Stability of Traveling Waves in Lasers*

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The equations of motion for a fully quantized system of atoms interacting with traveling waves is solved to terms of fourth order in the field strengths. From these, criteria are developed for the stability of traveling waves in solid and gaseous media. Expressions are given for the power outputs for both traveling and standing waves. The results for the latter agree with those obtained by Lamb in his semiclassical treatment of standing waves.

1. INTRODUCTION

 $\mathbf{R}_{\mathrm{theory}}^{\mathrm{ECENTLY}}$ Lamb has published a comprehensive theory of the behavior of standing waves in optical masers.¹ We wish here to add a brief note on some of the additional properties to be expected for individual traveling waves when they are not necessarily constrained to form standing waves. Our considerations apply to waves deflected by three or more mirrors so as to pass through the amplifying medium in the same direction on each traversal, rather than in opposite directions on successive traversals as in the usual laser configuration with just two mirrors.

In arriving at our results we proceed along a somewhat different route from Lamb. This serves to illustrate an alternative approach to the theory of optical masers which, though computationally about as simple as Lamb's, puts a greater emphasis on quantum features which must enter in future theories of noise fluctuations and of feeble signals.

Lamb calculates the increase in amplitude of classical waves contained in a laser in response to oscillations in the atomic polarizations produced by these waves. We prefer to ignore the polarizations, and calculate rather the changes in the numbers of photons populating the waves as the atoms make quantum hops down to less highly excited states. Mathematically, our approach emphasizes the diagonal elements of the density matrix (populations) for the single quantum system consisting of the atoms and radiation, whereas Lamb's focuses on the off-diagonal elements (polarizations) of the smaller density matrix for the atoms interacting with external, unquantized fields.

Traveling waves mean directed quanta of radiation. It is natural to quantize the motions of the atoms in units of the recoil they give to or receive from the fields in the processes of emission and absorption. This, together with the quantized radiation, results in a complete microscopic description of the states in which the momentum and energy of each particle and wave is completely specified. We integrate the equation of motion of the density matrix in this description to terms quadratic in the numbers of photons and derive from the diagonal elements of this matrix expressions for

saturation in the amplification of each wave and for interference between different waves.

In this paper expressions are obtained solely for the populations in the running waves; frequency locking or other effects of mutual shifts in the frequencies of the waves are ignored. The discussion is restricted, moreover, to amplifying media composed of stationary atoms or of gases in which effects of collisions can either be ignored or included in effective decay constants for the atoms. Explicit expressions are given only for the case that the effective decay constants for the two atomic states supporting the laser oscillations are equal. The chief new results obtained are expressions under these conditions for the amplification of each running wave in the presence of other running waves (Sec. 2) and criteria for the mutual stability of pairs of running waves in the absence of frequency locking (Sec. 3). In Sec. 4 the effect on the aggregate emission from the laser of the interference between pairs of waves is discussed and an expression equivalent to one given by Lamb¹ is derived for the emission in the case that a pair of traveling waves is constrained to form a standing wave.

2. AMPLIFICATION OF TRAVELING WAVES

Consider first, for simplicity, a single atom interacting with the running waves in the laser. The atom will normally be moving and may be in one of its excited states. Let the numbers of photons in the traveling waves be N_1, N_2, N_3, \cdots . Use the indices k, l, \cdots to represent individual states of the combined systemmoving atom plus radiation. The development of the system in time can then be written

$$i\hbar\partial\rho_{kt}/\partial t = \sum_{m} (H_{km}\rho_{ml} - \rho_{km}H_{ml}),$$
 (1)

where ρ is the density matrix for the system and H the energy. In a representation in which H is diagonal in the absence of interactions between the waves and the atoms the off-diagonal elements of H correspond to transitions in which the interactions cause (a) one photon in one of the waves to be created or destroyed and (b) the atom to be correspondingly de-excited or excited and given an increment of momentum equal to that absorbed or opposite to that emitted in the

^{*} Brief communications of some of the results of this paper have appeared in Bull. Am. Phys. Soc. 8, 530 (1963); 9, 560 (1964). ¹ W. E. Lamb, Jr., Phys. Rev. 134, A1429 (1964).

transition. The strength of such a transition can be found to be written as

$$H_{kl} = \hbar S_{kl} N_{kl}^{1/2}.$$
 (2)

Here S_{kl} is the strength when the wave causing the transition contains but one photon and N_{kl} is the number of photons in the wave when the atom is deexcited (i.e., N_{kl} is the larger of the numbers of photons in this wave in states k and l).

