Linearized Theory of Laser-Induced Instabilities in Liquids and Gases*

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A macroscopic linearized instability theory is developed to describe a class of instabilities resulting from optical-acoustic coupling of a laser beam in fluids and gases. A phenomenon of particular interest is the initial phase in the development of the high-intensity optical filaments which are observed when an intense laser beam propagates through certain fluids. It is suggested that for certain laser cell geometries and a sufficiently high power flux density, filament formation may be preceded by a breakdown in the mode structure of the incident laser beam as a result of coupling to the eigenmodes of the medium. The interaction mechanisms considered are stimulated Raman scattering, electrostriction, the high-frequency Kerr effect, and thermal-energy deposition, while the response of the laser-fluid system is described by Maxwell's equations combined with the appropriately modified conservation equations from hydrodynamics. On the basis of this model, it is proposed that inhomogeneities in the laser intensity, or in the density and temperature of the fluid, act as sources of instability growth for the induced waves which are generated when the primary and scattered optical waves interfere. The dispersion relation for the problem is derived and a procedure for calculating the growth rates of this instability is outlined. The method is illustrated by detailed computations on carbon disulfide covering a range of laser intensities, and it is shown that the laser-Stokes coupling terms do not significantly affect the initial growth rates. In the case of gases, where the electrostrictive effect can be ignored, analytic expressions for the spatial and temporal gains are derived. Under the assumption that the first-order contributions in the linear theory become important after they have undergone several e foldings, these results indicate—for a power flux density, optical-absorption coefficient product of 10^{-8} MW/cm^3—that mode degeneration is expected to occur in a laser beam which has propagated a distance of the order of a few kilometers through air at a pressure of 1 atm or which has a pulse length of several microseconds.

I. INTRODUCTION AND SUMMARY

RECENT experiments[1–3] have shown that high-intensity optical filaments are formed when an intense laser beam propagates a distance larger than some critical length through certain liquids. When this filament formation takes place in Raman-active media, it is invariably accompanied by the now widely discussed[4–7] phenomenon of anomalous Stokes gain. The question, therefore, arises whether this gain is a separate phenomenon explicable in terms of the process of stimulated Raman emission, or whether it is governed mainly by the mechanism responsible for filament formation. In support of the latter alternative, there is now evidence[1,8] indicating that, for a given power flux density in the incident laser beam, the critical length for the onset of anomalous Stokes gain is determined not so much by the Raman susceptibility of the medium, but rather by its optical Kerr constant—i.e., by the anisotropic polarizability of its constituent molecules—through the process of self-focusing. Essentially, a positive Kerr constant leads to an intensity-dependent phase velocity of the light waves such that, for a laser beam with an initial transverse intensity gradient, the parts of the wave front in the region of highest intensity lag increasingly further behind those in the neighboring regions. This results in a transverse shrinking, or self-focusing, of the laser beam which continues until it is limited by other processes, such as diffraction or induced-dipole moment saturation. It has been proposed[9] that the critical length already referred to is the self-focusing length, defined as that distance from the plane of incidence at which the optical intensity starts to become anomalously large. A critical power flux density can then be found by equating the self-focusing and diffraction lengths. Chiao, Garmire, and Townes,[10] Askarjan,[11] and Talanov[12] were among the first to point out the possibility of this self-focusing action of laser beams, while calculations of the above type were first carried out by Kelley.[9] Thus, critical lengths and power fluxes have been computed which for certain fluids are in reasonable agreement with experiment. However, this model does not seem capable of furnishing a satisfactory explanation of at

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[12] V. I. Talanov, Izv. Vysshikh Uchebn. Zavedenii, Radiofiz. **7**, 564 (1964) [English transl.: Radiophysics **7**, 254 (1964)].

least two closely related experimental facts. Firstly, experiments[2,3,13] indicate that the laser beam does not focus as a single unit. Instead, it breaks up into a number of randomly distributed filaments. Secondly, the self-focusing mechanism predicts a diameter for the filaments which is several orders of magnitude larger than the observed value.[2,3,14] It is here suggested that the initially single-mode structure of the laser radiation breaks up into several modes as a result of the interaction of the laser beam with the fluid through electrostriction, thermal-energy deposition, and the Kerr effect. To demonstrate the importance of including all of these processes and to obtain an estimate of the distance the beam has to travel into the medium before mode breakdown occurs, we develop in the present paper a linearized instability theory for the interacting laser-fluid system. The theory is phenomenological in the sense that a macroscopic theory is used to characterize the system, and the effects are assumed to be superimposable. Thus, the medium is described by its gross parameters, and microscopic processes like two-photon absorption are not accounted for, except insofar as they are already contained implicitly in the experimental values selected for such quantities as the optical Kerr constant, the Raman susceptibility, and the optical absorptivity.

In the model adopted here, it is assumed in the single-mode description of the linearized theory that the scattered optical waves resulting from the coupling of the incident coherent laser signal to the free modes of the liquid interferes coherently with the primary wave. As a consequence, interference waves are induced for each eigenmode of the liquid, and it is our aim to determine whether there exist modes that in the presence of an inhomogeneity lead to the excitation of rapidly growing instabilities. We should remark here that any such instabilities are a consequence of the coherent nature of the induced radiation. When this coherence is destroyed by instability growth, therefore, the present model is no longer adequate and must be replaced with that studied in an earlier paper.[15] The possibility of stimulated Raman scattering is taken into account by incorporating in the coupled Maxwell wave equations, polarization source terms whose strength and phase are measured by the Raman susceptibility. The coupling of the optical waves to the fluid is governed by the linearized wave equations and the classical conservation equations of hydrodynamics. In linearized form, the resultant set of simultaneous differential equations has solutions which are, by means of a Green's function, expressible as integrations over the free modes of the fluid. As will become apparent, the poles of the Fourier-Laplace transform of this Green's function are just the solutions of the dispersion relation for the problem. The stability of the laser-fluid system with respect to any given type of disturbance can, therefore, be deduced from the nature of these poles. For the calculation of the long-time response the precise nature of the disturbance is not critical: It may be due to small-scale variations in the laser intensity or in the density, or temperature, of the liquid. The analysis indicates that the nature of the processes contributing to the maximum over-all gain depends for a finite system on the duration of the laser pulse. For sufficiently short pulses (less than 1 nsec in the case of a 15-cm long carbon disulfide cell) the acoustic effects, such as electrostriction and thermal energy deposition, are small and maximum growth is exhibited by a Kerr-modulated electro-optic eigenmode which is phase-matched to the active field so that it propagates freely through the medium. As the pulse length is increased the acoustic processes become progressively more important until for very long pulse times almost all the gain comes from this source.

