PHYSICAL REVIEW A VOLUME 29, NUMBER 5 MAY 1984

Sideband interactions in homogeneously broadened saturable absorbers

H. L. Fragnito
Instituto de Física, Universidade Estadual de Campinas, Campinas 13100, São Paulo, Brazil

E. Palange
Istituto di Fisica, Universitá degli Studi di Roma, I-00100 Roma, Italy

C. H. Brito Cruz
Instituto de Física, Universidade Estadual de Campinas, Campinas 13100, São Paulo Brazil

F. DeMartini
Istituto de Fisica, Universitá degli Studi di Roma, I-00100 Roma, Italy
(Received 4 May 1983)

A theoretical study of amplitude (AM) and frequency (FM) modulation components of an optical wave interacting resonantly with a homogeneously broadened two-level system is presented. It is shown that the AM component is usually less attenuated than the FM component; however, under conditions which depend on the modulation frequency, relaxation times, and the saturation parameter, the reverse may occur. In strongly saturated media the AM component can be amplified, a process limited by the depletion of the carrier intensity as the wave propagates in the absorber. The two- and three-wave cases of unidirectional saturation spectroscopy are considered and the effects of depletion are discussed. In the two-wave (single-probe) case a third wave is generated which substantially modifies the absorption profile for the probe. An analysis is included of the sideband evolution in a laser with an intracavity saturable absorber. By analyzing the problem in the frequency domain, some new insights concerning the role of the saturable absorber in passively mode-locked lasers are presented.

I. INTRODUCTION

When a modulated light wave resonantly interacts with a two-level system, the population difference between these levels can follow the modulation if it involves frequencies which are not much greater than the inverse of the relaxation times of the system. At high intensities, when saturation is manifested, the induced nonlinear modulated polarization can significantly depart from that which is expected just from the saturation effect. The interaction is then better described in terms of frequency mixing between the Fourier components of the light wave: Each sideband (SB) interacts with the carrier to force a modulation in the population difference at the beat frequency; the modulated part of the population difference then interacts with the carrier to drive a coherent contribution to the SB polarization . As a result, the SB absorption coefficient can change dramatically and may even reverse its sign which, in a saturable absorber (SA) implies SB amplifcation[1] or amplification of slow intensity fluctuations.[2] SB amplification was first observed by Senitzky *et al.*[1] using millimeter waves, and later observed by Gordon *et al.*[3] in the infrared.

Frequency mixing of this type occurs in unidirectional saturation spectroscopy.[4–6] Here a weak probe wave (frequency ω_1) propagates collinearly with a strong or saturator wave (frequency ω_0) in a resonant medium. The population difference becomes modulated at the beat frequency $|\omega_1-\omega_0|$ modifying the absorption profile for the probe. A homogeneously broadened SA exhibits a profile characteristic of hole burning,[7,8] with "holes" which may penetrate the region of negative absorption leading to probe amplification.[9–12] In addition, a third wave at the "image band" frequency $\omega_{-1}=2\omega_0-\omega_1$ is generated in the medium and, eventually, as the waves propagate, the presence of this third wave can no longer be ignored since it interacts with both the carrier and the population pulsations to give an additional contribution to the probe. Thus, the absorption for the probe depends on the sample length; a dependence which is more pronounced for optically thick samples. In another configuration of unidirectional saturation spectroscopy, the third wave is initially present as a sideband of a pure amplitude modulated (AM) or pure frequency modulated (FM) field. This is the three-wave or two-probe case, where the absorption coefficient for the probes depends on the type of modulation.[4–6]

Physically, one can understand SB generation and amplification with a simple argument. The transmittance of each slab of a saturable medium depends on the intensity of the wave; thus, the transmittance for a modulated wave is also modulated. A saturable absorber then acts as a "modulator"[5] which produces further modulation on the wave and puts sidebands on both sides of the saturator carrier. One can also interpret SB interactions in terms of four-wave mixing,[13] transitions among dressed states[5,13] or still in terms of Raman scattering with the SB's being

Stokes and anti-Stokes waves.

SB and probe amplification have been studied theoretically using several approaches such as the quasimonochromatic wave approximation,[1] Bloch equations,[2] perturbation theory,[14] the dressed atom model,[15] and strong signal laser theory.[4–6] Sargent III (Ref. 6) has reviewed the collinear cases of saturation spectroscopy in both homogeneously and inhomogeneously broadened two-level systems. Boyd *et al.*[13] considered noncollinear propagation in homogeneously broadened systems. In these works, however, the problem of the propagation of the probes in the saturable absorber has been relagated or considered without taking into account the depletion of the saturator wave.[13] As we shall show, when SB amplification and generation are significant, the absorption from the carrier cannot be neglecteted and depletion is important in both modifying the absorption profile for the probe or probes, and limiting the maximum amplification factor for the sidebands with a given initial carrier intensity.

In the present paper we consider a modulated wave composed of a strong carrier (frequency ω_0, amplitude E_0) and two weak sidebands (frequencies $\omega_{\pm 1}=\omega_0 \pm \varpi$, amplitudes $E_{\pm 1} \ll E_0$) interacting in resonance with a homogeneously broadened saturable absorber. The SB amplification conditions and the propagation of the three Fourier components are studied. We assume that the depletion of the carrier is due only to absorption; the transfer of energy from this component to the sidebands (and vice versa) is neglected in the propagation equation for the pump since it contributes with terms of higher order in $E_{\pm 1}/E_0$. In our analysis we allow for arbitrary phases and amplitudes among the SB's (but retain the condition that $E_{\pm 1} \ll E_0$); we thus include the cases of pure AM and pure FM modulations, as well as other intermediate cases and, in particular, that of a single SB, where we study the evolution of the single-probe into the two-probe case by image band generation as the waves propagate in the absorber. It is shown that for slow modulation the resultant field approaches—but strictly never reaches—a state of pure amplitude modulation.

We consider in greater detail the case of the resonant carrier where the response of the medium to the sidebands is symmetric and the AM and FM components of the field evolve independently, a fact that simplifies the analysis. The extension to more general cases such as out of resonance carrier and inhomogeneously broadening is straightforward.

The paper is organized as follows. In Sec. II we derive the coupled equations which govern the steady-state interactions of any number of equidistant Fourier components of a light field with a homogeneously broadened two-level system. These formulas do not involve small perturbation approximations and are thus valid for any value of the Fourier amplitudes. In Sec. III we apply these equations to the case of two weak SB's and a strong carrier. The dependence of the SB's absorption coefficient on the beat frequency and saturation parameter and the conditions for SB amplification are then studied. In order to show explicitly the effects of depletion on the SB's evolution in a saturable absorber, the propagation equations for the three waves are solved numerically in the limit of slow modulation (Sec. IV) and, finally, in Sec. V we analyze the evolution of three running modes of a laser with an intracavity SA cell. Though the model here used for the laser is oversimplified, we find that, passing to the frequency domain, one obtains useful insights concerning the role played by the SA in the passive model locking of lasers.

II. GENERAL EQUATIONS

The equations for the macroscopic polarization P associated with the transition, and the population difference per unit volume ΔN of a two-level system interacting with a radiation field E at optical frequencies are[16]

$$\frac{\partial^2 P}{\partial t^2}+\frac{2}{T_2}\frac{\partial P}{\partial t}+\Omega^2 P=\frac{2\mu^2\Omega E}{3\hbar}\Delta N L \ , \tag{2.1}$$

$$\frac{\partial \Delta N}{\partial t}+\frac{\Delta N-\Delta N^e}{T_1}=-\frac{2E}{\hbar\Omega}\frac{\partial P}{\partial t} \ , \tag{2.2}$$

where T_1 and T_2 are the longitudinal (recovery time) and transversal (inverse of linewidth) relaxation times, respectively; Ω is the central frequency of the homogeneously broadened absorption line; μ is the electric dipole moment of the transition; ΔN^e is the equilibrium value of ΔN (i.e., in the absence of the field) and $L=(\eta^2+2)^2/9$ is a Lorentz local field correction factor (η is a background index of refraction).

