spontaneous and thermal noise in the medium and the field. The contribution of these two factors near and well away from the excitation threshold of the Raman laser is discussed.

1. There has been a considerable increase in the interest in tunable Raman lasers since 1970 when Patel et al. succeeded in exciting stimulated Raman scattering (SRS) in a resonator in which the Stokes frequency was tuned by using a magnetic field to vary the period of spin oscillations in a semiconductor.[1] This was followed by studies of the various characteristics of the pulsed and continuous operation of such lasers (usually referred to as spin-flip Raman lasers)[2-4] and, in particular, by measurements of the line width of the Raman radiation.[5] However, the theoretical line width was not considered although there was undoubtedly considerable interest in this quantity, especially since the tunable Raman laser has been finding extensive application in spectroscopy.[6]

Before the line width of the Raman laser can be determined, it is necessary to investigate the width of the spectrum of the Stokes component of SRS in the resonator. This problem is solved in the present paper for the case of the excitation of SRS by a traveling pump wave within the framework of the classical analysis. A derivation is given of the line width of the Raman laser and it is shown that its value is determined by the additive contributions of two terms, one of which is connected with the effect of the nonmonochromatic nature of the pump and the other with spontaneous and thermal noise producing a natural broadening of the Stokes spectrum. The contribution of the two factors near and well away from the threshold for the excitation of the Raman laser is discussed.

We note that the results obtained below refer not only to traveling-wave spin-flip Raman lasers, but also to other continuous generators whose operation is based on SRS in a resonator transparent to the external pump.

2. We shall consider a Raman laser whose working material is located in a circular resonator in which the pump wave produces SRS. We shall describe SRS by the standard set of truncated equations for the complex amplitudes of the exciting $\mathscr{E}_0$, scattered $\mathscr{E}_1$, and phonon Q waves:[7] 1)

$$\left(\frac{1}{v_{0,1}}\frac{\partial}{\partial t}+\frac{\partial}{\partial z}+\frac{\alpha_{0,1}}{2}\right)\mathscr{E}_{0,1}=ig_{0,1}\mathscr{E}_{1,0}\left\{\begin{matrix}Q\\Q^*\end{matrix}\right\}+f_{0,1}, \quad (1)$$

$$\frac{\partial Q}{\partial t}+T_2^{-1}Q=ig_3\mathscr{E}_0\mathscr{E}_1^*+\zeta(z,t). \quad (2)$$

These two equations take into account extraneous sources of noise which are responsible for spontaneous Raman scattering. The spectral source—correlation functions are, respectively, given by

$$\langle f_{0,1}(\omega,z)f_{0,1}^*(\omega',z')\rangle=\frac{\Theta(\omega_{0,1})\alpha_{0,1}}{v_{0,1}\varepsilon_0 S}\delta(\omega-\omega')\delta(z-z'), \quad (3)$$

$$\langle\zeta(\omega,z)\zeta^*(\omega',z')\rangle=\frac{\Theta(\Omega_0)}{2\pi\Omega_0^2\rho_0T_2S}\delta(\omega-\omega')\delta(z-z'). \quad (4)$$

In these expressions, T_2 and Ω_0 are, respectively, the relaxation time and frequency of optical phonons, $\omega_1=\omega_0-\Omega_0$ is the frequency of the Stokes radiation, S is the cross-sectional area of beams in the resonator, ϵ_0 is the permittivity, ρ_0 is the density of the medium, and

$$\Theta(\omega)=\frac{\hbar\omega}{2}+\frac{\hbar\omega}{\exp(\hbar\omega/kT)-1}.$$

We shall assume that the circular resonator in which SRS takes place is tuned to the Stokes component and is transparent to the pump. The reflection coefficient of the mirror for Raman radiation is denoted by R, and the distributed losses in the medium are the same for both waves, $\alpha=\alpha_0=\alpha_1$. We shall confine our attention to the case when the width $\delta\omega_0$ of the pump spectrum is small in comparison with the line width T_2^{-1} of spontaneous scattering. This condition is usually satisfied in experiments.[3-5]