The paper is arranged as follows. In Sec. II, the set of coupled equations relating the physical variables of the optical radiation and the fluid is derived. These are then linearized and the existence of a differential operator acting on the variables is deduced formally from a consistency criterion. In Sec. III, the Green's function is determined and the nature of its singularities examined. The analysis is illustrated by the detailed calculations for carbon disulfide presented in the first part of Sec. IV. The second part of this section pertains to gases. Here, the analytic derivation of general expressions for the spatial and temporal gains is followed by an application to the case of air. Section V contains a brief recapitulation of the main results of this investigation.

II. THE LINEARIZED DIFFERENTIAL EQUATIONS

In the following, it is assumed that the incident radiation passes through a fluid that is nonmagnetic and electrically nonconducting. In addition to stimulated Raman scattering, the main mechanisms coupling the laser wave to the fluid are supposed to be the high-frequency Kerr effect, electrostriction, and thermal-energy deposition.

The modification of the dielectric constant ϵ resulting from the interaction induces a nonlinear polarization wave. Since only processes involving the acoustical and molecular, rather than the electronic, states of the molecules constituting the liquid are important here, the change of ϵ over an optical wavelength is small compared with that over a typical acoustical length. Thus, in Maxwell's wave equation for the electric field vector $\mathbf{E}$ in regions free from charges and currents, i.e.,

$$\nabla^2\mathbf{E}+\nabla(\mathbf{E}\cdot\nabla\ln\epsilon)=\frac{1}{c^2}\frac{\partial^2}{\partial t^2}(\epsilon\mathbf{E}); \qquad (1)$$

[13] Y. R. Shen and Y. Shaham, Phys. Rev. Letters **15**, 1008 (1965).
[14] Ya. B. Zel'dovich and Ya. P. Raizer, JETP Pis'ma Redaktsiyu **3**, 137 (1966) [English transl.: JETP Letters **3**, 86 (1966)].
[15] K. A. Brueckner and S. Jorna, Phys. Rev. Letters **17**, 78 (1966).

the term involving $\nabla\epsilon$ can be neglected. For a Raman-active medium with a Raman susceptibility χ_R, therefore, the laser and Stokes field intensities E_L and E_S, respectively, satisfy the coupled equations[16]

$$\nabla^2\mathbf{E}_L=\frac{1}{c^2}\frac{\partial^2}{\partial t^2}(\epsilon+\epsilon_2\langle E_L^2\rangle_{\rm av}+4\pi\chi_R{}^L\langle E_S^2\rangle_{\rm av})\mathbf{E}_L, \quad (2)$$

$$\nabla^2\mathbf{E}_S=\frac{1}{c^2}\frac{\partial^2}{\partial t^2}(\epsilon+\epsilon_2\langle E_S^2\rangle_{\rm av}+4\pi\chi_R{}^S\langle E_L^2\rangle_{\rm av})\mathbf{E}_S, \quad (3)$$

where ϵ is the density- and temperature-dependent part of the dielectric constant, ϵ_2 is the optical Kerr constant, and the angular brackets indicate averaging over times that are long compared with an optical period. We have also made the reasonable assumption that $\epsilon_L=\epsilon_S=\epsilon$.

The change in the dielectric constant is, in turn, related to the varying properties of the fluid which, in the macroscopic model adopted here, are governed by the following equations of classical hydrodynamics: momentum,

$$\rho\frac{d\mathbf{v}}{dt}=-\nabla p_{\rm hydr}+\mathbf{f}_{\rm es}+\mathbf{f}_{\rm visc}, \quad (4)$$

energy,

$$\rho T\frac{dS}{dt}=\kappa\nabla^2T-\nabla\cdot\mathbf{F}+\varphi_\eta, \quad (5)$$

matter,

$$\frac{d\rho}{dt}+\rho\nabla\cdot\mathbf{v}=0, \quad (6)$$

in which $\mathbf{v}$ is the "particle" velocity, i.e., the velocity of the center of the volume element, ρ the density, T the temperature, $p_{\rm hydr}$ the pressure in the absence of the field, S the entropy, κ the thermal conductivity of the medium, and $\mathbf{F}$ the time-averaged Poynting vector. The electrostrictive and viscous force densities $\mathbf{f}_{\rm es}$ and $\mathbf{f}_{\rm visc}$, respectively, are defined by the relations

$$\mathbf{f}_{\rm es}=\frac{\rho}{8\pi}\nabla\left\{E^2\left(\frac{\partial\epsilon}{\partial\rho}\right)_T\right\}-\frac{E^2}{8\pi}\left(\frac{\partial\epsilon}{\partial T}\right)_\rho\nabla T, \quad (7)$$

$$\mathbf{f}_{\rm visc}=(\eta+\eta')\nabla(\nabla\cdot\mathbf{v})+\eta\nabla^2\mathbf{v}, \quad (8)$$

in which η is the coefficient of shear viscosity and η' is the "second" or compressional viscosity coefficient. The viscous dissipation function φ_η can be shown[17] to be given by

$$\varphi_\eta=\sum_{i,j,k}\sigma_{ijk}\frac{\partial v_i}{\partial x_k}, \quad (9)$$

with

$$\sigma_{ijk}=\eta\left(\frac{\partial v_i}{\partial x_k}+\frac{\partial v_k}{\partial x_i}\right)+\eta'\delta_{ik}\frac{\partial v_j}{\partial x_j}. \quad (10)$$

[16] Y. R. Shen and N. Bloembergen, Phys. Rev. **137**, 1787 (1965).