We consider a radiation field composed by a superposition of linearly polarized plane monochromatic waves copropagating in the z direction:

$$E(z,t)=\sum_n E_n \cos(\omega_n t-k_n z+\phi_n) \ . \tag{2.3}$$

Here ω_n and ϕ_n are frequency (rad/sec) and phase, respectively, of the nth Fourier component, and $k_n=\eta\omega_n/c$ (c is the speed of light).

We shall assume that the frequency separations are all multiples of the quantity ϖ:

$$\omega_{n+1}-\omega_n=\varpi, \quad n=0,\pm 1,\pm 2,\dots \ . \tag{2.4}$$

This form of the field is adequate to describe a multimode laser or a modulated wave with beat frequency ϖ. We express the polarization as

$$P(z,t)=\tfrac{1}{2}\sum_n P_n \exp[i(\omega_n t-k_n z+\phi_n)]+\text{c.c.} \tag{2.5}$$

On the right-hand side of Eq. (2.2) there will appear terms such as

$$\frac{\hbar\omega_n}{2\Omega}P_n^* E_m \exp[i(\omega_m-\omega_n)t-i(k_m-k_n)z+i(\phi_m-\phi_n)]+\text{c.c.} \ ,$$

which force a modulation in the population difference at the beating frequency $|m-n|\varpi$. It is convenient to separate the modulated and nonmodulated parts of ΔN:

$$\Delta N=N_0+\left[\tfrac{1}{2}\sum_{q\ge 1} N_q \exp(iq\varpi t)+\text{c.c.}\right] . \tag{2.6}$$

The steady-state solutions are then

$$N_0=\Delta N^e-i\frac{T_1}{2\hbar\Omega}\sum_n(\tilde{\tilde{E}}^*_n\tilde{\tilde{P}}_n-\tilde{\tilde{E}}_n\tilde{\tilde{P}}^*_n)\omega_n \tag{2.7}$$

$$N_q=\frac{iT_1}{\hbar\Omega(1+iq\varpi T_1)}\sum_n(\tilde{\tilde{E}}_n\tilde{\tilde{P}}^*_{n-q}\omega_{n-q}-\tilde{\tilde{E}}^*_n\tilde{\tilde{P}}_{n+q}\omega_{n+q}) , \tag{2.8}$$

$$\tilde{\tilde{P}}_n=-\frac{i\mu^2T_2L}{3\hbar a_n}\left[\tilde{\tilde{E}}_nN_0+\tfrac{1}{2}\sum_q(\tilde{\tilde{E}}_{n+q}N^*_q+\tilde{\tilde{E}}_{n-q}N_q)\right] , \tag{2.9}$$

where

$$a_n=(\Omega^2-\omega_n^2+2i\omega_n/T_2)(T_2/i2\Omega)\simeq 1-iT_2(\Omega-\omega_n) \tag{2.10}$$

$$\tilde{\tilde{P}}_n=P_n\exp(-ik_nz+i\phi_n) ,$$
$$\tilde{\tilde{E}}_n=E_n\exp(-ik_nz+i\phi_n) . \tag{2.11}$$

The propagation law for the nth mode is, from the Maxwell equations,

$$\frac{\partial}{\partial z}(E_ne^{i\phi_n})=-\frac{i\omega_n}{2\eta\epsilon_0c}P_ne^{i\phi_n} , \tag{2.12}$$

where ϵ_0 is the dielectric permissivity of vacuum. From this equation we obtain the absorption coefficient for the nth mode

$$\alpha(\omega_n)\equiv-\frac{1}{E_n^2}\frac{\partial E_n^2}{\partial z}=\frac{i\omega_n}{2\eta\epsilon_0cE_n}(P_n-P_n^*) . \tag{2.13}$$

This completes the set of coupled mode equations which describes the evolution of the modes as the wave propagates through the resonant medium in the steady state. The population pulsations at the frequency $q\varpi$ combine with the fields at ω_{n-q} and ω_{n+q} to give a contribution to the polarization at ω_n, but all modes contribute to N_q so that all modes are coupled in an intricate way.

Neglecting the population pulsations one obtains

$$P_n=-\frac{i\mu^2T_2L\,\Delta N^eE_n}{3\hbar a_n\left[1+(\mu^2LT_1T_2/3\hbar^2\Omega^2)\sum_n E_n^2\omega_n^2/|a_n|^2\right]}$$

and in that case the polarization at ω_n depends on the amplitudes of all modes but not on the relative phases $\phi_n-\phi_m$. On the other hand, when the population pulsations are taken into account, the sum term in the rhs of Eq. (2.9) gives a contribution to P_n which explicitly depends on the relative phases among the modes. Following Sargent III (Ref. 6) we shall call these two types of contributions incoherent (phase independent, associated to the dc value of the population difference) and coherent (associated to the population pulsations) polarizations. While the incoherent part of the polarization always represents absorption at ω_n (depending naturally on the sign of ΔN^e), the coherent contribution can lead to an increase or decrease of the absorption and even amplification for that mode, depending on the amplitudes and phases of all the modes.

Some of the modes can actually be amplified in the absorber. However, the energy is not supplied by the medium. The power absorbed per unit volume is $E\partial P/\partial t$, and for the coherent part of the polarization this quantity averages to zero in a time long compared with the beat periods. Thus, the amplification of the favored modes is at the expense of the increased attenuation of others. If Eq. (2.3) represents a light pulse (or a train of light pulses) then the parametric processes described by the coherent polarizations have the effect of redistributing the available energy among the modes. In the time domain this results in pulse reshaping.

We also note that even if a given mode is initially absent (for example, $E_n=0$ at $z=0$) it will be created and will grow as the wave propagates in the absorber, driven by the coherent part of the polarization at ω_n. Furthermore, the induced polarization at ω_n has a definite phase; thus, the coherent effects tend to broaden the spectra and to correlate the phases which, in the time domain, means pulse compression. This action of the SA as a mode expander[17] is precisely the key for understanding, from the frequency domain point of view, the generation of ultrashort pulses in passively mode-locked lasers. In principle, only two adjacent modes are necessary to generate all the others in the SA. During the transient response of a two-level system, however, a single mode can evolve into a modulated wave, with the generated sidebands beating at the Rabi frequency. This last effect (optical nutation) does not occur in the steady state and will not be considered here, but one can expect some type of resonance when the interacting modes beat at a frequency close to the Rabi flopping frequency.[13]

The relative importance of the modulated to the nonmodulated field-dependent parts of the population difference can be expressed by the quotients

$$|\tfrac{1}{2}N_q/(N_0-\Delta N^e)| .$$

If all the modes under the wings of the resonance curve have equal amplitudes and phases, then these quotients are of the order of

$$1/[1+(q\varpi T_1)^2]^{1/2} .$$

Thus, pulsations—and with them, the coherent effects—can be neglected only if $\varpi T_1\gg 1$. In a laser cavity with mirror separation of 1 m, the longitudinal modes beat at about 9.4×10^8 rad/sec, and if these modes interact in a SA with a recovery time of 10 psec (as is typical in dyes used for passive mode locking of solid-state lasers) we have

$$1/[1+(q\varpi T_1)^2]^{1/2}=99.99\%$$

for $q=1$, and 50% for $q=184$. This means not only that the coherent effects are important, but also that the coupling between modes n and $n\pm q$ is as important as the coupling between adjacent modes (n and $n\pm1$), where q

can have values surprisingly high. Hundreds, and even thousands[18] of modes couple simultaneously in pulsed—passively mode-locked—solid-state lasers; thus, when applied to these devices, frequency domain theories, where only a few oscillating modes[19] or only adjacent coupling are considered,[20–23] are not expected to give accurate descriptions of the pulse narrowing effect.

