PHYSICAL REVIEW A VOLUME 12, NUMBER 4 OCTOBER 1975

# Transient behavior of the amplification of the Stokes field by thermal Rayleigh and Brillouin effects

A. Bambini, R. Vallauri, and M. Zoppi

*Consiglio Nazionale delle Ricerche, Laboratorio di Elettronica Quantistica, Via Panciatichi 56/30-50127 Firenze, Italy*

(Received 3 February 1975; revised manuscript received 19 May 1975)

We derive analytical solution for a signal field amplified by stimulated Rayleigh and Brillouin effects. The presence of light absorption is taken into account. The solution describes the transient behavior at all frequency differences between the pump and the signal waves. The validity is demonstrated for the simple case of no absorption, and the particular time behavior at $\omega = 0$ is stressed.

## I. INTRODUCTION

In 1970 Pohl and Kaiser published an article[1] that can be considered the conclusion of the work of many years on the study of the transient behavior of the amplification of an electromagnetic wave interacting with a strong field by density and temperature fluctuations. The very accurate experimental techniques used allowed measurement of the very fast phonon lifetime and investigation in the transient regime to ascertain the characteristic time before the steady state is reached.[2-6] The addition of small quantities of dyes also allowed the study of the influence of heat absorption on the Brillouin line of gain.

The differential-equation system of Ref. 1 was solved by Pohl and Kaiser by introducing a series expansion in the $z$ coordinate to first order, with various expressions for the input and pump waves as a function of time. The same theoretical method was used by Rangnekar and Enns, who extended the calculation to second order.[7] They were also interested in the transient behavior near the Rayleigh line which was studied in previous works.[8,9] In this region the first-order perturbation approach used by Pohl and Kaiser in Ref. 1 fails.

A different approach was used by Rother[10] in order to solve the differential-equation system (15)–(17) of Ref. 1. He succeeded in integrating by Fourier-transforming the system and by separating the contribution to the spectrum of the amplified signal into three different parts, namely, the Rayleigh and Brillouin (Stokes and anti-Stokes) frequencies. In particular, he was able to explain the experimental results obtained[11] when the interaction between a strong pumping field at frequency $\omega_L$ and a signal wave at the same frequency occurs in liquid $CCl_4$.

We have obtained a nonperturbative solution to Eqs. (15)–(17) of Ref. 1 in terms of the amplitude of the signal field for arbitrary frequencies of this field. As will become apparent, this solution allows one to consider the influence of all the components of the amplified field on any particular region of the spectrum.

## II. THEORY

We start from Eqs. (15)–(17) of Ref. 1, which we write out in the proper form

$$\left(\frac{k^2v^2}{\gamma} - \omega^2 + i\omega\Gamma_B + (2i\omega + \Gamma_B)\frac{d}{dt} + \frac{d^2}{dt^2}\right)\rho(z,t) + \frac{k^2v^2\beta\rho_0}{\gamma}\,T(z,t) = \frac{\gamma^e k^2}{8\pi}\,E_L E_S^*(z,t)\,, \tag{1}$$

$$-(\gamma-1)\left(i\omega + \frac{d}{dt}\right)\rho(z,t) + \beta\rho_0\left(i\omega + \tfrac{1}{2}\gamma\Gamma_R + \frac{d}{dt}\right)T(z,t) = \frac{\gamma\gamma^a k}{8\pi v}\,E_L E_S^*(z,t)\,, \tag{2}$$

$$\frac{d}{dz}E_S^*(z,t) = -i\,\frac{\gamma^e\omega_S}{4nc\rho_0}\,E_L\,\rho(z,t)\,e^{-\alpha z}\,. \tag{3}$$

This set of differential equations describes, in the slowly-varying-amplitude approximation, the interaction of two electromagnetic fields: a Stokes wave, with amplitude $E_S(z,t)$, traveling in the $-z$ direction with frequency $\omega_S$ and propagating vector $k_S$, and a strong field, with amplitude $E_L$, traveling in the $+z$ direction with frequency $\omega_L$ and propagating vector $k_L$. The interaction occurs through density and temperature fluctuations. These fluctuations have amplitudes indicated by $\rho(z,t)$ and

$T(z,t)$, respectively, and propagate in the $+z$ direction with frequency $\omega$ and propagating vector $k$. Energy and momentum conservation requires that $\omega_L - \omega_S = \omega$ and $k_L + k_S = k \simeq 2k_L$. Other parameters are defined in Ref. 1.

In our derivation we neglect any depletion of the field $E_L$ and consider a step-function pulse input $E_S$, i.e., $E_L(z,t) = E_L$ independent on $z$ and $t$, and $E_S(L,t) = E_S^{(0)}$ denotes the signal amplitude which we assume to be constant at any time $t>0$. Here, $L$ denotes the input position of the Stokes wave.