[17] F. V. Hunt, J. Acoust. Soc. Am. **27**, 1019 (1955).

To relate the energy equation (5) to the remaining conservation equations, we postulate that the thermodynamic state of the fluid is uniquely specified by three local-state variables—such as the pressure p, specific volume V, and temperature T—whose interdependence is expressed by a functional equation of state of form $S=S(T,V)$ or $S=S(T,p)$.[18] Thus $T=T(V,p)$, which together with the alternative expressions for the equations of state just given, yields[19]

$$\rho T\frac{dS}{dt}=\rho C_v\frac{dT}{dt}-\frac{C_v(\gamma-1)}{\beta}\frac{d\rho}{dt}, \quad (11)$$

where $C_v(C_p)$ is the specific heat at constant volume (pressure), $\gamma=C_p/C_v$, and β is the cubical expansion coefficient defined as $(1/V)(\partial V/\partial T)_p$.

Owing to their complexity, it is not possible to obtain the exact solutions for the above set of simultaneous differential equations in an explicit form. However, they may be solved approximately by invoking a perturbation technique developed by Eckart.[20] In this scheme, it is supposed that each of the physical variables in the problem is expressible as the sum of its slowly varying zero-order component and a small correction. Working only to first order, we denote the zero- and first-order contributions in the perturbation expansion by subscripts 0 and 1, respectively. The incident laser and the Stokes waves are assumed to be described by infinite plane, linearly polarized waves with the well-defined frequencies ω_L and ω_S, and the corresponding wave numbers k_L and k_S. After the perturbation has acted let the fields be represented by

$$\mathbf{E}_L=\tfrac{1}{2}\hat{y}\{(E_0{}^L+E_1{}^L)e^{i[\omega_L t-(k_L-i\alpha/2)z]}+\text{c.c.}\}, \quad (12)$$

$$\mathbf{E}_S=\tfrac{1}{2}\hat{y}\{(E_0{}^S+E_1{}^S)e^{i[\omega_S t-(k_S-i\alpha/2)z]}+\text{c.c.}\}, \quad (13)$$

where α is the optical absorption coefficient in cm^{-1}, and c.c. stands for "complex conjugate." In addition, we set

$$\rho=\rho_0+\rho_1, \quad T=T_0+\theta_1,$$

$$\epsilon\equiv\epsilon(\rho,T)=\epsilon_0+\rho_1\left(\frac{\partial\epsilon}{\partial\rho}\right)_T+\theta_1\left(\frac{\partial\epsilon}{\partial T}\right)_\rho,$$

$$p\equiv p(\rho,T)=p_0+\rho_1\left(\frac{\partial p}{\partial\rho}\right)_T+\theta_1\left(\frac{\partial p}{\partial T}\right)_\rho$$

$$=p_0+\frac{v_s^2}{\gamma}\rho_1+\frac{v_s^2}{\gamma}\beta\rho_0\theta_1, \quad (14)$$

in which v_s is the speed of sound in the medium, while the other quantities are as defined before. In the last

[18] For complete generality this relation should read $S=S(T,p,E)$ since the state of the fluid depends also on the electric field intensity. However, it can readily be shown that in the present case the additional terms arising therefrom are negligible.

[19] M. W. Zemansky, *Heat and Thermodynamics* (McGraw-Hill Book Company, Inc., New York, 1957), 4th ed., Chap. 13.

[20] C. Eckart, Phys. Rev. **73**, 68 (1948).

of these expressions, use has been made of well-known relations from thermodynamics.[19]

If we now assume that the "particle" velocity v is a first-order quantity, i.e., that it is small compared with the sound velocity, we need no longer distinguish between Eulerian and Lagrangian representations in the first-order calculations, so that $d/dt=\partial/\partial t$. Combining Eqs. (4)–(8) and (11) with (12)–(14), and retaining only the dominant terms, we obtain the first-order equations

$$\left(\frac{\partial^2}{\partial t^2}-\frac{v_s^2}{\gamma}\nabla^2\right)\rho_1-\frac{\beta\rho_0 v_s^2}{\gamma}\nabla^2\theta_1 = \frac{-1}{24\pi}(\epsilon_0+2)(\epsilon_0-1)\nabla^2(E_0E_1), \quad (15)$$

$$\frac{C_v(\gamma-1)}{\beta}\frac{\partial\rho_1}{\partial t}-\rho_0 C_v\frac{\partial\theta_1}{\partial t}=-\frac{\alpha n_0 c}{4\pi}E_0E_1, \quad (16)$$

with $E_0E_1=E_0^LE_1^L+E_0^SE_1^S$ and where, in addition to the already defined parameters, we have introduced the index of refraction $n_0\equiv\epsilon_0^{1/2}$. The first of these equations incorporates the effect of the body forces on the properties of the fluid, while the second takes account of the deposition of thermal energy by the incident radiation. The density dependence of the dielectric constant has been assumed to be given by the Lorentz-Lorenz formula: $\rho_0(\partial\epsilon/\partial\rho)_T=\frac{1}{3}(\epsilon_0+2)(\epsilon_0-1)$. In calculating $(\nabla\cdot\mathbf{f}_{es})_1$ we have retained only the leading contributions. In particular, terms arising from the second member of (7) have been neglected in view of the relative smallness of $(\partial\epsilon/\partial T)_\rho$, which Oster[21] estimates to be about $10^{-5}°C^{-1}$.

To complete the set of equations, we finally need a relationship between E_1, θ_1, and ρ_1. This is given by the Maxwell wave equations (2) and (3). The linearized equation for the laser wave is

$$\left[\nabla^2-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\epsilon^{(0)}\right]E_L^{(1)}=\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\epsilon^{(1)}E_L^{(0)}, \quad (17)$$

with a similar equation for the Stokes wave. In (17) $\epsilon^{(0)}$ and $\epsilon^{(1)}$ are the zero- and first-order parts of the intensity-, density-, and temperature-dependent dielectric constant.