III. SIDEBAND INTERACTIONS

In this section we consider a field composed by a strong carrier at a frequency ω_0 and two weak sidebands at frequencies $\omega_{\pm1}=\omega_0\pm\bar{\omega}$. The mode coupled equations of Sec. II, solved to all orders in the carrier amplitude but to first order in the SB's amplitudes $E_{\pm1}$, give

$$N_0=\Delta N^e/(1+\gamma)\ , \tag{3.1}$$

$$N_1=2\Delta N^e[(\chi_1^{(3)}/\chi_1)\tilde{\tilde{E}}{}_0^*\tilde{\tilde{E}}_1+(\chi_{-1}^{(3)*}/\chi_{-1}^*)\tilde{\tilde{E}}_0\tilde{\tilde{E}}{}_{-1}^*]\ , \tag{3.2}$$

$$\tilde{\tilde{P}}_0=\epsilon_0\chi_0N_0\tilde{\tilde{E}}_0/\Delta N^e\ , \tag{3.3}$$

$$\tilde{\tilde{P}}_1=(\epsilon_0\chi_1/\Delta N^e)(N_0\tilde{\tilde{E}}_1+\tfrac{1}{2}N_1\tilde{\tilde{E}}_0)\ , \tag{3.4}$$

$$\tilde{\tilde{P}}_{-1}=(\epsilon_0\chi_{-1}/\Delta N^e)(N_0\tilde{\tilde{E}}_{-1}+\tfrac{1}{2}N_1^*\tilde{\tilde{E}}_0)\ , \tag{3.5}$$

where we introduced the linear susceptibilities

$$\chi_n=-i\mu^2T_2L\Delta N^e/(3\epsilon_0\hbar a_n)\ , \tag{3.6}$$

in terms of which we define the saturation parameter

$$\gamma=iT_1\omega_0E_0^2\epsilon_0(\chi_0-\chi_0^*)/(2\hbar\Omega\Delta N^e)\ , \tag{3.7}$$

and the "third-order susceptibility"

$$\chi_1^{(3)}=\frac{\chi_1(\omega_1\chi_1-\omega_0\chi_0^*)/(1+\gamma)E_s^2}{\gamma(\omega_{-1}\chi_{-1}^*-\omega_1\chi_1)+\omega_0(\chi_0^*-\chi_0)(1+i\omega_1T_1-i\omega_0T_1)}\ . \tag{3.8}$$

$\chi_{-1}^{(3)}$ is obtained from this expression by changing the sign of the subindices of ω_n and χ_n. The saturation field is

$$E_s=E_0/\sqrt{\gamma}\ . \tag{3.9}$$

The coherent contribution to the carrier polarization does not appear in Eq. (3.3) since it is of second order in $E_{\pm1}$. That contribution is necessary to preserve the energy balance of the parametric process. Thus, in the above equations energy is not conserved, the small energy unbalance being of the order of $(E_{\pm1}/E_0)^2$. In this approximation the carrier propagates as if it solely saturates the medium. As a consequence of the homogeneous broadening, the carrier also saturates the response of the medium to the sidebands. The incoherent polarization can be described in terms of an effective first-order susceptibility

$$\chi_n^{(1)}=\chi_n/(1+\gamma)\ . \tag{3.10}$$

The term "susceptibility" applied to $\chi^{(1)}$ and $\chi^{(3)}$ is used here in an extended sense, for they are quantities which depend on the intensity of the carrier.

The polarizations at the sideband frequencies are given by

$$\tilde{\tilde{P}}_{\pm1}=\epsilon_0\chi_{\pm1}^{(1)}\tilde{\tilde{E}}_{\pm1}+\epsilon_0\chi_{\pm1}^{(3)}\tilde{\tilde{E}}_0\tilde{\tilde{E}}{}_0^*\tilde{\tilde{E}}_{\pm1}+\epsilon_0\chi_{\mp1}^{(3)*}(\chi_{\pm1}/\chi_{\mp1}^*)\tilde{\tilde{E}}_0\tilde{\tilde{E}}_0\tilde{\tilde{E}}{}_{\mp1}^*\ . \tag{3.11}$$

The coherent contributions represent Raman-type frequency mixing processes in which the carrier is scattered at the SB's frequencies by the population pulsations. The second term in Eq. (3.11) represents a self-phase matched process, while the third term depends on the phase mismatch $\Delta\phi$ defined below. These two coherent contributions are not, in general, in phase, and they interfere in sucy a way that the absorption coefficient for each SB may either increase or decrease with respect to that given by the imaginary part of $\chi_1^{(1)}$, depending on the frequencies and amplitudes of the three Fourier components and on the phase mismatch

$$\Delta\phi=2\phi_0-(\phi_1+\phi_{-1})\ . \tag{3.12}$$

The absorption coefficients for the three Fourier components are readily obtained after substituting (3.3) and (3.11) into (2.13). To investigate the conditions for SB amplification we shall assume the carrier to be in resonance $|\omega_0-\Omega|\ll 1/T_2$. For $\omega_0=\Omega$, χ_0 is purely imaginary and as the wave propagates in the absorber the saturation parameter decreases according to [see Eq. (2.13)]

$$\partial\gamma/\partial z=-\alpha_0\gamma/(1+\gamma)\ , \tag{3.13}$$

where $\alpha_0=i\omega_0\chi_0/\eta c$ is the linear absorption coefficient at resonance.

For $\bar{\omega}\ll\omega_0=\Omega$ Eq. (3.11) reduces to

$$P_{\pm1}=\epsilon_0\chi_{\pm1}^{(1)}E_{\pm1}+\epsilon_0\chi_{\pm1}^{(3)}(E_0^2E_{\pm1}+E_0^2E_{\mp1}e^{i\Delta\phi}) \tag{3.14}$$

and the third-order susceptibility simplifies to

$$\chi_1^{(3)}=-\chi_{-1}^{(3)*}=-\chi_1^{(1)}(1+ir\beta/2)/E_s^2[\gamma+(1+i\beta)(1+ir\beta)]\ , \tag{3.15}$$

where

$$\beta=\bar{\omega}T_1$$

and

$$r=T_2/T_1\ . \tag{3.16}$$

The main features of SB evolution will be better appreciated if we look at the modulation components of the field. Equation (2.3) can for $\langle n\rangle<2$ be recast into the form

$$E(z,t)=E_0'(z,t)\cos[\omega_0t-k_0z+\phi'(z,t)]\ , \tag{3.17}$$

where the slowly varying amplitude and phase are given by

$$E_0'=[(E_0+E_1\cos A_1+E_{-1}\cos A_{-1})^2+(E_1\sin A_1-E_{-1}\sin A_{-1})^2]^{1/2}\ ,$$
$$\phi'=\phi_0+\tan^{-1}\left[\frac{E_1\sin A_1-E_{-1}\sin A_{-1}}{E_0+E_1\cos A_1+E_{-1}\cos A_{-1}}\right]\ , \tag{3.18}$$

where

$$A_{\pm 1}=\varpi(t-\eta z/c)\pm\phi_{\pm 1}\mp\phi_0 .$$

To first order in $E_{\pm 1}/E_0$, (3.17) represents a wave which is harmonically modulated at ϖ both in amplitude (AM) and phase or frequency (FM):

$$E_0'=\tfrac{1}{2}E_0+\tfrac{1}{2}E_{\rm AM}e^{i\varpi(t-\eta z/c)}+\text{c.c.} ,$$
$$E_0\phi'=\tfrac{1}{2}E_0\phi_0-\tfrac{1}{2}iE_{\rm FM}e^{i\varpi(t-\eta z/c)}+\text{c.c.} \qquad (3.18')$$

The (complex) amplitudes of the AM and FM components are

$$E_{\rm AM}=E_1e^{i(\phi_1-\phi_0)}+E_{-1}e^{-i(\phi_{-1}-\phi_0)}$$
$$E_{\rm FM}=E_1e^{i(\phi_1-\phi_0)}-E_{-1}e^{-i(\phi_{-1}-\phi_0)} . \qquad (3.19)$$

From (2.12) and (3.14) we see that the AM and FM components evolve independently following the propagation equations

$$\frac{\partial E_{\rm AM}}{\partial z}=-\frac{i\omega_0}{2\eta c}(\chi_1^{(1)}+2\chi_1^{(3)}E_0^2)E_{\rm AM} ,$$
$$\frac{\partial E_{\rm FM}}{\partial z}=-\frac{i\omega_0}{2\eta c}\chi_1^{(1)}E_{\rm FM} . \qquad (3.20)$$