To solve Eq. (1) write $H_{kk} - H_{ll} = \hbar \omega_{kl}$ for the difference in total energies of states k and l and make the substitution $\rho_{kl}(t) = \rho_{kl}^{0}(t) \exp(-i\omega_{kl}t)$. The development of ρ^0 in time is given by the simple operator equation

$$i\hbar\partial\rho^{0}{}_{kl}/\partial t = \sum_{k',l'} T_{k\,l,k'\,l'}\rho^{0}{}_{k'\,l'},\qquad(3)$$

where

$$T_{kl,k'l'}(t) = (1 - \delta_{kk'} \delta_{l'l}) (H_{kk'} \delta_{l'l} - \delta_{kk'} H_{l'l}) \\ \times e^{i(\omega_{kl} - \omega_{k'l'})t}.$$
(4)

Equation (3) can be solved by iteration. This gives

$$\rho^{0}{}_{kl}(t) = \sum_{k'l'} \tau_{kl,k'l'}(t,0) \rho^{0}{}_{k'l'}(0)$$
(5)

for the state of the system at time *t* in terms of its state at time t=0, where

$$\tau_{kl,k'l'}(t,0) = \delta_{kl,k'l'} - \frac{i}{\hbar} \int_0^t dt' T_{kl,k'l'}(t') \\ - \frac{1}{\hbar^2} \int_0^t dt' \int_0^{t'} dt'' \sum_{k''l''} T_{kl,k''l''}(t') T_{k''l'',k'l'}(t'') \\ + \cdots . \quad (6)$$

In Eq. (6) "+..." means terms of order $(-iT/\hbar)^3$, $(-iT/\hbar)^4$, ... are to be added with triple, quadruple, ···integrations, and summations over two pairs, three pairs, ... of indices.

It is not necessary to include all states in the summations in Eq. (6). In particular, if the two excited atomic states which interact with the laser radiation decay to noninteracting states with the single decay constant γ , this decay can be taken into account by ignoring the latter states in performing the summations in Eq. (6), then multiplying the expression obtained in this way for ρ^0 by exp $(-\gamma t)$. Suppose this is the case and that at time t=0 the atom is excited and the state of the system is $l: \rho_{ll}(0) = 1$. Write $\rho_{kk,ll}(t)$ for the probability that by time t > 0 the system will be found in state k, in which the atom has been stimulated to emit a photon into wave k but has not yet decayed to an inert state. Then when the integrations are performed in Eq. (6), including terms through T^4 , the probability that the atom be in state k after an initial excitation into state l is

$$\rho_{kk,ll}(t) = 2S_{kl}^2 N_{kl} t^2 e^{-\gamma t} \left[\left[(1 - \cos x) / x^2 \right] + t^2 \sum_{m \neq k} S_{ml}^2 N_{ml} f(x, \omega_{ml} t) + t^2 \sum_n S_{kn}^2 N_{kn} f(x, \omega_{kn} t) \right], \quad (7)$$

where $x = \omega_{kl} t$ and

$$xf(x,y) = \frac{1}{y-x} \left[\frac{\cos x - 1}{x^2} - \frac{\cos y - 1}{y^2} \right] + \frac{\sin x}{xy} + \frac{\cos x - 1}{xy} \left[\frac{1}{x} + \frac{1}{y} \right] + \frac{\cos (y-x) - 1}{(y-x)y^2} .$$
 (8)

In the integrations leading to Eq. (7) terms of odd order in T do not contribute; those of sixth and higher order contribute negligibly when the laser operates close to threshold. The leading term in Eq. (7), of second order in T or S, or of first order in the number of photons N, expresses the portion of the response which is free of interference or saturation. The remaining terms, of order S^4 or N^2 , determine the levels of the oscillations in the laser and the dependence of the output for each oscillation on the amplitudes and frequencies of the other oscillations.