By (14), therefore, (15) and (16) are reducible to equations involving only ρ_1 and θ_1. These can be solved when the perturbations are prescribed; in terms, say, of their Fourier transforms. The consistency criterion for nontrivial solutions then states that ρ_1 and θ_1 must both satisfy an equation of form

$$\mathfrak{L}u=0. \quad (18)$$

The action of the differential operator $\mathfrak{L}$ on u is discussed in Sec. 3.

[21] G. Oster, Chem. Rev. 43, 319 (1958).

III. DERIVATION AND ANALYSIS OF THE DISPERSION RELATION

Within the framework of our assumptions, Eqs. (15) and (16), together with (17), constitute a complete first-order description of the fluid's response, in the absence of any inhomogeneities. Disturbances do, however, exist, either in the incident laser intensity or in the density and temperature of the fluid, which may cause a drastic change in the character of the propagating light wave. For example, the coupling to the incident beam of perturbations in the macroscopic fluid density can under certain conditions lead to an enhancement in the coherent oscillations of the beam at the expense of the incident signal energy. The disturbances can, therefore, act as sources for growing temporal and spatial instabilities. To determine the nature of such unstable modes in the laser-fluid system, and to calculate their initial growth rates, we solve (15) and (16) subject to the presence of a source function in one of the physical variables.

Let the source function be represented by $s(t,z,\mathbf{r}_T)$, where $\mathbf{r}_T$ is the position vector transverse to the z axis. The solution of (18) then takes the form

$$u(t,z,\mathbf{r}_T)=\int G(t-t',z-z',\mathbf{r}_T,\mathbf{r}_T')\times s_u(t',z',\mathbf{r}_T')dt'dz'd^2\mathbf{r}_T', \quad (19)$$

where the kernel G is the Green's function for the operator $\mathfrak{L}$ giving the response to an impulse stimulus at the point $(t',z',\mathbf{r}_T')$. An expression for the function u in terms of the parameters of the problem can now be derived by resolving G and s_u in terms of the eigenmodes of the fluid, each of which is defined by a frequency ω and wave numbers k_x, k_y, and k_z. The causality condition, which states that for times $t<0$ the perturbations must equal zero, can be conveniently satisfied by taking the Laplace transform for the time coordinate. Since any disturbance propagates at a finite speed, the Fourier transform is appropriate for the spatial coordinates.

The notation is, without much loss of generality, simplified considerably by selecting a particular transverse eigenmode. We choose $k_y=0$, and postulate a plane boundary at $z=0$ so that k_x is restricted to be real. The optimum value of k_x depends on the nature of the dominant interaction and will be determined later. Since we are only interested in the asymptotic behavior of the integral in (19), the detailed form of s_u does not have to be specified. It merely needs to be stated that the only singularities contributing to maximum instability growth are those of the Green's function. This condition would be met, for instance, by any reasonable source function of finite spatial extent, modulated by a real frequency. In the present problem, this degree of erudition is unnecessary and it suffices to represent the source function by a Dirac δ function in time and space. The Fourier-Laplace transform of the

Green's function of the operator $\mathfrak{L}$, therefore, enables (19) to be reduced to

$$u(t,z,x)=\frac{u_0}{(2\pi)^3}\int_{L_\omega}\int_{\mathbf{k}}\tilde{G}(\omega,k_z,k_x)e^{i(\omega t-\mathbf{k}\cdot\mathbf{r})}d\omega d^2\mathbf{k}, \quad (20)$$

where u_0 is the strength of the source. This is substituted in (15) and (16), where the contribution from E_0E_1 will now be determined via a relation similar to (17) with the Stokes wave coupled in by means of (3) and (13). The expression for E_0E_1 is most conveniently derived by first expressing the laser and Stokes fields in real form. It is evident that the nonlinear terms in the field equations couple the scattered waves resulting from the acoustic interaction. Thus, these waves cannot be assumed to propagate independently of one another, and we must write[22]

$$\begin{aligned}E_L&=E_0{}^L\cos(\omega_L t-k_L z)+E_+{}^L\cos(\omega_+{}^L t-\mathbf{k}_+{}^L\cdot\mathbf{r})\\&\quad+E_-{}^L\cos(\omega_-{}^L t-\mathbf{k}_-{}^L\cdot\mathbf{r}),\\E_S&=E_0{}^S\cos(\omega_S t-k_S z)+E_+{}^S\cos(\omega_+{}^S t-\mathbf{k}_+{}^S\cdot\mathbf{r})\\&\quad+E_-{}^S\cos(\omega_-{}^S t-\mathbf{k}_-{}^S\cdot\mathbf{r}),\end{aligned} \quad (21)$$

in which for the laser perturbations

$$\omega_\pm{}^L=\omega_L\pm\omega, \quad k_\pm{}^L=k_L\hat{z}\pm\mathbf{k},$$

with $L\rightarrow S$ for the corresponding Stokes perturbations, and where ω and $\mathbf{k}$ represent the frequency and wave vector of the perturbation. Substitution into the coupled Maxwell equations (2) and (3) yields, after averaging over times that are long compared with an optical period, the zero-order results

$$\begin{aligned}k_L{}^2&=(\omega_L{}^2/c^2)[\epsilon_0+\tfrac{1}{2}\epsilon_2(E_0{}^L)^2+2\pi\chi_R(E_0{}^S)^2],\\k_S{}^2&=(\omega_S{}^2/c^2)[\epsilon_0+\tfrac{1}{2}\epsilon_2(E_0{}^S)^2+2\pi\chi_R(E_0{}^L)^2].\end{aligned} \quad (22)$$