Thus, the coherent effects act only on the AM component, which may then suffer increased or reduced attenuation (with respect to the FM component) depending on the sign of the imaginary part of $\chi_1^{(3)}$. The real and imaginary parts of $\chi^{(3)}=\chi^{(3)'}+i\chi^{(3)''}$ are

$$\chi_{\pm 1}^{(3)'}=\pm\frac{\alpha_0\eta c}{D\omega_0E_s^2}\beta(2+3r+\gamma r+r^3\beta^2) ,$$
$$\chi_{\pm 1}^{(3)''}=\frac{\alpha_0\eta c}{D\omega_0E_s^2}[2(\gamma+1)+(\gamma r^2-3r)\beta^2-r^3\beta^4] , \qquad (3.21)$$

where

$$D=2(\gamma+1)(1+r^2\beta^2)[(1+\gamma)^2+(1+r^2-2\gamma r)\beta^2+r^2\beta^4] . \qquad (3.22)$$

If $\chi^{(3)''}>0$ the saturable absorber favors the amplitude modulations, i.e., the AM component is less attenuated in the absorber than the FM component. From (3.21) we see that this occurs if

$$\gamma>(r^3\beta^4+3r\beta^2-2)/(r^2\beta^2+2) \qquad (3.23)$$

or, equivalently, if

$$\varpi<(1/\sqrt{2}T_2)\{\gamma r-3+[(\gamma r-3)^2+8r(\gamma+1)]^{1/2}\}^{1/2} . \qquad (3.23')$$

In the short dipole lifetime limit ($r=0$) the AM component is favored regardless of the value of the saturation parameter, and if $r\neq 0$ saturation extends the interval of modulation frequencies where the AM component is favored from

$$\varpi<(1/\sqrt{2}T_2)[(9+8r)^{1/2}-3]^{1/2}$$

to that given in (3.23′). If $\chi^{(1)''}+2\chi^{(3)''}E_0^2>0$ then the AM component (and thus the SB's of an AM wave) are amplified in the medium, i.e., the modulation depth of the field amplitude increases with distance. From (3.21), (3.22), and (3.6) we have

$$\chi^{(1)''}+2\chi^{(3)''}E_0^2=\frac{2\alpha_0\eta c}{D\omega_0}(1+r^2\beta^2)[\gamma^2-1-(1+\gamma r)\beta^2] \qquad (3.24)$$

and the condition for AM amplification comes to be

$$\gamma>\tfrac{1}{2}[r\beta^2+(4+4\beta^2+r^2\beta^4)^{1/2}] . \qquad (3.25)$$

Only if $\gamma>1$ is there a beat frequency interval where AM amplification can occur:

$$\varpi<(1/T_1)[(\gamma^2-1)/(1+\gamma r)]^{1/2} .$$

In unidirectional saturation spectroscopy[6] two cases are of interest: In the first the SB's have equal amplitudes and are phased to give pure amplitude or pure frequency modulation; this is the two-probe or three-wave case. In the second or two-wave case, a single probe is used. Consider first the three-wave case with $E_1=E_{-1}=E_{\rm SB}$ and with arbitrary phase relationship $\Delta\phi$. From (3.14) we have

$$\frac{\partial}{\partial z}(E_{\pm 1}e^{i\phi_{\pm 1}})=-\frac{i\omega_0}{2\eta c}[\chi_{\pm 1}^{(1)}+E_0^2\chi_{\pm 1}^{(3)}(1+e^{i\Delta\phi})]\times E_{\rm SB}e^{i\phi_{\pm 1}} . \qquad (3.26)$$

For $\Delta\phi=\pm\pi$ we have pure FM ($E_{\rm AM}=0$), the coherent contributions interfere destructively, and the three waves propagate as if there were no interactions between them. If $\Delta\phi=0,\pm 2\pi$ we have pure AM ($E_{\rm FM}=0$), and the coherent contributions interfere constructively minimizing the absorption coefficient for the SB's:

$$\alpha_{\rm SB}(\text{AM})=-\frac{\omega_0}{\eta c}(\chi^{(1)''}+2E_0^2\chi^{(3)''}) , \qquad (3.27)$$

where the quantity in parentheses was given in (3.24). If (3.25) is satisfied the SB's are amplified.

It is interesting to observe that since the SB's suffer equal but opposite dispersion ($\chi_1^{(1)'}=-\chi_{-1}^{(1)'}$ and $\chi_1^{(3)'}=-\chi_{-1}^{(3)'}$), an initially pure AM or FM wave conserves its character, i.e., the phase relationship is maintained. From (3.26) we have

$$\frac{\partial\Delta\phi}{\partial z}=-\frac{\omega_0}{\eta c}E_0^2\chi^{(3)''}\sin\Delta\phi .$$

Thus, if $\Delta\phi=0,2\pi$ (AM), or if $\Delta\phi=\pm\pi$ (FM), then $\Delta\phi$ is a constant of propagation. Furthermore, for other values of $\Delta\phi$ the SB's attenuate at different rates, and it may occur that only one of the SB's experiences gain. To illustrate this fact consider the case $r=0$ and $\Delta\phi=\pi/2$; from (3.26) we obtain

$$\alpha_{\pm 1}=\alpha_0(\beta^2+\gamma+1\mp\gamma\beta)/(1+\gamma)(\beta^2+\gamma^2+2\gamma+1)$$

and if $\gamma(\beta-1)>\beta^2+1$ then one of the sidebands is amplified while the other is attenuated.

Consider now the single probe case. At $z=0$ only one

SB is present, for example, $E_1 \neq 0$ and $E_{-1}=0$. The absorption coefficient for the probe is given (at $z=0$) by

$$\alpha_1 = -\frac{\omega_0}{\eta c}(\chi^{(1)''}+E_0^2\chi^{(3)''}) \tag{3.28}$$

and has been discussed by several authors.[3–14] We shall concern ourselves here with another aspect of the problem. From (3.19) we have, at $z=0$, $E_{AM}=E_{FM}$; however, as the waves propagate, these modulation components suffer different attenuations, and at $z\neq 0$ the probe absorption coefficient is no longer given by (3.28). It is clear from (3.14) that even when $E_{-1}(0)=0$, there is an induced polarization at the image band frequency $\omega_{-1}=2\omega_0-\omega_1$:

$$P_{-1}=\epsilon_0\chi_{-1}^{(3)}E_0^2E_1e^{i(2\phi_0-\phi_1)}$$

which is responsible for the generation of the image band at a rate given by

$$\frac{\partial}{\partial z}(E_{-1}e^{i\phi_{-1}}) = -\frac{i\omega_0}{2\eta c}E_0^2\chi_{-1}^{(3)}E_1e^{i(2\phi_0-\phi_1)} . \tag{3.29}$$

In order to see what the effects are of this third wave let us analyze them in the slow modulation limit ($\beta \rightarrow 0$) where there is no dispersion. The probe is initially attenuated with an absorption coefficient $\alpha_0/(1+\gamma)^2$, the carrier with $\alpha_0/(1+\gamma)$, and the image band grows at a rate

$$\frac{\partial E_{-1}}{\partial z} = \frac{\alpha_0\gamma}{2(1+\gamma)^2}E_1$$

and phase $\phi_{-1}=2\phi_0-\phi_1$, which is proper for amplitude modulation. After a propagation distance of the order of

$$2(1+\gamma)^2/\alpha_0\gamma ,$$

the amplitude of the generated SB reaches the order of magnitude of that of the probe and the total field approaches that of a pure AM wave. If the carrier intensity has not been depleted too much so that the saturation parameter is still greater than one, then both SB's are amplified (we shall see in Sec. IV that this occurs if at $z=0$, $\gamma > 7$). This shows that there is a substantial modification of the probe absorption coefficient as the waves propagate.