The summations on m and n run through all those states, excluding m = k, which can be connected to states l and k by the interaction between the atoms and the radiation: In states of type m the atom is de-excited and an extra photon is present in wave m; in states of type nthe atom has been re-excited by absorbing a photon from wave n. The term in the second summation with n=l is a special case and corresponds to re-excitation from state k back into state l by absorption of a photon from the wave k which initially caused the atom to de-excite. This term has the simple form

$$S_{kl}^2 N_{kl} f(x,x) = -2S_{kl}^2 N_{kl} x^{-4} (2 - 2\cos x - x\sin x);$$

it expresses saturation in the response of the atom to radiation in wave k in the absence of other radiation.

Equations (7) and (8) give the time development of a system consisting of a single atom plus radiation. For a laser, in which there are many atoms, the density matrix at time t is equal to the sum of the density matrices $\rho(t-t_0)$ for the individual atoms excited at times t_0 . Suppose the excitation occurs preferentially into the upper laser state of the atoms at a rate R(t) which varies little during several natural atomic decay times and that the resonant frequency for all of the atoms is the same. Then, for equal decay constants, the rate at which the photon population in wave k increases from

A 1652

stimulated emission is

$$\frac{dN^{(l)}{}_{k}(t)}{dt} = \gamma \int_{-\infty}^{t} R(t_{0})e^{-\gamma(t-t_{0})}\rho^{0}{}_{kk,ll}(t-t_{0})dt_{0} \\
= 2\gamma^{-2}R(t)\frac{S_{kl}^{2}N_{kl}(t)}{1+\bar{x}^{2}} \\
\times \left[1 - \frac{\sum_{m \neq k}\gamma^{-2}S_{ml}^{2}N_{ml}(t)I(\bar{x},\omega_{ml}/\gamma)}{1+\bar{x}^{2}} - \frac{\sum_{n}\gamma^{-2}S_{kn}^{2}N_{kn}(t)I(\bar{x},\omega_{kn}/\gamma)}{1+\bar{x}^{2}}\right], \quad (9)$$

where $\bar{x} = \omega_{kl} / \gamma$ and

$$I(x,y) = \frac{4 + x^2 + x^4 + 2xy^3 + y(1-x^2)(x+2y)}{(1+y^2)[1+(x-y)^2]} .$$
(10)

The first term in Eq. (9) gives the growth of the wave in the absence of interference or saturation; the term in the second summation with n=l and I(x,y)=I(x,x)=4, gives the saturation in the growth of the wave in the absence of other waves; the remaining terms in the summations, containing factors I(x,y) with $y \neq x$, express the effects of interference between pairs of waves. The waves may be described fully by adding a term $-\gamma_{ck}N_k$ to the right side of Eq. (9) to represent the losses in the cavity due to incoherent scattering from the atoms, absorption of the radiation in or transmission through the mirrors, etc. (These processes may reasonably be assumed to be such that γ_{ck} is independent of N_k .)

3. STABILITY OF TRAVELING WAVES

In sparsely populated waves for which the gain exceeds the losses the numbers of photons can be expected to grow toward the equilibrium values obtained by setting Eq. (9), including the loss term for each wave, equal to zero. Whether such an equilibrium with finite populations in each wave will actually be reached depends on the magnitudes of the factors I(x,y)coupling the pairs of running waves. As a rule, if the couplings are too strong the least disturbance from the equilibrium values must result in some of the waves growing larger and others growing smaller and finally disappearing. For example, when there are just two waves k and m with the same polarization whose strengths S_{rs} of interaction with one photon are the same, it is readily seen that Eq. (9) predicts that the populations will tend to move away from their equilibrium values unless the product I(x,y)I(y,x) for the two waves is less than 4. (We have here assumed the populations are large enough to set $N_{ml} \approx N_m \approx N_{kn}$ and $N_{kl} \approx N_k$, where N_m and N_k are the average numbers of photons in waves m and k.) When this inequality is satisfied, as when the frequencies x and y of the two waves differ considerably, the waves can be expected once at equilibrium to coexist indefinitely.