We assume for simplicity that the Stokes shifts are small, and that the phase mismatch between the modes characterized by the phase factors $(\omega_+t-\mathbf{k}_+\cdot\mathbf{r})$ and $(\omega_-t-\mathbf{k}_-\cdot\mathbf{r})$ is such that these modes may be considered to move independently. The terms involving the same phase factors can then be equated, and we have from Eqs. (2), (3), and (21), that to first order,

$$\begin{aligned}E_+{}^L+E_-{}^L&=\left(\frac{kk_L}{n_0}\right)^2E_0{}^L\xi_L\left[4\pi\chi_RE_0{}^S(E_+{}^S+E_-{}^S)\right.\\&\quad\left.+\tfrac{1}{3}(\epsilon_0+2)(\epsilon_0-1)\frac{\rho_1}{\rho_0}\right],\\E_+{}^S+E_-{}^S&=\left(\frac{kk_L}{n_0}\right)^2E_0{}^S\xi_S\left[4\pi\chi_RE_0{}^L(E_+{}^L+E_-{}^L)\right.\\&\quad\left.+\tfrac{1}{3}(\epsilon_0+2)(\epsilon_0-1)\frac{\rho_1}{\rho_0}\right],\end{aligned} \quad (23)$$

where

$$\begin{aligned}\xi_L&=\left(k^2-\frac{8\pi\epsilon_2}{n_0{}^3c}P_Lk_L{}^2\right)-4\left(\frac{n_0k_L}{ck}\right)^2\left(\omega-\frac{c}{n_0}k_z\right)^2,\\\xi_S&=\left(k^2-\frac{8\pi\epsilon_2}{n_0{}^3c}P_Sk_L{}^2\right)-4\left(\frac{n_0k_L}{ck}\right)^2\left(\omega-\frac{c}{n_0}k_z\right)^2,\\k^2&=k_z{}^2+k_x{}^2,\\P_L&=(n_0c/8\pi)(E_0{}^L)^2,\\P_S&=(n_0c/8\pi)(E_0{}^S)^2.\end{aligned} \quad (24)$$

Eliminating $(E_+{}^S+E_-{}^S)$ with the aid of the second equation in (23) yields finally that

$$E_0E_1=\frac{8\pi}{3}\frac{k_L{}^2}{n_0{}^3c}(\epsilon_0-1)(\epsilon_0+2)\frac{\rho_1}{\rho_0}\times\frac{\xi_SP_L+\xi_LP_S+2\lambda P_LP_S}{\xi_S\xi_L-\lambda^2P_LP_S}, \quad (25)$$

in which

$$\lambda=(32\pi^2/n_0{}^3c)\chi_Rk_L{}^2.$$

Substitution for E_0E_1 into (15) and (16) yields two equations involving ρ_1 and θ_1, and setting the determinant of the coefficients equal to zero we obtain the dispersion relation

$$D(\omega,k_x,k_z)=0, \quad (26)$$

where

$$\begin{aligned}D=&[\omega^2-v_s{}^2k^2-i\omega(2\eta+\eta')k^2/\rho_0](\xi_S\xi_L-\lambda^2P_LP_S)\\&+(P_L\xi_S+P_S\xi_L+2\lambda P_LP_S)k^2(\iota+i\delta/\omega).\end{aligned} \quad (27)$$

We have adopted the following further abbreviations:

$$\begin{aligned}\iota&=[\tfrac{1}{3}(\epsilon_0+2)(\epsilon_0-1)]^2k_L{}^2/\rho_0n_0{}^3c,\\\delta&=\tfrac{2}{3}(\epsilon_0+2)(\epsilon_0-1)\beta v_s{}^2\alpha k_L{}^2/\rho_0n_0{}^2C_p.\end{aligned} \quad (28)$$

Evidently, $\tilde{G}=D^{-1}$ and the singularities of $\tilde{G}$ are just the solutions of the dispersion relation (26).

It may at this stage be instructive to discuss the origin of the various contributions in the dispersion relation, and from the way they are combined therein gain an intuitive insight into the nature of the various phenomena that take place in the laser-fluid system. The last term in Eq. (27) represents the body forces on the medium arising from electrostriction and thermal-energy deposition. These forces couple together the free-running modes of the system which are described by the zeros of the rest of the equation. The vanishing of the first term in (27), i.e.,

$$\omega^2=v_s{}^2k^2+i\omega(2\eta+\eta')k^2/\rho_0,$$

represents the damped acoustic modes of the liquid moving at sound velocity. The vanishing of the second factor corresponds to an electro-optic mode propagating without density fluctuation. In the absence of the Stokes wave, this free mode satisfies the condition $\xi_L=0$,

[22] Since the coupling parameter involves the Kerr constant, it appears at first sight that the cross terms involving both scattered waves may be ignored. However, as was pointed out to us by Professor N. Kroll, this assumption is in general invalid, particularly in the case where the Kerr-effect modulated waves are phase matched to the primary waves.

which by Eq. (24) is satisfied when

$$\omega-\frac{c}{n_0}k_z=\pm\frac{c}{2n_0k_L}\{k^2(k^2-k_0^2)\}^{1/2}, \tag{29}$$

where

$$k_0^2=8\pi\epsilon_2P_Lk_L^2/cn_0^3. \tag{30}$$

This result predicts a convective instability whose maximum spatial growth, obtained by keeping ω real[23,24] and maximizing the imaginary part of the right-hand side of Eq. (29), occurs for $k^2=\frac{1}{2}k_0^2$. For this value of k^2, the growing electro-optic mode is described by

$$k_z=\frac{n_0}{c}\omega+i\frac{1}{4}\frac{k_0^2}{k_L}, \tag{31}$$

propagating at the group velocity

$$\mathrm{Re}\left(\frac{\partial k_z}{\partial\omega}\right)^{-1}=\frac{c}{n_0}, \tag{32}$$

i.e., at the speed of light in the medium. The corresponding spatial gain is given by

$$\mathrm{Im}k_z=\tfrac{1}{4}(k_0^2/k_L), \tag{33}$$

in agreement with that found by Bespalov and Talanov,[25] and subsequently by Chiao, Kelley, and Garmire,[26] for the static mode $\omega=0$. The process described here is essentially that of matching the phases of the primary and index modulated waves so as to maximize the amount of energy transferred from the active to the passive fields. All other combinations of frequency and wave number result in phase mismatches which, because of the resulting destructive interference, reduce this energy transfer and hence the gain.