In general, beyond the slow modulation limit, the phase of the generated band will be different from that for AM, and it will change with distance. If (3.23) is not satisfied, for example, the FM component is less attenuated than the AM one, and the two waves evolve into three waves phased for FM. If (3.23′) is satisfied but

$$\bar{\omega} > [(9+8r)^{1/2}-3]^{1/2}/\sqrt{2}T_2 ,$$

then the waves will tend initially to AM but, after some distance, due to depletion, the saturation parameter will not satisfy (3.23) and the field will tend to a FM wave. One can see that the field may approach a state of pure AM or FM modulation, but it will never reach such states. This is because a pure AM or FM character is preserved during propagation; thus, if at some point one of the modulation components vanishes, it must also vanish for all subsequent and precedent positions. Furthermore, there is a limit for the relative growth of the image band, i.e., the ratio

$$E_{FM}(z)/E_{AM}(z)$$

does not tend to zero. One can solve Eq. (3.20) as a function of γ and, using (3.13), obtain

$$E_{AM}(z)=E_{AM}(0)\left[\frac{\gamma(z)}{\gamma_0}\right]^{\psi/2} \times \left[\frac{1+i\beta+\psi\gamma(z)}{1+i\beta+\psi\gamma_0}\right]^{(1+\psi)/2} , \tag{3.30}$$

$$E_{FM}(z)=E_{FM}(0)\left[\frac{\gamma(z)}{\gamma_0}\right]^{\psi/2} ,$$

where $\psi=1/(1+ir\beta)$ and γ_0 is the saturation parameter at $z=0$. From this equation we have

$$E_{FM}(z)/E_{AM}(z)=[E_{FM}(0)/E_{AM}(0)] \times \left[\frac{1+i\beta+\psi\gamma(z)}{1+i\beta+\psi\gamma_0}\right]^{(1+\psi)/2} . \tag{3.31}$$

For $z\rightarrow\infty$ we have $\gamma(z)\rightarrow 0$ so that the ratio (3.31) tends to a nonvanishing value. For example, in the slow modulation case considered by Senitzky *et al.*,[1] $\beta=0$ and

$$\lim_{z\rightarrow\infty}[E_{AM}(z)/E_{FM}(z)]=[E_{AM}(0)/E_{FM}(0)](1+\gamma_0) .$$

In the single probe case where $E_{AM}(0)=E_{FM}(0)$, the AM component becomes $(1+\gamma_0)$ times the FM component for $z\rightarrow\infty$.

The probe absorption profile in the two-wave case is thus a complicated function of the sample length; a dependence which is more pronounced for optically thick samples where depletion is important. Unfortunately, the easier it is to detect absorption differences, the higher the effects of depletion are; this is because the probe absorption coefficient is proportional to α_0. In the three-wave case the z dependence is only through the saturation parameter so that the probes absorption coefficient is always given by (3.27), and (3.24). An additional obvious advantage of the three-wave configuration is that only one laser is required. Both methods give the same information about the two-level system. A disadvantage of the three-wave configuration may arise in systems with short relaxation times where one would need to modulate the laser at rather high frequencies.

Similar effects due to population modulations also occur in the counter propagation configuration of saturation spectroscopy, where the probe and saturator waves propagate in opposite directions but have the same frequency. The population difference is spatially modulated and the saturated wave is back scattered to give a coherent contribution to the probe. For a review on this subject see (Ref. 24) and for the analogies between temporal and spatial modulations see (Ref. 5).

IV. SIDEBAND PROPAGATION

The propagation equations for the carrier and the two side bands are coupled differential equations with coefficients which are functions of distance to account for depletion. Some remarks on the evolution of the SB's have been discussed in Sec. III. Here we shall solve these equations numerically for some representative cases in the slow modulation limit ($\beta=0$). We shall consider SB's phased for AM ($\Delta\phi=0$); in the $\beta=0$ limit this phase relationship is a constant of propagation. If an arbitrary phase mismatch would be considered, an additional equation for $\Delta\phi$ should be included. We emphasize that, in spite of our assumption ($\Delta\phi=0$), the total field may have a FM component since this vanishes only if $\Delta\phi=0$ *and* $E_1=E_{-1}$.

For $\Delta\phi=0$ the propagation equations for the SB's are

$$\frac{\partial E_{\pm 1}}{\partial z}=-\frac{\alpha_0}{2(1+\gamma)^2}(E_{\pm 1}-\gamma E_{\mp 1}) \tag{4.1}$$

and for the carrier or, equivalently, for the saturation parameter we have Eq. (3.13). After a redefinition of the time origin we can write the AM and FM components as real amplitudes,

$$\begin{aligned} E_{\rm AM}&=E_1+E_{-1}\ ,\\ E_{\rm FM}&=E_1-E_{-1}\ , \end{aligned} \tag{4.2}$$

which evolve according to

$$\begin{aligned} \frac{\partial E_{\rm AM}}{\partial z}&=\frac{\alpha_0(\gamma-1)}{2(\gamma+1)^2}E_{\rm AM}\ ,\\ \frac{\partial E_{\rm FM}}{\partial z}&=-\frac{\alpha_0}{2(\gamma+1)}E_{\rm FM}\ . \end{aligned} \tag{4.3}$$

A pure FM wave can be represented in this formalism by allowing negative values for the amplitude of one of the SB's (thus having $E_{\rm AM}=0$). This case however, does not show interesting features since the coherent contributions vanish.

According to (4.3) the FM component attenuates with the same absorption coefficient as the carrier, while if $\gamma>1$ the AM component is amplified. Significant amplification occurs along an interaction path of the order of

$$2(\gamma+1)^2/\alpha_0(\gamma-1)\ .$$

We have also seen in Sec. III that the process of SB generation has a characteristic length

$$2(\gamma+1)^2/\alpha_0\gamma\ .$$

At these propagation distances in the absorber the depletion of the carrier intensity is not negligible and, consequently, the exponential (Beer's) law does not apply. We can express the solutions of (4.3) in terms of the saturation parameter $\gamma=\gamma(z)$ and the initial values $[\gamma_0=\gamma(0)]$. From (3.30)

$$\begin{aligned} E_{\rm AM}(z)&=E_{\rm AM}(0)\sqrt{\gamma/\gamma_0}(\gamma_0+1)/(\gamma+1)\ ,\\ E_{\rm FM}(z)&=E_{\rm FM}(0)\sqrt{\gamma/\gamma_0}\ . \end{aligned} \tag{4.4}$$

Integration of Eq. (3.13) gives an implicit equation for $\gamma(z)$:

$$\gamma e^{\gamma}=\gamma_0 e^{\gamma_0}e^{-\alpha_0 z}\ . \tag{4.5}$$

With the help of these equations the general behavior of the SB's evolution can be analyzed. We shall refer to Figs. 1 and 2 and discuss separately the cases of AM and SB generation.