We shall write the solution of Eq. (2) in integral form and assume that the field $\mathscr{E}_0$ and $\mathscr{E}_1$ vary slowly along the time axis in comparison with the relaxation time T_2. We then expand the integrand into a Doppler series. If we substitute the result in Eq. (1), we finally obtain

$$\left(\frac{1}{v_{0,1}}\frac{\partial}{\partial t}+\frac{\partial}{\partial z}+\frac{\alpha}{2}\right)\mathscr{E}_{0,1}=\mp g_{0,1}g_3\left(\mathscr{E}_{0,1}|\mathscr{E}_{1,0}|^2-T_2\mathscr{E}_{1,0}\frac{\partial}{\partial t}\left\{\begin{matrix}\mathscr{E}_0 & \mathscr{E}_1^*\\ \mathscr{E}_0^* & \mathscr{E}_1\end{matrix}\right\}\right)$$
$$+ig_{0,1}\mathscr{E}_{1,0}\int_0^t\exp\left(-\frac{u}{T_2}\right)\zeta(z,t-u)\,du+f_{0,1}(z,t). \quad (5)$$

3. We now use Eq. (5) to set up the equations for the phases φ_0 and φ_1 of the exciting and scattered waves

$$\left(\frac{1}{v_{0,1}}\frac{\partial}{\partial t}+\frac{\partial}{\partial z}\right)\varphi_{0,1}-g_{0,1}g_3T_2^2A_{1,0}^2\frac{\partial\Phi}{\partial t}$$
$$=g_{0,1}\frac{A_{1,0}T_2}{A_{0,1}}(\zeta'\cos\Phi+\zeta''\sin\Phi)+\frac{1}{A_{0,1}}(f_{0,1}'\cos\varphi_{0,1}+f_{0,1}''\sin\varphi_{0,1}). \quad (6)$$

In these expressions $\Phi=\varphi_0-\varphi_1$, ζ' and ζ'' are the real and imaginary parts of the noise source

$$\frac{1}{T_2}\int_0^t\exp\left(-\frac{u}{T_2}\right)\zeta(z,t-u)\,du,$$

and $f_{0,1}'$ and $f_{0,1}''$ are the real and imaginary parts of the source $f_{0,1}$. The boundary conditions for the circular resonator must be written in the form

$$\varphi_0(z=0,t)=\varphi_0(t),\quad \varphi_1(z=0,t)=\varphi_1(z=l,t)+\xi(t). \quad (7)$$

In these expressions, $\xi(t)$ is the correction to the phase of the Raman wave, which is due to the influence of zero-point vacuum fluctuations which penetrate the semitransparent mirror (within the framework of the classical analysis which we are giving here). Simple calculations show that

$$\langle \xi(t)\xi(t')\rangle = \frac{(1-R)\Theta(\omega_1)}{4\pi v\varepsilon_0 S A_1^2(0)}\delta(t-t'). \quad (8)$$

Since the line width of the Raman radiation in the continuous state is determined by phase fluctuations, the determination of the line width requires the solution of Eq. (6) subject to the boundary conditions given by Eq. (7). It is clear from Eq. (6) that relatively small fluctuations in the amplitudes of the interacting waves have no substantial effect on the phase fluctuations. We shall use this fact in the determination of the line width and neglect the effect of small changes in the amplitude due to spontaneous and thermal noise, zero-point fluctuations penetrating the resonator through the semitransparent mirror, and possible small fluctuations in the pump intensity at the entrance to the nonlinear medium.[2)] The amplitudes A_0 and A_1 in Eq. (6) may be regarded (in the continuous state) as functions of only the longitudinal coordinate z, and their steady-state distribution may be described by

$$\left(\frac{\partial}{\partial z}+\frac{\alpha}{2}\right)A_{0,1} = \mp g_{0,1}g_3T_2A_{0,1}A_{1,0}^2 \quad (9)$$

with the boundary conditions

$$A_0^2(0)=A_{00}^2, \quad A_1^2(0)=RA_1^2(l). \quad (10)$$

To simplify our analysis, we shall also assume that the velocities of the two waves are equal, i.e., $v = v_0 = v_1$. Since the functions φ_0 and φ_1 are "slow," it can be shown that the statistical properties of the right-hand sides of Eq. (6) do not depend on the values of Φ, φ_0, and φ_1. In other words, Eq. (6) is statistically equivalent[9] to the set of equations, the left-hand sides of which coincide with Eq. (6) and the right-hand sides are, correspondingly, equal to

$$\frac{g_{0,1}A_{1,0}T_2}{A_{0,1}}\zeta' + \frac{1}{A_{0,1}}f'_{0,1}.$$

We must now set up the equations for Φ. Subtracting the second equation in Eq. (6) from the first, we have

$$\frac{1}{v}\frac{\partial\Phi}{\partial t}+\frac{\partial\Phi}{\partial z} - g_3T_2(g_0A_1^2-g_1A_0^2)\frac{\partial\Phi}{\partial t} = \left(g_0\frac{A_1}{A_0}-g_1\frac{A_0}{A_1}\right)T_2\zeta' + \frac{f_0'}{A_0}-\frac{f_1'}{A}. \quad (11)$$