In the presence of a Stokes wave, the electro-optic mode satisfies the more complicated relation

$$\xi_S\xi_L=\lambda^2P_LP_S.$$

This is once more a wave with the Kerr effect and the index modulation resulting from the superposition of the laser and Stokes waves matched to the interference pattern of the optical waves. Again, other wave mixtures are possible, but since they do not correspond to the proper phase-match, destructive interference would result, preventing the propagation of an electro-optic eigenmode.

In addition to the electro-optic type of instability there also exist more slowly convecting acoustically driven modes which are excited by the body forces in the medium resulting from the processes of electrostriction and thermal-energy deposition. In general, neither unstable mode propagates independently of the other due to the link between the optical intensity and the density changes caused by the body forces. Thus, in addition to exciting the acoustic instabilities, the driving terms in the last member of Eq. (27) mix these with the electro-optic modes. The determination of the precise mixture of these distinct types of instability yielding maximum over-all gain depends, as will be shown in the succeeding section, on the length of the cell and the group velocity of the unstable mode in relation to the duration of the laser pulse. However, if in anticipation of a conclusion reached later in this section the laser-Stokes coupling terms are neglected, we can already make the tentative observations that for long cells or short laser pulses the most important unstable mode is that driven by the Kerr effect with a gain and group velocity given by (33) and (32), respectively, while for short cells or long laser pulses the more slowly propagating acoustic type of instability dominates.

We now turn to a discussion of the integral in (20). To satisfy the causality condition, the Laplace contour L_ω is to be chosen such that it passes below all singularities of $\tilde{G}$ in the ω plane. In other words, if the real k_z axis is mapped onto the ω plane with the dispersion relation as mapping function, L_ω might be as illustrated in Fig. 1. Two branches of some arbitrary dispersion relation are shown, and the sole restraint on σ is that it must exceed the maximum value of the negative imaginary part, $-\mathrm{Im}\omega$, of ω for real k_z. The integral in (20) can now be evaluated for $t>0$ and $z>0$ by closing the contour in the ω plane in an anticlockwise sense and that in the k_z plane in a clockwise sense.

In deriving the most rapidly growing unstable modes from the dispersion relation, we make extensive use of results obtained by Bers and Briggs[23] and discussed exhaustively by Briggs in his monograph[24] on electron-stream interaction with plasmas. The following remarks are included for the sake of completeness. They constitute only a very sketchy outline of the more pertinent features of the analysis, and the above sources should certainly be consulted for the detailed justification of the instability criteria adopted hereinafter.

It will be clear from the preceding considerations that the integral in (20) diverges exponentially in time if, for real k_z, the dispersion relation admits of solutions for ω which have a negative imaginary part. Moreover, the maximum temporal growth rate is given by

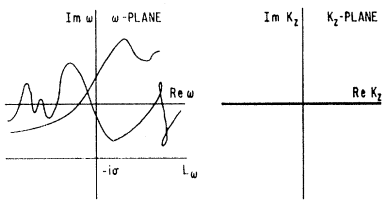

FIG. 1. Laplace and Fourier contours for an arbitrary dispersion relation.

[23] A. Bers and R. J. Briggs, Massachusetts Institute of Technology Research Laboratory of Electronics Quarterly Progress Report No. 71, 1963, p. 122 (unpublished).

[24] R. J. Briggs, *Electron-Stream Interaction with Plasmas* (The MIT Press, Cambridge, Massachusetts, 1964).

[25] V. I. Bespalov and V. I. Talanov, JETP Pis'ma Redaktsiyu 3, 471 (1966) [English transl.: JETP Letters 3, 307 (1966)].

[26] R. Y. Chiao, P. L. Kelley, and E. Garmire (to be published).

$\max(-\mathrm{Im}\omega)$ for real k_z. In the absence of any absolute instabilities,[27] this growing mode will convect through the medium at a certain speed, v_0 say, where

$$v_0=\frac{\partial}{\partial k_z}(\mathrm{Re}\omega)\Big|_{k_z=k_0}$$

and k_0 satisfies the equation

$$\frac{\partial}{\partial k_z}(\mathrm{Im}\omega)=0.$$

The corresponding spatial gain is then $\{\max(-\mathrm{Im}\omega)\}/v_0$. When dealing with a finite system, the speed v_0 may be such that during the time of interaction the peak of the convective instability propagates beyond its confines. In that case, a different mode may yield larger spatial growth, and the maximum of $-\mathrm{Im}\omega(v)/v$ will provide a more significant estimate of the strength of the instability. The speed v is now the group velocity of the unstable mode whose temporal growth rate is $-\mathrm{Im}\omega(v)$.

To derive the functional dependence of the temporal gain on v, it is convenient to make a Galilean transformation to a reference frame in which the convective instability is at rest; i.e., in the moving reference frame the convective instability becomes an absolute instability. Accordingly, we set $z=vt$ in (20) and investigate the long-time behavior of $u(t,vt)$. Note that the replacement $z=vt$ corresponds to the change of variable $\omega\rightarrow\omega'=\omega-k_zv$. The transformed integral can be evaluated by the method of steepest descents, and the asymptotic behavior of u is determined by the solutions of the saddle-point equation $\partial\omega'/\partial k_z=0$, for each value of v. In fact, if these solutions are denoted by, say, $\omega_s(v)$ and $k_s(v)$, it follows that $u\sim e^{i\omega_s(v)t}t^{-1/2}$ for large t. Thus, $-\mathrm{Im}\omega_s(v)$ is the growth rate "seen" by some measuring device moving at speed v through the medium. (This is strictly true only for $v^2\ll c^2$, since otherwise relativistic effects necessitate the replacement of the Galilean by the more cumbersome Lorentz transformation.) We make the parenthetical comment here that while $k_s(v)$ is generally complex, the above considerations indicate that it is real when $v=v_0$, the group velocity of the waves with maximum temporal gain. There is usually more than one pair of solutions to the saddle-point equation, and the choice of ω_s and k_s specifying the position of the saddle point to be employed in the integration is subject to an important restriction, a consequence of the fact that a physical variable, such as u in (19), must be described by a single-valued function. This point has been treated in detail by Briggs,[24] and, briefly, the condition is as follows. Since $\partial\omega'/\partial k_z=0$ implies that $\partial\tilde{G}/\partial k_z=0$, $k_z=k_s$ is a double pole of $\tilde{G}$, and the path of integration in the k_z plane must be chosen to pass through the double pole resulting from a merging of those roots which for $\mathrm{Im}\omega\rightarrow-\infty$ lie on opposite sides of the real k_z axis. This limitation on the choice of k_s has an interesting physical interpretation. Following Briggs, we suppose that an infinite medium is excited by a spatial impulse source located at some point $z=d$ whose temporal growth rate exceeds that of any unstable mode capable of being generated in the system. The resulting waves must all decay away from the origin of the source, so that it can be determined which wave numbers arise for $z>d$ and which for $z<d$. If the time-dependent nature of the source is now altered by decreasing its growth rate, a complex $\omega=\omega_s$ may be found for which the wave numbers on opposite sides of $z=d$ are equal. At this frequency, therefore, a kind of spatial "resonance" is possible, since a response that varies smoothly through the point $z=d$ can be set up in the absence of a source. Clearly, the matching of the wave numbers for $\omega=\omega_s$ corresponds to the merging of the poles in the k_z plane referred to above.