AM waves. If, initially, $E_{-1}=E_1=E_{\rm SB}$, then the FM

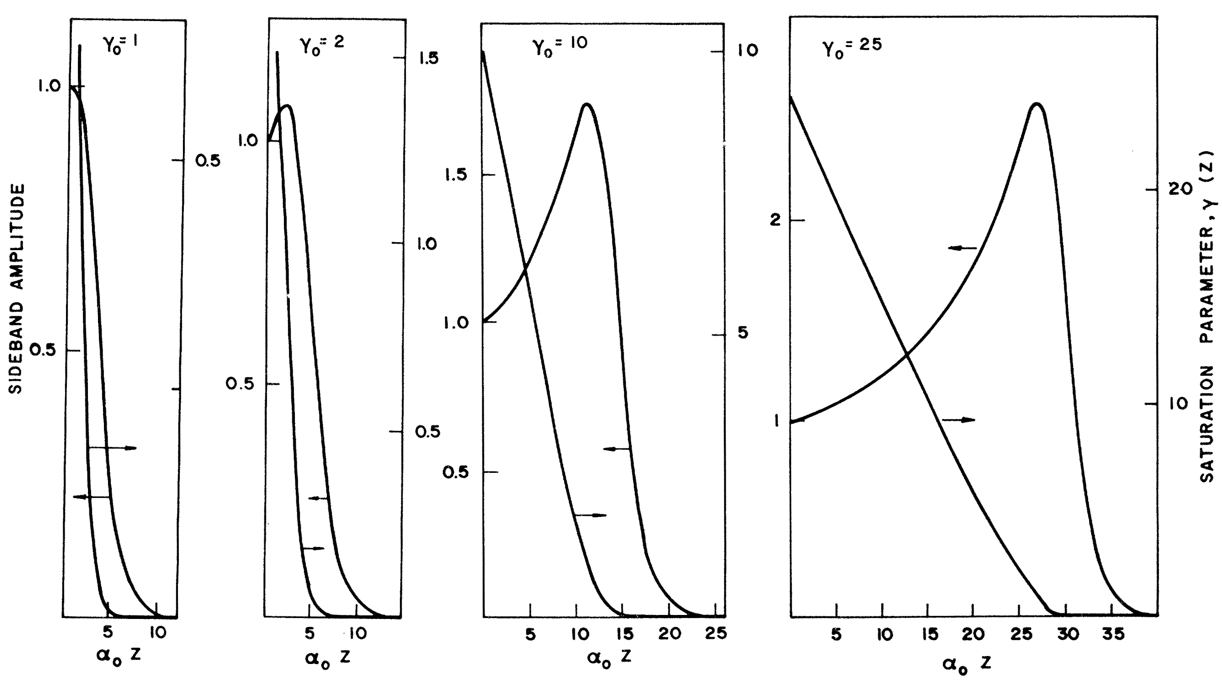


FIG. 1. Propagation of an AM wave. Arrows indicate scale readouts for SB amplitude (←) and saturation parameter (→).

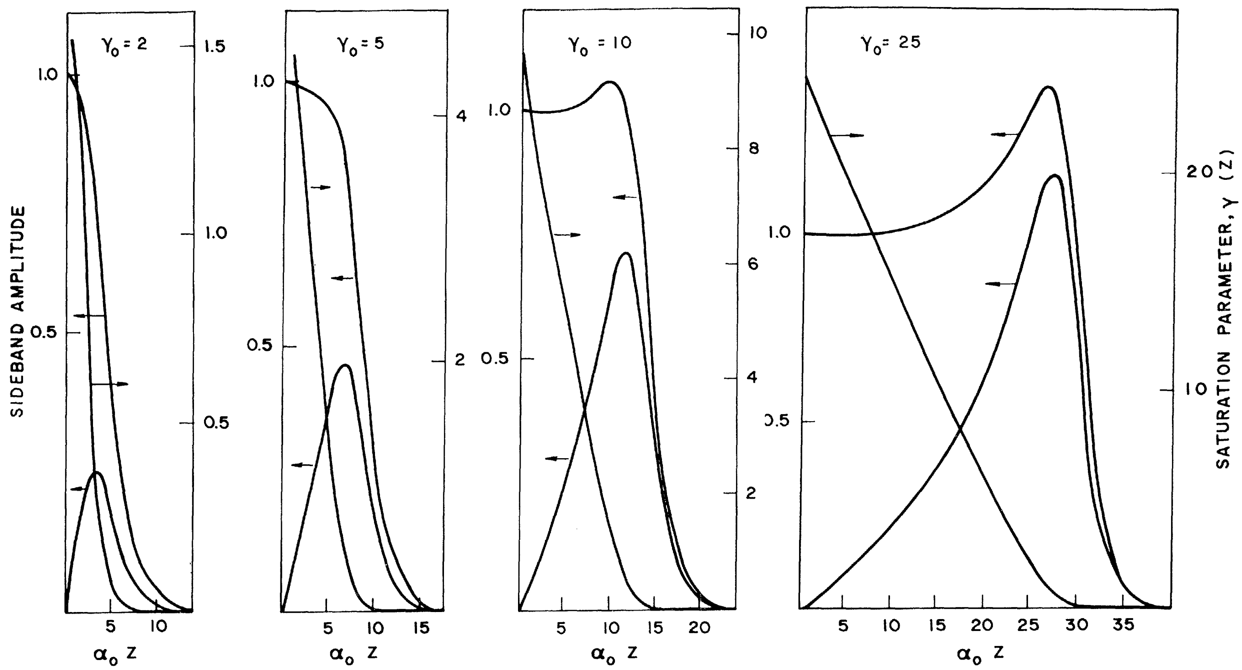


FIG. 2. Sideband generation (curves starting at the origin) and evolution of the initially present SB.

component vanishes for all z and the wave continues to be pure AM. (This is also true even for rapid modulations, as was shown in Sec. III.) In Fig.1 we plotted the SB amplitudes normalized to their initial value [i.e., $E_{SB}(z)/E_{SB}(0)$] while for the carrier we plotted $\gamma(z)$, which is proportional to $E_0^2(z)$.

The SB's are amplified if $\gamma_0 > 1$ and amplification continues until a point z_0 is reached where $\gamma(z_0)=1$. After that point the SB's are attenuated. From the above equations we deduce

$$z_0=(\gamma_0-1+\ln\gamma_0)/\alpha_0$$

provided that $\gamma_0 > 1$. At this point the SB's reach the maximum amplitude

$$E_{SB}^{max}=E_{SB}(z_0)=E_{SB}(0)(\gamma_0+1)/\sqrt{4\gamma_0}\ .$$

A large value of γ_0 leads to a high amplification factor $E_{SB}^{max}/E_{SB}(0)$ but to a slow rate of growth $(1/E_{SB})(\partial E_{SB}/\partial z)$. This rate of growth is a measure of the rapidity of the SB amplification process and is maximum for $\gamma=3$.

SB generation. Here at $z=0$ only one SB is present and we have, initially, equal AM and FM components: $E_{AM}(0)=E_{FM}(0)$. In Fig. 2 we plotted the evolution of the two sidebands for several values of the initial saturation parameters. Both SB amplitudes are normalized to the initial value of the preexistent band.

The image SB is generated and amplified even for $\gamma < 1$, while the preexistent sideband is initially attenuated; but after some point it can start to grow. This behavior can be understood as follows: The new SB grows at the expenses of the energy of both the carrier and the other SB, so that this last is always initially attenuated; however, as the new SB grows the total field tends to a pure AM wave, and if γ is still greater than one then both SB's can be amplified. From Eqs. (4.1)–(4.5) we can see that this would occur if $\gamma_0 > 7$, and in this case the preexistent SB (probe) attains a minimum at z^+ and a maximum at z^-, where z^+ and z^- are given by

$$\gamma(z^\pm)=\tfrac{1}{2}(\gamma_0-1)\pm\tfrac{1}{2}[(\gamma_0-1)^2-4\gamma_0-8]^{1/2}\ .$$

The amplitude of the generated SB is maximum at a point $z=z'$, when

$$\gamma(z')=-\tfrac{1}{2}(\gamma_0+3)+\tfrac{1}{2}(\gamma_0^2+10\gamma_0+9)^{1/2}\ .$$

The characterization of these extremal points (z_0, $z^\pm$, and z') can be of help in the design of experiments on SB amplification and/or SB generation. For a given initial saturation parameter, one can optimize the product $\alpha_0 l$ (varying the concentration or the SA cell length l) so that SB amplification is maximum and, thus, easier to measure. To be specific, consider experiments with $\gamma_0=3$, then the best choices are as follows: for SB amplification of AM waves $z_0=l$ gives $\alpha_0 l \simeq 3.1$; for SB generation $z'=l$ gives $\alpha_0 l \simeq 4.4$. Probe amplification requires $\gamma_0 > 7$; thus, if $\gamma_0=10$ the best choice to observe the effect is $l=z^-$ or $\alpha_0 l \simeq 10.2$.

V. SIDEBAND EVOLUTION IN A LASER CAVITY

The amplification of sidebands in a saturable absorber is limited—for a given initial saturation parameter—by the depletion of the carrier intensity. If a cell with the SA is placed inside a laser cavity, the gain of the laser medium can compensate the loss in the absorber in each round trip, and the SB amplitudes grow due to both the parametric amplification in the SA and to the laser gain. A quantitative analysis of SB generation and amplification in this case may be of help in understanding the passive mode-locking mechanism in the frequency domain. The SA used for passive mode locking of giant pulse solid-state lasers, for example, is a broadband-fast bleachable dye in solution in a solvent which exhibits negligible dispersion to the laser modes. For most purposes the dye is well described by a homogeneously broadened two-level system[18,25] with a recovery time which is small compared with the inverse of the beat frequency between the modes, and the slow modulation limit is sufficient to describe the steady-state interactions between the modes in the dye.