Solving Eq. (11) and substituting the solution into Eq. (6), we can determine φ_0 and φ_1. Next, using the boundary conditions, we find the phase of the Stokes radiation in the z = 0 plane and its Fourier component $\varphi_{1\omega}(0)$. We shall require the formula for the Fourier component near the zero frequency. Omitting the intermediate steps, we reproduce here the final result:

$$\varphi_{1\omega}|_{\omega\to 0} = L/M,$$

$$L = i\omega\varphi_{0\omega}(0)\int_0 dz\, g_1g_3T_2A_0^2(z) + \int_0 \frac{g_1A_0(z)}{A_1(z)}\zeta_\omega'(z)\,dz + \int_0^l \frac{f_{1\omega}'(z)}{A_1(z)}dz + \xi(\omega), \quad (12)$$

$$M = i\omega\left(\frac{l}{v}+\int_0^l g_1g_3T_2A_0^2(z)\,dz\right).$$

4. We now proceed directly to the determination of the spectrum of the Stokes component. We use the well-known formula relating the spectrum width $\delta\omega$ with the power spectrum of frequency fluctuations S_ω:[9]

$$\delta\omega = 2\pi \lim_{\omega\to 0} S_\omega, \quad (13)$$

where

$$\langle\varphi_\omega\varphi_{\omega'}\rangle = \frac{S_\omega}{\omega^2}\delta(\omega-\omega'). \quad (14)$$

Substituting for $\varphi_{1\omega}(0)$ in Eqs. (13) and (14), and using Eqs. (3), (4), and (8) and the fact that

$$\int_0^l \frac{A_0^2(z)}{A_1^2(z)}dz = \frac{\alpha}{2g_1g_2T_2}\int_0^l \frac{dz}{A_1^2(z)} + \frac{1-R}{2g_1g_2T_2A_1^2(0)}, \quad \int_0^l A_0^2(z)\,dz = \frac{\alpha l - \ln R}{2g_1g_2T_2}$$

[the last two results follow from the amplitude equation (9) and the boundary conditions (10)], we find the width of the spectrum of the Stokes radiation

$$\delta\omega = \delta\omega_1^{(1)} + \delta\omega_1^{(2)}. \quad (15)$$

In this expression $\delta\omega_1^{(1)}$ is the broadening of the spectrum due to the nonmonochromatic nature of the pump:

$$\delta\omega_1^{(1)} = \frac{(T_2/T_p)^2}{(1+T_2/T_p)^2}\delta\omega_0, \quad (16)$$

where $T_p = 2[v(\alpha - \ln R/l)]^{-1}$ is the characteristic damping time for the amplitude of the Raman radiation in the resonator and $\delta\omega_0 = 2\pi \lim S_{0\omega}$ is the width of the pump spectrum. The value of $\delta\omega_1^{(2)}$ which characterizes the so-called natural broadening of the line is given by

$$\delta\omega_1^{(2)} = \frac{\pi v\omega_1(\Theta(\Omega_0)/\Omega_0+\Theta(\omega_1)/\omega_1)}{Sl^2\varepsilon_0(1+T_2/T_p)^2}\left(\alpha\int_0^l\frac{dz}{A_1^2(z)}+\frac{1-R}{A_1^2(0)}\right). \quad (17)$$

It is interesting to note that the value of $\delta\omega_1^{(1)}$ when T_2 is replaced with the damping time for the free wave coincides with the corresponding term in the formula for the line width of the localized parametric generator excited by nonmonochromatic radiation.[10] The expression for $\delta\omega_1^{(2)}$ in the resonator case ($R \to 1$) is similar to the expression for the natural line width of the laser and the parametric oscillator obtained within the framework of the localized model.

It follows from Eq. (17) that the natural width of the spectrum is determined by the additive contribution of spontaneous noise in the medium [term proportional to $\Theta(\Omega_0)/\Omega_0$] and the thermal noise associated with the electromagnetic field in the resonator and on the semitransparent mirror [term proportional to $\Theta(\omega_1)/\omega_1$]. In the quantum region ($\hbar\Omega_0$, $\hbar\omega_1 \gg kT$), the contribution of the two factors which lead to the natural line broadening is the same. If, on the other hand, $\hbar\Omega_0 \ll kT \ll \hbar\omega_1$, or $\hbar\Omega_0 \ll \hbar\omega_1 \ll kT$, the natural width of the spectrum is determined by spontaneous noise in the medium; in the opposite case ($\hbar\omega_1 \ll kT \ll \hbar\Omega_0$, or $\hbar\omega_1 \ll \hbar\Omega_0 \ll kT$), the value of $\delta\omega_1^{(2)}$ is determined by the thermal noise in the field.[3)]