In accordance with the above prescription, the saddle-point equation for the present problem follows from (26) by setting $\partial D(\omega',k_z)/\partial k_z=0$, with $\omega'=\omega-vk_z$. When this is combined with (26), there results a twelfth-order equation for ω_s in which, for each particular choice of v, only one root survives on application of the merging-pole condition. The spatial gain is then simply the ratio of $-\mathrm{Im}\omega_s$ and the appropriate value of v. We would emphasize that the growing modes thus determined are a consequence of the coherence of the interacting waves. When this coherence becomes degraded due to the instability development, the present model breaks down and becomes replaced with that based on the eikonal approximation discussed in an earlier paper.[15] For large k_x, these approaches merge in the diffraction limit $k_x^2=k_zk_L$, as is readily verified by substitution in the respective dispersion relations.

In the next section, we study a particular case in detail. Before doing so, however, we wish to make a significant simplification. The range of the Raman susceptibility for the liquids that are of interest in connection with the problem of filament formation is 10^{-14}–10^{-12} esu, while that for the high-frequency Kerr constant is 10^{-12}–10^{-11} esu. Typically, $k_L\approx10^5$ cm^{-1} and, as will be demonstrated shortly, the optimum value of k^2 is governed by the magnitude of the Kerr term in the quantity ξ_L defined in (24) or by the acoustic properties of the medium, depending on the mode under consideration. For these values of the parameters, therefore, it is clear from (27) and (24) that the contributions to the dispersion relation of the laser-Stokes coupling terms are small, even for a laser power flux density of 100 MW/cm^2 and a Stokes conversion rate of a few percent.

IV. APPLICATIONS

A. Application to Carbon Disulfide

The structure of (27) is quite complicated and the general solution may be obtained only by extensive

[27] The definition of an absolute instability as it is used here is given in Chap. 2 of Ref. 24.

numerical study of its zeros. It is, however, of interest to discuss separately the acoustically driven instabilities and those of a predominantly electro-optic origin, since they have widely different regions of importance. As noted at the end of the preceding section, we may make the simplification of dropping the Stokes terms in the equation. The dispersion relation thus reduces to

$$\left[\omega^2-v_s^2k^2-i\omega(2\eta+\eta')\frac{k^2}{\rho_0}\right]\times\left[\left(\omega-\frac{c}{n_0}k_z\right)^2-(k^2-k_0^2)\left(\frac{kc}{2n_0k_L}\right)^2\right]=k^4\left(\frac{c}{2n_0k_L}\right)^2P_L\left(\iota+i\frac{\delta}{\omega}\right). \quad (34)$$

We have already seen that the fast-moving instability is that due to the electro-optic effect. Optimum gain resulted with $k^2=\frac{1}{2}k_0^2$. For the static mode $\omega=0$, we can define an angle $\theta=k_x/k_L$ which yields a measure of the maximum divergence of the scattered beams. Since $k_L^2\gg k_0^2$, we have for the optimum case

$$\theta_{\text{opt}}=\frac{k_0}{k_L\sqrt{2}}=\left(\frac{4\pi\epsilon_2}{cn_0^3}P_L\right)^{1/2}. \quad (35)$$

The transverse scale of the instability development, $\pi/(k_x)_{\max}$, provides an estimate of the initial diameter of the optical filaments. To illustrate, the appropriate data for carbon disulfide are $\epsilon_2=7.5\times10^{-11}$ esu, $\epsilon_0=2.66$, and, typically, $\omega_L=2.7\times10^{15}$ sec^{-1}. Thus, for a power flux density of 40 MW/cm^2, $\pi/(k_x)_{\max}\approx150\,\mu$ which, of course, exceeds the observed filament diameter[2,3] ($\approx50\,\mu$) as this is ultimately governed by strictly nonlinear processes not covered by the present theory. It is the smallness of this quantity in comparison with the over-all diameter of the incident laser beam that justifies our representing the primary radiation by a plane wave. For these values of the parameters and the power flux density, the gain for this mode is from (33) given by

$$k_0^2/4k_L\simeq0.22\text{ cm}^{-1},$$

or about 3 e foldings in a cell 15 cm long. The dependence of gain on θ or k_x follows directly from (29) and is illustrated in Fig. 2. The gain does not fall off to zero at $\sqrt{2}\theta_{\text{opt}}$ due to the presence of acoustical effects. Similar results have been deduced by Bespalov and Talanov,[25] and more recently by Chiao, Kelley, and Garmire.[26] (Evidently, there is a close link between the resonance-type criterion invoked here for the determination of θ_{opt} and the phase-matching condition in the "weak-wave retardation" scheme employed in the latter reference.) Equally valid results would have been obtained with k_x and k_y interchanged, so that the scattered radiation forms a cone around the z axis whose apical angle is approximated by θ_{opt}.