We have performed calculations for a thin absorption cell with small signal transmission $1/e$ per pass, which is about the typical values used in pulsed solid-state lasers. Only one mode (traveling wave) has been assumed to be present with enough amplitude to act as the saturator wave; and two weak modes, symmetrically spaced in frequency with respect to the first, were assumed to act as sidebands phased for amplitude modulation. We used the equations of Sec. IV to describe the propagation in the absorber and assumed that the action of the laser medium was that of multiplying the amplitude of the three waves by the same amplification factor G in each round trip (G includes stimulated emission and all losses in the cavity other than that due to the SA). All frequency dependences have been ignored, as well as any intensity dependence (saturation) of the laser gain. A constant loop gain is a good approximation[26] in pulsed solid-state lasers at least while the total energy of the pulse cannot significantly deplete the number of active atoms. If G would overcompensate the loss in the SA, the sidebands will grow without limit; eventually, the SB amplitudes would reach values comparable to that of the carrier and our equations would not be applicable. Consequently, we have limited ourselves to values of G a little less than the SA loss. The case of exact compensation is treatable analytically and will be presented at the end of this section.

SB generation. (See Fig. 3.) In Fig. 3(a) the loop gain G is too low to compensate the depletion of the carrier, but is nevertheless enough to allow amplification of both SB's (recall that, in contrast to Sec. IV, in the absence of laser gain the preexistent SB is amplified only if $\gamma_0 > 7$). When the SA loss is almost compensated [Figs. 3(b) and 3(c)] the sideband amplitudes are equalized in few round trips, indicating a nearly pure amplitude modulation.

AM waves. (See Fig. 4.) In Figs. 4(a) and 4(b) we have the same initial conditions as that of Figs. 3(c) and 3(b), respectively, except that now the SB amplitudes are equal from the beginning. The corresponding curves are very similar, with the only difference in the scales for the SB amplitudes, which are now almost doubled.

Comparing the curves for $\gamma_0 = 1$ and $\gamma_0 = 5$ we note that the SB amplitudes reach lower values for higher values of the saturation parameter. At large values of γ_0 the amplification process in the absorber is less efficient and, since the absorption for the carrier is reduced by the strong saturation, the laser gain factor necessary to compensate the losses is also reduced. In fact, note that for $\gamma_0 = 1$ we have only reduced attenuation for the sidebands in the SA but not amplification; thus, the SB amplification observed in Fig. 4(a) is due only to the laser gain.

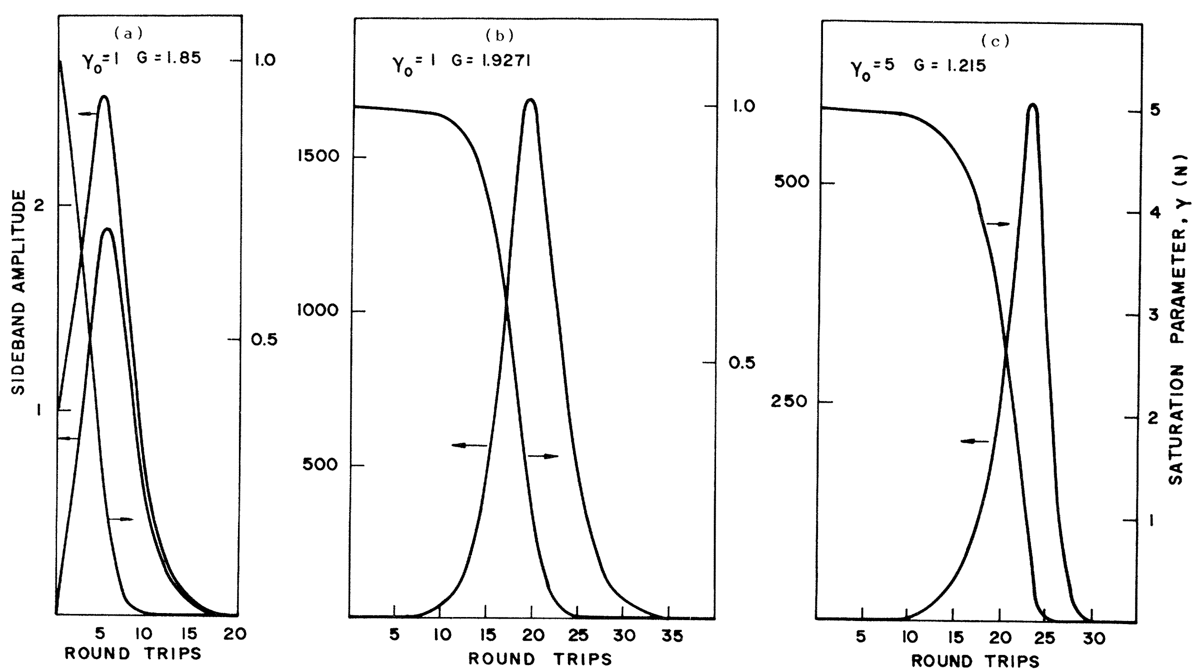
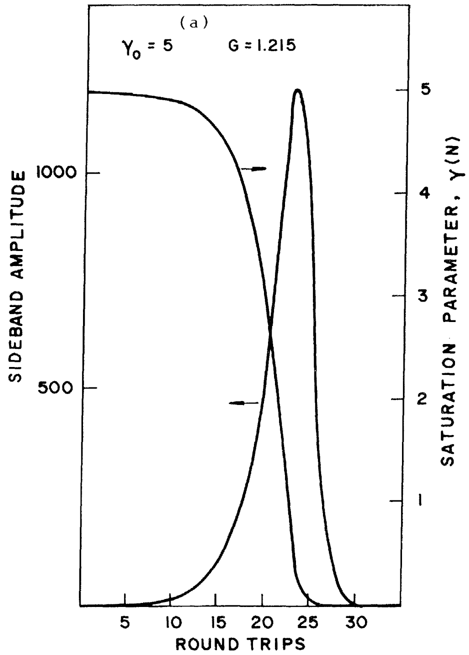


FIG. 3. Sideband generation and evolution of the initially present sideband inside a laser cavity. For the scales used in (b) and (c) the differences in amplitude of the two sidebands are not resolved, indicating a rapid evolution into a pure AM wave.



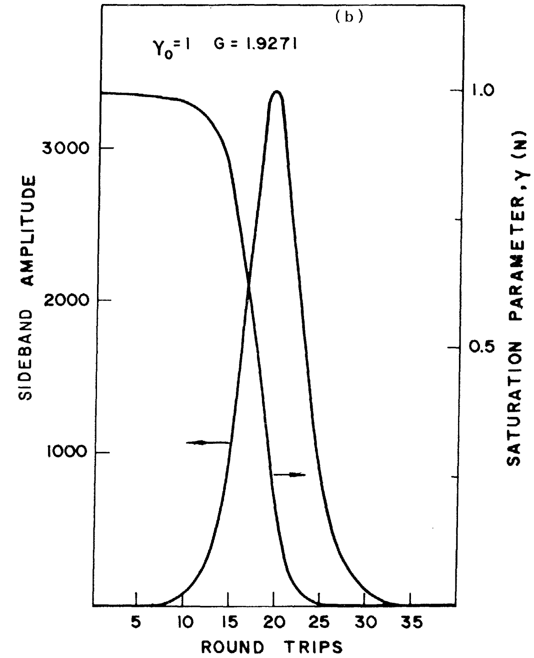


FIG. 4. Evolution of the sidebands of a pure AM wave inside the laser cavity. Compare with Figs. 3(b) and 3(c).