We take the Laplace transform of Eqs. (1)–(3) using the above stated initial conditions in conjunction with $\rho(0)=0$, $\dot\rho(0)=0$, and $T(0)=0$. Then the resulting equations are solved, and the slowly varying amplitude of the Stokes field turns out to be

$$E_S^* = \frac{1}{2\pi i}\int \frac{E_S^{(0)}}{p}\exp\left[A(z)\left(p + \tfrac{1}{2}\gamma\Gamma_R - \frac{\gamma^a}{\gamma^e}\,\omega_B + i\omega\right)(p-\Omega_1)^{-1}(p-\Omega_2)^{-1}(p-\Omega_3)^{-1}\right]e^{pt}dp\,, \tag{4}$$

where

$$A(z) = i(\gamma^e k E_L)^2\omega_S(e^{-\alpha z} - e^{-\alpha L})/32\pi n c\rho_0\alpha\,. \tag{5}$$

The $\Omega_j$ are the roots of the characteristic equation derived from the Laplace transform of Eqs. (1) and (2), and are given by

$$\Omega_1 = -i\omega - \tfrac{1}{2}\Gamma_R\,,$$

$$\Omega_2 = -i(\omega + \omega_B') - \tfrac{1}{2}\Gamma_B'\,,$$

$$\Omega_3 = -i(\omega - \omega_B') - \tfrac{1}{2}\Gamma_B'\,,$$

where $\omega_B'$ represents the frequency shift of the Brillouin peak from the frequency of the incident radiation, and is given approximately by the well-known Brillouin relation.[12] $\Gamma_B'$ is given by $\Gamma_B + \frac{1}{2}(\gamma-1)\Gamma_R$. For detailed calculations of $\omega_B'$ and $\Gamma_B'$, we refer to Ref. 7.

In order to get an analytical expression for the Stokes field $E_S$, we must calculate the residues of the four poles of the integrand in Eq. (4). The residue in the pole $p=0$ is easily derived and yields the steady-state contribution to the electromagnetic field. For the other three singularities, we proceed as follows. First of all we expand the exponent in the integrand:

$$A(z)\left(p + \tfrac{1}{2}\gamma\Gamma_R - \frac{\gamma^a}{\gamma^e}\,\omega_B + i\omega\right)\Big/[(p-\Omega_1)(p-\Omega_2)(p-\Omega_3)] = A(z)\left(\frac{B_1}{p-\Omega_1} + \frac{B_2}{p-\Omega_2} + \frac{B_3}{p-\Omega_3}\right), \tag{6}$$

where

$$B_1 = [\Omega_1 + i\omega + \tfrac{1}{2}\gamma\Gamma_R - (\gamma^a/\gamma^e)\omega_B]/(\Omega_1-\Omega_2)(\Omega_1-\Omega_3)\,, \tag{7a}$$

$$B_2 = [\Omega_2 + i\omega + \tfrac{1}{2}\gamma\Gamma_R - (\gamma^a/\gamma^e)\omega_B]/(\Omega_2-\Omega_1)(\Omega_2-\Omega_3)\,, \tag{7b}$$

$$B_3 = [\Omega_3 + i\omega + \tfrac{1}{2}\gamma\Gamma_R - (\gamma^a/\gamma^e)\omega_B]/(\Omega_3-\Omega_1)(\Omega_3-\Omega_2)\,. \tag{7c}$$

Then for each singularity we divide the exponential into two parts, one of which is singular while the other one is regular. For example, for the $\Omega_1$ singularity we get

$$\exp\left(A(z)\sum_{j=1}^{3}\frac{B_j}{p-\Omega_j}\right)\exp(pt) = \exp\left(A(z)\frac{B_1}{p-\Omega_1} + (p-\Omega_1)t\right) \times \exp\left(A(z)\sum_{j=2}^{3}\frac{B_j}{p-\Omega_j} + \Omega_1 t\right). \tag{8}$$

By the use of the generating function for Bessel functions,

$$\exp\left[\frac{1}{2}\,x\left(y - \frac{1}{y}\right)\right] = \sum_{s=-\infty}^{+\infty} y^s J_s(x)\,, \tag{9}$$

it is possible to expand the integrand in a series of $(p-\Omega_j)^l$, where $l$ ranges from $-\infty$ to $+\infty$, and to extract the $l=-1$ contribution. By this method we evaluate the inverse Laplace transform on the right-hand side of Eq. (4). The expression for the slowly varying amplitude $E_S(z,t)$ is inserted in Eq. (9) of Ref. 1, obtaining for the Stokes field $\mathcal{E}_S$ (which contains also the optical frequencies)