It is readily seen that the condition for stability is not satisfied for waves of nearly the same frequency propagating in the same or opposite directions through stationary media. Thus in a stationary triangular laser with a stationary solid amplifying medium, a standing wave reflected around the perimeter of the triangle should be unstable against growth of one of its constituent running waves at the expense of the other. For the standing wave to become stable the medium must move or the triangle to which the mirrors are attached must rotate. For the special case that the radiation is resonant with the atoms when they are at rest, the condition for coexistence of the two running waves when the atoms are moving is that the two Doppler-shifted frequencies of the waves as viewed by the moving atoms satisfy the condition $\bar{x}^2 = \bar{y}^2 > \frac{1}{5}$.

For gaseous media, which consist of atoms moving with many different velocities, it is necessary to sum the individual contributions (9) for the stimulated emission from each atom to obtain the aggregate emission and interference. The criteria for stability will now, as for the moving but not for the stationary solid, be different for pairs of waves propagating in the same and in opposite directions. Pairs of waves of nearly the same frequency propagating in the same direction should still be unstable against growth of one wave at the expense of the other. But pairs of waves propagating in opposite directions should now normally be stable.

This latter result is a consequence of the fact that, just as for the moving solid, the Doppler shifts experienced by the moving atoms are different for the oppositely directed waves. The quantity (x-y) occurring in Eq. (10) can, accordingly, be expected to be large, and the contribution to I(x,y) small, for many of the atoms in the gas. (This will be true of course even when the frequencies of the traveling waves are nearly equal.) Small values for I(x,y) mean greater stability.

Quantitatively, it is found, upon integrating over velocities in Eq. (9), that the total rate of stimulated emission into wave k when there are just two oppositely directed waves k and m is

$$\frac{dN^{(l)}{}_{k}(t)}{dt} = 2\pi R \gamma^{-1} S_{kl}^{2} N_{kl} \left[1 - \frac{2S_{kl}^{2} N_{kl}}{\gamma^{2}} - \frac{(S_{ml}^{2} N_{ml} + S_{kn}^{2} N_{kn})}{\gamma^{2} + \omega_{av}^{2}} \right]$$
(11)

in the limit of a very broad Gaussian distribution of velocities. Here $2\omega_{av} = \omega_{kl} + \omega_{ml} = \omega_{kl} + \omega_{kn}$. The criterion for stable coexistence of the pair of waves is readily seen to be $(1 + \omega_{av}^2/\gamma^2) > 1$. As this condition is satisfied for nearly all pairs of oppositely directed waves, a gas laser might be expected normally to oscillate simultaneously in all waves which would oscillate by themselves in the absence of interference between oppositely directed waves.

In any real gas, of course, there is a finite distribution of velocities. Equation (11) is applicable only to the extent that the rate of excitation $R=d^2(n_{up}-n_{lo})/dtd(\omega v/c)$ at time t, with ω =angular frequency of the radiation and v=velocity of the atoms, varies inappreciably over the resonances in Eq. (9) at $\omega_{kl}, \omega_{ml}, \omega_{kn}, \omega_{km}, \omega_{nl}=0$. When the variation over the resonances is taken into account, the criterion for stability is found to become somewhat more stringent. This means that instabilities should actually occur somewhat before $\omega_{av}=0$; that is, they should occur whenever the laser is tuned so that the two waves in a pair are approximately resonant simultaneously with some fraction of the atoms.

Our discussion in this section has, of course, consistently ignored effects due to collisions except insofar as they can be incorporated into the effective decay constant γ , so that our results can be expected to apply strictly, if at all, only for dilute gaseous media. We have assumed, moreover, that the coupling between the waves occurs entirely within the amplifying medium, not, e.g., at mirrors or other surfaces where partial reflections might enhance the stability of standing waves.