Case of exact compensation of the losses

We call $E_{\pm1}(N)$ and $\gamma(N)$ the SB amplitudes and the saturation parameter which enter the SA cell (length l) after N round trips (Fig. 5). $E'_{\pm1}(N)$ and $\gamma'(N)$ are the same parameters after two passages through the SA cell. After two passages through the laser medium we have

$$E_{\pm1}(N+1)=GE'_{\pm1}(N)\ ,$$
$$\gamma(N+1)=G^2\gamma'(N)\ . \qquad (5.1)$$

The primed parameters are related to the unprimed ones by [see Eqs. (4.2), (4.4), and (4.5)]

$$E'_{\pm1}(N)=\tfrac{1}{2}[\gamma'(N)/\gamma(N)]^{1/2}\left[E_{\rm AM}(N)\left\{\frac{1+\gamma(N)}{1+\gamma'(N)}\right\}\pm E_{\rm FM}(N)\right]\ , \qquad (5.2)$$

$$\gamma'(N)=\gamma(N)\exp[-2\alpha_0 l+\gamma(N)-\gamma'(N)]\ . \qquad (5.3)$$

To solve these equations we need to express $\gamma'(N)$ as a function of $\gamma(N)$. Unfortunately, Eq. (5.3) is implicit and transcendental, and can solved only within some approximations. On the other hand, if the laser gain exactly compensates the depletion of the carrier,

$$\gamma(N+1)=\gamma(N)=\text{const}=\gamma_0\ ,$$

we then have

$$\gamma'(N)=\gamma(N)/G^2$$

and the equations can be solved exactly:

$$E_{\pm1}(N+1)=\tfrac{1}{2}[E_{\rm AM}(N)G^2(1+\gamma_0)/(G^2+\gamma_0)\pm E_{\rm FM}(N)]\ . \qquad (5.4)$$

From this equation one can see that while the FM component remains constant in each round trip, the AM component grows in a geometrical progression:

$$E_{\rm AM}(N)=[G^2(1+\gamma_0)/(G^2+\gamma_0)]^N E_{\rm AM}(0)\ .$$

The ratio of this geometric progression is maximum—and consequently the sidebands grow faster—if $G=\gamma_0$ or, equivalently, if the transmission of the SA cell is such that the saturation parameter falls to $1/\gamma_0$ in two passages (one round trip) through the cell. Under this optimum condition the SA amplifies the SB's of AM waves in one transit and attenuates by the same factor in the next, thus the SA has no overall effect on the SB amplitudes, which then grow due to the laser gain.

This oversimplified model of a laser with an intracavity SA, constant loop gain, and only three modes, is of course a poor approximation of real devices, where the parameters are frequency and time dependent and a huge number

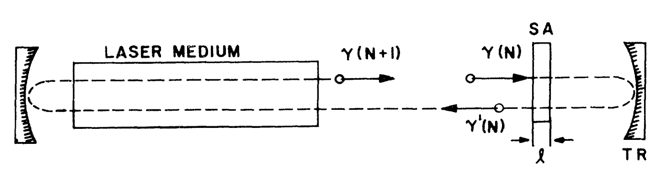


FIG. 5. Definition of the values of the saturation parameter inside the laser cavity. The mirror to the right is a total reflector.

of modes may oscillate. Nevertheless, we can obtain some ideas on how the coherent mode interactions work in the SA of passively mode-locked lasers and regenerative amplifiers. In frequency domain theories of passive mode locking, the role of the SA is described as that of the SB generator and phase-difference "locker" (see Ref. 17 for background discussions on these lines). These effects have been explained as a result of adjacent mode coupling through a third-order nonlinear polarization.[20–22] As discussed in Sec. II, the hypothesis of only adjacent coupling is susceptible to severe criticism, but even in this case our simple analysis with depletion taken into account seems to indicate that the SA may play an important role as a selective mode filter; at some stage of pulse formation in the laser cavity, during the nonlinear loss regime, the stronger modes, which do not enjoy the coherent contributions from population pulsations, are strongly attenuated in the SA; while the weaker modes (generated in part in the SA by frequency mixing) are much less attenuated, and even slightly amplified in the SA, and can then grow due to the laser gain. The net effect is a smoothing of the envelope of the pulse spectra. The selectivity is against stronger modes and also against weaker modes which do not have the proper phase for amplitude modulations.

This selective filter action of the SA would be of help in compensating both the differences in the mode losses of the cavity and, in homogeneously broadened lasers, the gain reduction due to gain saturation caused by the dominant mode.

If the main features remain the same when the extrapolation to real devices is made, one expects that efficient mode locking in lasers will be dictated by the ability of the system in prolonging the duration of the nonlinear loss regime, where the selective filter action of the SA is more efficient.

ACKNOWLEDGMENTS

It is pleasure to acknowledge Professor Alvin Kiel, Professor Cid Araújo, and Professor G. H. C. New for helpful discussions, and Istituto Italo Latino Americano for financial support.

[1] B. Senitzky, G. Gould, and S. Cutter, Phys. Rev. 130, 1460 (1963).

[2] S. L. McCall, Phys. Rev. A 9, 1515 (1974).

[3] P. L. Gordon, S. E. Schwarz, C. V. Shank, and O. R. Wood, Appl. Phys. Lett. 14, 235, (1969).

[4] M. Sargent III, P. E. Toschek, and H. G. Danielmeyer, Appl. Phys. 11, 55 (1976).

[5] M. Sargent III and P. E. Toschek, Appl. Phys. 11, 107 (1976).

[6] M. Sargent III, Phys. Lett. C 43, 223 (1978).

[7] B. H. Soffer and B. B. McFarland, Appl. Phys. Lett. 8, 166 (1966).

[8] S. E. Schwarz and T. Y. Tan, Appl. Phys. Lett. 10, 4 (1967).

[9] B. R. Mollow, Phys. Rev. A 5, 2217 (1972).

[10] F. Y. Wu, S. Ezekiel, M. Ducloy, and B. R. Mollow, Phys. Rev. Lett. 38, 1077 (1977).

[11] A. M. Bonch-Bruevich, V. A. Khodovoi, and N. A. Chigir, Zh. Eksp. Teor. Fiz. 67, 2069 (1974) [Sov. Phys.—JETP 40, 1027 (1975)].

[12] S. G. Rautian and I. I. Sobel'man, Zh. Eksp. Teor. Fiz. 41, 456 (1961) [Sov. Phys.—JETP 14, 328 (1962)].

[13] R. W. Boyd, M. Raymer, P. Narum, and D. J. Harter, Phys. Rev. A 24, 411 (1981).

[14] N. Bloembergen and Y. R. Shen, Phys. Rev. 133, A37 (1964).

[15] C. Cohen-Tanoudji and S. Reynaud, J. Phys. B 10, 345 (1977).

[16] E. T. Jaines and F. W. Cummings, Proc. IEEE, 51, 89 (1963). The local field correction factor and the molecular orientation averaging factor are discussed in R. H. Pantell and H. E. Puthoff, *Fundamentals of Quantum Electronics* (Wiley, New York, 1969).

[17] See, for example, M. Sargent III, M. O. Scully, and W. E. Lamb, Jr., *Laser Physics* (Addison-Wesley, Mass. 1974), Chap. 9; A. Yariv, *Quantum Electronics,* 2nd ed. (Wiley, New York, 1967), Chap. 11.

[18] G. H. C. New, Proc. IEEE 67, 380 (1979).

[19] H. Statz, G. A. De Mars, and C. L. Tang, J. Appl. Phys. 38, 2212 (1967).

[20] H. Statz, J. Appl. Phys. 38, 4648 (1967).

[21] S. E. Schwarz, IEEE J. Quantum Electron. QE-4, 509 (1968).

[22] G. A. Sacchi, G. Sancini, and O. Svelto, Il Nuovo Cimento 48B, 58 (1967).

[23] O. P. McDuff and S. E. Harris; IEEE J. Quantum Electron. QE-3, 101 (1967).

[24] S. Haroche and F. Hartmann, Phys. Rev. A 6, 1280 (1972).

[25] R. R. Cubeddu and O. Svelto, IEEE J. Quantum Electron. QE-5, 495 (1969).

[26] W. R. Sooy, Appl. Phys. Lett. 7, 36 (1965).