$$\mathcal{E}_S(z,t) = \tfrac{1}{2}E_S^{(0)}e^{\alpha z/2}\Big\{\exp\left[A(z)\left(\frac{\gamma^a}{\gamma^e}\,\omega_B - \tfrac{1}{2}\gamma\Gamma_R - i\omega\right)\Big/\Omega_1\Omega_2\Omega_3\right]e^{-i(\omega_S t + k_S z)} + C_1(z,t)e^{-i(\omega_L t + k_S z)}e^{-\Gamma_R t/2} + C_2(z,t)e^{-i[(\omega_L+\omega_B)t + k_S z]}e^{-\Gamma_B' t/2} + C_3(z,t)e^{-i[(\omega_L-\omega_B)t + k_S z]}e^{-\Gamma_B' t/2} + \text{c.c.}\Big\}\,, \tag{10}$$

where

$$C_j(z,t)=\sum_{m=1}^{\infty} b_m(\Omega_j)\left(-\frac{A(z)B_j}{t}\right)^{m/2} J_m\{2[-A(z)B_jt]^{1/2}\}\ , \tag{11}$$

where $b_m(\Omega_j)$ are coefficients independent of time $t$; e.g., for $j=1$ it reads

$$b_m(\Omega_1)=\frac{(-1)^m}{(m-1)!}\frac{d^{m-1}}{dp^{m-1}} \times\left\{\frac{1}{p}\exp\left[A(z)\left(\frac{B_2}{p-\Omega_2}-\frac{B_3}{p-\Omega_3}\right)\right]\right\}\Bigg|_{p=\Omega_1}.$$

The physical meaning of the result derived in Eq. (10) is easily understandable. The first term represents the input wave spatially amplified in the steady-state regime. The other three components describe the transient behavior as the sum of a Rayleigh process (term labeled by 1) and a Brillouin process (the anti-Stokes and Stokes components labeled by 2 and 3, respectively). The frequencies of these three components are imposed by the thermal fluctuations stimulated in the medium by the coupling of the two electromagnetic fields.

In order to compare our results with experimental data, one should take the square modulus of the electric field expressed by Eq. (10), because any detector reveals the intensity rather than the field itself. Because the time resolution of photodetectors is approximately 0.3 nsec, one can drop the fast oscillations which arise taking the square modulus of the electric field.

First we wish to consider the case in which the Rayleigh component vanishes. This can be achieved by setting $\alpha=0$ and $\lambda=0$. These two conditions mean that both the thermal heating and the presence of spontaneous temperature fluctuations are neglected (i.e., $\gamma^a=0$ and $\Gamma_R=0$). In this physical situation the argument of the Bessel functions on the right-hand side of Eq. (11) is real or pure imaginary, as $j$ is equal to 2 or 3, respectively. The coefficient $C_2(z,t)$ turns out to be a sum of Bessel functions evaluated on the real axis and tends to zero as the argument (i.e., time $t$) increases. In contrast, $C_3(z,t)$ tends to infinity as the time $t$ increases, being the sum of Bessel functions evaluated on the imaginary axis. We find the well-known result that anti-Stokes components cannot be excited by the stimulated Brillouin effect.

Away from the limit $\alpha=0$, we study the behavior of the amplified signal at $\omega=0$. In the range of frequencies near the Rayleigh line the process is recognized to be wholly transient during the time duration of experimental pulses.[5,11] This result was confirmed by Darée and Kaiser,[13] who analyzed their experimental results using Rother's solution[10] in order to take into account the time variation of the pumping field.

Even if our solution is restricted to the case in which the laser intensity is kept constant during the interaction, the present treatment makes it possible to distinguish the influence of different components on the total amplified intensity. From Eq. (10) we see that the main contribution to the intensity comes from the Rayleigh component (namely, the square modulus of the term labeled by 1) and from the beating between the Rayleigh component and the stationary term. These two terms have a decay constant approximately equal to $1/\Gamma_R$, thus confirming that the temperature fluctuations drive these components. But, in addition to this, we can also take into account the contribution of other components. In particular, it turns out that the beatings between the Stokes component (labeled by 3) and the stationary and Rayleigh components are important in some cases, namely, when the phonon lifetime is so long that it becomes comparable with the time duration of the temperature fluctuations. This result is apparent in Figs. 1 and 2.