4. POWER OUTPUTS FOR STANDING AND TRAVELING WAVES

Not only the criteria for stability but also the ratios of the power outputs for standing and traveling waves differ qualitatively for solid and gaseous media. The power outputs are, of course, proportional to the numbers of photons inside the cavity. For solids comprised of stationary atoms with a single resonant frequency, these numbers may be found at equilibrium from Eq. (9) augmented by the dissipation $-\gamma_{ck}N_k$ for each wave. Suppose the dissipations are equal and the strengths S_{rs} are equal. Consider a lone traveling wave $(N_k \text{ or } N_m \neq 0, \text{ all other } N_r = 0)$ of frequency ω and, alternatively, a standing wave of the same frequency composed of two such waves $(N_k = N_m \neq 0, \text{ all }$ other $N_r=0$). It is found from Eq. (9) that the output is larger, by $\frac{3}{2}$, for the traveling wave than for the standing wave composed of the pair of traveling waves. This is true independently of the frequency ω .² As indicated in the last section, the standing wave in such a solid amplifying medium is, moreover, unstable against growth of one of the constituent traveling waves at the expense of the other: in the absence of such constraints as highly reflecting mirrors in a two-mirror cavity the laser operates in that mode of oscillation which gives the greater output. The same ratio $\frac{3}{2}$ is obtained when Eq. (9) is summed over atoms with different resonant frequencies. Thus this ratio and these conclusions can be expected to apply generally to stationary solid amplifying media in nonmoving enclosures.

For gaseous media, the ratio of the power outputs for the two kinds of waves depends on the frequency of the radiation and is usually appreciably less than $\frac{3}{2}$. According to Eq. (11), augmented by the dissipation term, the equilibrium population in an isolated standing wave for large Doppler broadening is

$$2N = \gamma^2 S^{-2} \left[1 - \frac{\gamma \gamma_o}{2\pi R S^2} \right] \left[1 + \frac{1}{1 + (\omega_{\rm av}/\gamma)^2} \right]^{-1}, \quad (12)$$

where ω_{av} is now the difference between the frequency of the standing radiation and the resonant frequency of the atoms when they are not moving. The population at equilibrium in a running wave is $N = \frac{1}{2}\gamma^2 S^{-2}(1-\gamma\gamma_c/2\pi RS^2)$. This is half that given by Eq. (12) for the pair of waves when ω_{av} is large, and equal to that for the pair of waves when $\omega_{av} = 0$. Thus, in the limit of large Doppler broadening, the ratio of the outputs for gaseous media should not exceed 1 for any frequency.

It was found in the last section that in the same limit of large Doppler broadening the standing wave in a gas is at all frequencies as stable as either of its constituent traveling waves. In a real gas, for which a traveling wave becomes the more stable for a small realm of frequencies about $\omega_{av}=0$, a somewhat greater power output can be expected in this realm for the lone traveling wave than for the standing wave, at least if the gas is dilute and the decay constants equal.

It is interesting to observe, in closing, that Eq. (12) predicts for a gas a possible decrease in populations, and hence of output, as the frequency of a standing wave is tuned through the atomic resonance. If the excitation Rof atoms with the correct speed to interact resonantly with the radiation does not vary too rapidly near $\omega_{av} = 0$, the output is predicted there to decrease by as much as 50%. It should decrease by exactly this much for a single standing wave and an infinitely broad distribution of velocities. For a finite distribution of velocities the decrease of R with increasing ω_{av} leads through the first quantity in brackets in Eq. (12) to a less conspicuous dip in output. When R in addition varies appreciably within each resonance, the required alterations in Eq. (11) are found to promote through the second quantity in brackets in Eq. (12) a somewhat deeper dip (assuming the radiation is constrained to remain in the form of a standing wave). The net effect is normally a dip of less than 50%. Expression (12) for the possible decrease in output as the cavity is tuned, through resonance with the internal frequency of the atoms, agrees with the expression for this dip derived directly for standing waves by Lamb.³

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 $^{^{2}}$ A simple, qualitative interpretation of this effect has been given by the author in Nature 201, 911 (1964).

³ Reference 1, Eq. (96).