5. Let us consider Eqs. (16) and (17) in greater detail. We begin with the natural line broadening. To determine $\delta\omega_1^{(2)}$, we must find the spatial distribution of the intensity of the Raman radiation in the resonator, i.e., solve Eq. (9). We shall not write out here the solution of Eq. (9) in the general form, but confine our attention to the detailed analysis of the natural line width for $\alpha l \ll 1$ which is the case of interest in practice.

The dependence of $m_1 = A_1^2$ and $m_0 = A_0^2$ on the longitudinal coordinate is

$$m_1 = \frac{\Gamma m_1(0)\exp(-\alpha z)}{2[\gamma_0 m_1(0)+\gamma_1 m_0(0)\exp(-\Gamma z)]}, \quad m_0 = \frac{\Gamma - 2\gamma_0 m_1}{2\gamma_1}, \quad (18)$$

where $\Gamma = 2\gamma_0 m_1(0) + 2\gamma_1 m_0(0)$ and $\gamma_{0,1} = g_{0,1}g_2T_2$. The value of $m_1(0)$ is obtained by solving the transcendental equation

$$(Re^{-\alpha l} - e^{-\Gamma l})/(1-Re^{-\alpha l}) = \gamma_0 m_1(0)/\gamma_1 m_0(0). \quad (19)$$

The integral I in Eq. (17) is given by

$$I=\int_0^l \frac{dz}{m_1}=\frac{2\gamma_0 l}{\Gamma}\left(1+\frac{\gamma_1 m_0(0)}{\gamma_0 m_1(0)}\frac{1-e^{-(\Gamma-\alpha)l}}{(\Gamma-\alpha)l}\right). \qquad (20)$$

The generation threshold is given by

$$2\gamma_1 m_{0\,\mathrm{th}}(0)l=\alpha l-\ln R. \qquad (21)$$

If $1-R \ll 1$ and $\alpha l \ll 1$, we have the threshold condition $2\gamma_1 m_{0\mathrm{th}}(0)l \ll 1$. Near the threshold, the gain per pass is small, i.e., $2\gamma_1 m_0(0)l \ll 1$. For a small (but sufficient to enable us to neglect amplitude fluctuations) excess over the threshold $[m_0(0)-m_{0\mathrm{th}}(0) \ll m_{0\mathrm{th}}(0)]$ we have the approximate formulas

$$m_1(0)\approx\frac{m_0(0)-m_{0\,\mathrm{th}}(0)}{3m_{0\,\mathrm{th}}(0)\gamma_0 l}, \qquad I=\frac{l}{m_1(0)}. \qquad (22)$$

When $2\gamma_1 m_0(0)l \gtrsim 1$ (substantial gain per pass)

$$m_1(0)=\frac{R\exp(-\alpha l)}{1-R\exp(-\alpha l)}\frac{\gamma_1}{\gamma_0}m_0(0), \qquad I=\frac{l}{m_1(0)}. \qquad (23)$$

The natural line width for both the above cases is

$$\delta\omega_1^{(2)}=\frac{v\omega_1(\Theta(\Omega_0)/\Omega_0+\Theta(\omega_1)/\omega_1)}{lT_pP_1(1+T_2/T_p)^2}, \qquad (24)$$

where $P_1 = v\epsilon_0 m_1(0)S/2\pi$ is the power carried by the Raman radiation in the z = 0 plane. We have used the fact that for $1-R \ll 1$ we can replace $1-R$ by $-\ln R$.

We now note the following point. The formula given by Eq. (24) was obtained on the approximation $R \to 1$, and if we express P_1 in terms of the power of the radiation dissipated in the medium and through the resonator walls, i.e., $P_1 = P_{1D}vT_p/2l$, then the corresponding expression for $\delta\omega_1^{(2)}$ when T_2 is replaced by the damping time for the free wave becomes identical with that obtained earlier for the natural line width of the parametric generator.[10] We can then use the analogy between the formulas for the line width of the parametric and quantum-mechanical generators, which was noted in[10]. To achieve this, the expression $\Theta(\Omega_0)/\hbar\Omega_0 + \Theta(\omega_1)/\hbar\omega_1$ will be written in the form

$$1+\bar{n}_{\Omega_0}+\bar{n}_{\omega_1} \quad (\bar{n}_{\Omega_0}=[\exp(\hbar\Omega_0/kT)-1]^{-1} \text{ and } \bar{n}_{\omega_1}=[\exp(\hbar\omega_1/kT)-1]^{-1}$$

which gives the mean numbers of the optical phonons and Stokes photons in equilibrium. Equation (24) with $\bar{n}_{\Omega_0}+\bar{n}_{\omega_1}$ replaced by the sum of the number of thermal photons and photons responsible for spontaneous emission, is identical with the formula for the line width due to the quantum-mechanical generator (see, for example,[11]).