In Fig. 1 we show the contribution of the above-mentioned terms for liquid-$CCl_4$ parameters. In this case the ratio $\Gamma_B/\Gamma_R$ is $\simeq 36$, and it appears that only the steady-state and Rayleigh components are important. Thus, the approximation used by

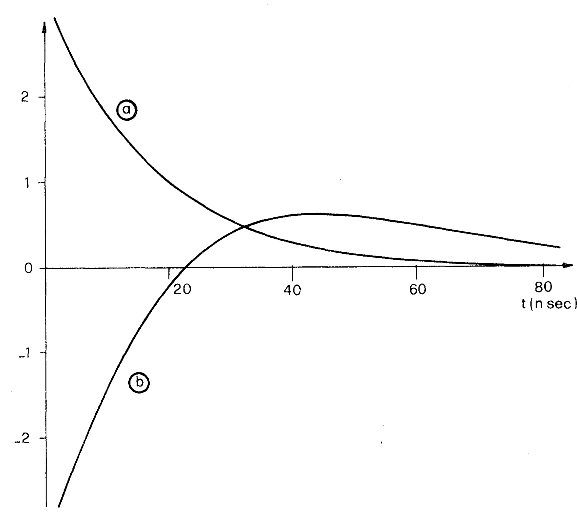


FIG. 1. Various contributions to the amplified intensity of the signal field vs time. The material parameters are those of liquid $CCl_4$ (Ref. 7); $\alpha=0.06\ \text{cm}^{-1}$, interaction length = 0.3 cm, and laser intensity $P_L=1.6\times10^{15}$ W/cm$^2$. Curves a and b show the behavior of the $|C_1|^2\exp(-\Gamma_R t)$ and $2\text{Re}[C_0C_1^*\exp(-\frac{1}{2}\Gamma_R t)]$ terms, respectively. The other contributions to the intensity are not to scale. Here, $C_0$ indicates the amplitude of the stationary term.

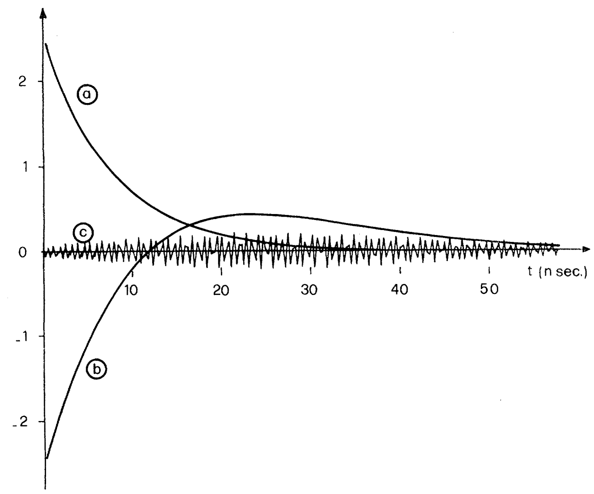


FIG. 2. Contributions to the amplified intensity using liquid-$CS_2$ parameters (Ref. 1), with the same values for $\alpha$, $P_L$, and interaction length as for Fig. 1. Curve c shows the beating between the Brillouin and the stationary and Rayleigh components.

Rother, who separated the contribution of the Stokes and Rayleigh poles, can adequately describe the transient regime.

In Fig. 2 the results obtained using the liquid-$CS_2$ parameters are reported. The ratio $\Gamma_B/\Gamma_R$ is now $\simeq 1.21$, and the beating between the Stokes component and the stationary and Rayleigh components is not negligible during all the transient regime. The beating occurs with a frequency modulation equal to $\omega_B$.

## III. CONCLUSIONS

In conclusion, we have derived an expression for the Stokes field amplified by the interaction with a pump field via thermal fluctuations, which seems to be easily handled both in the steady state and in the transient regime. The solution is valid for arbitrary interaction length, extending the range of validity of Pohl and Kaiser's results. Furthermore, the method used allows the description of the amplification of a signal field interacting with a strong field for any frequency difference between the two waves.

[1]D. Pohl and W. Kaiser, Phys. Rev. B 1, 31 (1970).
[2]D. Pohl, M. Mayer, and W. Kaiser, Phys. Rev. Lett. 20, 366 (1968).
[3]D. Pohl, I. Reinhold, and W. Kaiser, Phys. Rev. Lett. 20, 1141 (1968).
[4]M. Maier, Phys. Rev. 166, 113 (1968).
[5]W. Rother, D. Pohl, and W. Kaiser, Phys. Rev. Lett. 22, 915 (1969).
[6]A. Laubereau, W. English, and W. Kaiser, IEEE J. Quantum Electron. QE-5, 410 (1969).
[7]S. S. Rangnekar and R. H. Enns, Can. J. Phys. 49, 2307 (1971).
[8]I. P. Batra and R. H. Enns, Can. J. Phys. 47, 1283, 2265 (1969); Phys. Lett. 28A, 591 (1969).
[9]R. H. Enns and S. S. Rangnekar, Phys. Lett. 34A, 249 (1971).
[10]W. Rother, Z. Naturforsch. A 25, 1120 (1970).
[11]W. Rother, H. Meyer, and W. Kaiser, Z. Naturforsch. A 25, 1136 (1970).
[12]L. Landau and E. Lifschitz, *Électrodynamique des milieux continus* (Mir, Moscow, 1969), p. 499.
[13]K. Darée and W. Kaiser, Phys. Rev. Lett. 26, 816 (1971).