We must now analyze the expression given by Eq. (24). Near the generation threshold, P_1 tends to zero [see Eq. (22)] and the natural line broadening can be quite large. However, if the gain per transit through the resonator is greater than unity, i.e., $2\gamma_1 m_0(0)l \gtrsim 1$, the Raman power becomes considerable and the natural line width can be very small. For example, when $\Omega_0/\omega_1 = 0.2$, we have $\gamma_1/\gamma_0 = 0.8$ and when $Re^{-\alpha l} = 0.8$, we have $P_1(0) = 3P_0(0)$. When $P_0(0) = 1$ W, $T_p = 2.5 \times 10^{-10}$ sec, $T_2 = 10^{-11}$ sec, $v = 2 \times 10^{10}$ cm/sec, $\omega_1 = 4 \times 10^{14}$ sec^{-1}, $T = 2°$ K, $l = 1$ cm, the natural broadening is $\delta\omega_1^{(2)} = 0.5$ sec^{-1}. The output power $P_{1\,\mathrm{out}} = (1-R)P_1(0)$ is 0.6 W (when $R \approx 0.8$).

When $R \ll 1$ and the excess over the threshold is $m_0 = m_{0\mathrm{th}}(0) \gtrsim (\gamma_1 l)^{-1}$, the value of $m_1(0)$ can be obtained from the formula ($\alpha l = 0$)

$$m_1(0)\approx\gamma_1 R m_0(0)/\gamma_0. \qquad (25)$$

The natural line width is

$$\delta\omega_1^{(2)}=\frac{v^2\omega_1(\Theta(\Omega_0)/\Omega_0+\Theta(\omega_1)/\omega_1)}{2l^2(1+T_2/T_p)^2P_1}. \qquad (26)$$

When $P_0(0) = 5$W, $R = 0.1$, $\gamma_1/\gamma_0 \simeq 0.8$, the Raman radiation power is 0.4 W. For $\omega_1 = 4 \times 10^{14}$ sec^{-1}, $T = 2°$K, $l = 1$ cm, $v = 2 \times 10^{10}$ cm/sec, the natural broadening is $\delta\omega_1^{(2)} = 8$ sec^{-1}.

We now consider the contribution to broadening due to the fact that the pump is nonmonochromatic. When $T_2 \ll T_p$, we have

$$\delta\omega_1^{(1)}=(T_2/T_p)^2\delta\omega_0. \qquad (27)$$

If we suppose, following[5], that $T_2 = 10^{-11}$ sec, $T_p = 2.5 \times 10^{-10}$ sec, $\delta\omega_0 = 5 \times 10^6$ sec^{-1}, we obtain $\delta\omega_1^{(1)} = 8000$ sec^{-1}, which is much greater than $\delta\omega_1^{(2)}$. The approximate result $\delta\omega_1 \approx \delta\omega_1^{(1)} = 8000$ sec^{-1} is in very approximate correspondence with the measured value of the line width of Raman radiation in InSb.[5] It follows that the foregoing analysis leads to the conclusion that the line broadening in the Raman laser is largely determined by the nonmonochromatic nature of the pump and to reduce $\delta\omega_1$ for $\delta\omega_0 < T_2^{-1}$, the width of the spectrum of the exciting radiation or the parameter T_2/T_p must be reduced.

1) In deriving Eqs. (1) and (2), we neglect the change in the population difference between the vibrational levels in the course of SRS. [8].

2) It is important to note, however, that near the generation threshold, when the mean value of the Stokes component is comparable with the magnitude of its amplitude fluctuations, the effect of the latter may turn out to be important. Such small excesses above the threshold will not be considered here.

3) When account is taken of the change in the difference between the populations of vibrational levels during SRS, the natural line width $\delta\omega_1^{(2)}$ will, in general, be slightly different, but an estimate of this quantity can be obtained by substituting the value of Θ which is a function of the new effective temperature (determined with allowance for the change in the populations) into the previous formula. However, in most cases of interest in practice, the change in the populations during SRS is quite negligible. [8]

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