We now set out to calculate the spatial gain and group velocity for weakly driven acoustical modes. The

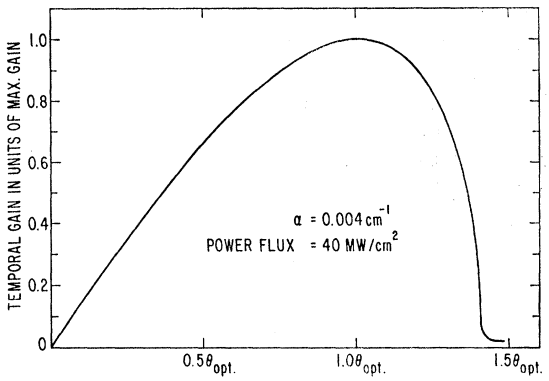

FIG. 2. Temporal gain for carbon disulfide as a function of θ in the vicinity of the Kerr-matched mode.

maximum growth rates in this case are expected to occur for the largest value of k_x compatible with its reality, imposed by our assumption of plane waves at the origin $z=0$. The limit is set by acoustic damping, and we must have

$$(2\eta+\eta')k_x\ll\rho_0v_s.$$

For most liquids this means that we may take 10^4 cm^{-1} for the maximum value of k_x. Since k_x^2 is here much larger than k_0^2 and k_z^2, the dispersion relation to be analyzed takes the form

$$(\omega^2-v_s^2k_x^2)\left[\left(\omega-\frac{c}{n_0}k_z\right)^2-A\right]=AP_L\left(\iota+i\frac{\delta}{\omega}\right), \quad (36)$$

where

$$A=k_x^4(c/2n_0k_L)^2.$$

We expect the weakly driven modes to lie close to those of the free system and hence we write

$$\omega=v_sk_x+i\omega_i, \quad (37)$$

where $|\omega_i|\ll v_sk_x$. Substituting some typical values for the fluid parameters in the expressions for ι and δ given by Eq. (28), it is readily verified that for the values of ω and k_x adopted here $|\delta/\omega|\ll\iota$ so that the last term on the right of (36) may be ignored. The functional dependence of the gain on the propagation speed of the unstable modes is calculated by adopting the technique outlined in Sec. III. We introduce a Galilean transformation and set

$$\omega'=\omega-v_gk_z, \quad (38)$$

where ω' is subject to the condition $\partial\omega'/\partial k_z=0$, or, alternatively, ω satisfies $\partial\omega/\partial k_z=v_g$. The spatial gain is then given by $-\text{Im}\omega'/v_g$. Differentiation of Eq. (36) with respect to k_z yields, therefore, that

$$2\omega v_g\left[\left(\omega-\frac{c}{n_0}k_z\right)^2-A\right]+2\left(v_g-\frac{c}{n_0}\right)(\omega^2-v_s^2k_x^2)\left(\omega-\frac{c}{n_0}k_z\right)=0, \quad (39)$$

where the term involving the absorption coefficient has again been neglected as we are here only interested in the region for which $v_g \ll c/n_0$.

To solve these simultaneous equations for ω_i and k_i, the imaginary part of k_z, we note that for power flux densities of the order of 40 MW/cm² the magnitude of the driving terms in Eq. (36) is small. Thus, we must choose

$$k_r = \frac{n_0}{c} A^{1/2} = -\frac{1}{2}\frac{k_x^2}{k_L}, \tag{40}$$

whence it follows that $\omega_r \ll (c/n_0)k_r$. Equating real parts and retaining only the dominant terms, we deduce from Eq. (36) that

$$\omega_i\left(\omega_i - \frac{c}{n_0}k_i\right) = \frac{n_0 A P_L \iota}{c\, 4\omega_r k_r}. \tag{41}$$

From the real part of (39), it follows that

$$\omega_i = -\frac{c}{n_0}\frac{v_g k_i}{(c/n_0 - 2v_g)}. \tag{42}$$

Combining Eqs. (38), (41), and (42), we obtain the result that the spatial gain is given by

$$-\mathrm{Im}\frac{\omega'}{v_g} = k_i - \frac{\omega_i}{v_g} = \left[\frac{k_x \iota}{2v_s v_g k_L}\left(1 - \frac{n_0 v_g}{c}\right)P_L\right]^{1/2}. \tag{43}$$

The physical constants for carbon disulfide appearing in the above expressions have the values: $n_0 = 1.63$, $\rho_0 = 1.26$ g/cc, $v_s = 1.15\times10^5$ cm/sec, $C_p = 9.86\times10^6$ erg/g°C, $\beta = 1.2\times10^{-3}$/°C, $2\eta+\eta' = 0.726$ P, $\omega_L = 2.7\times10^{15}$ sec^{-1}. The quantity $2\eta+\eta'$, occurring in the viscous damping coefficient, has been determined by Truesdell[28] from acoustical absorption data. For these values of the parameters and $k_x = 10^4$ cm^{-1} the spatial gain, g say, becomes from Eq. (43)

$$g = 1.6\times10^3 (P_L/v_g)^{1/2}, \tag{44}$$

for $v_g \ll c/n_0$, and where the laser power flux P_L is in MW/cm². For $v_g = 2\times10^8$ cm/sec and $P_L = 40$ MW/cm² the gain of the acoustically driven instability is about 0.7 e foldings per cm, or more than three times the Kerr gain. Exactly at $v_g = 0$, the gain goes to zero as is obvious when the substitution is made in Eqs. (39) and (36).

To determine the growth rate for other than the extreme cases discussed above, numerical evaluation of the dispersion relation is necessary. Under these conditions the strongly driven mode departs very appreciably from the free-running eigenmodes of the medium, with the wave-number-frequency relation shifted markedly from the simple acoustic or electro-optic match. The results of the calculations are presented graphically in Figs. 3–5. Figure 3 is a plot of the temporal growth rate versus the speed of propagation of the unstable mode, for a fixed laser intensity and various values of the optical absorptivity α. Figure 4 expresses the relationship between the spatial gain and the propagation speed of the acoustically driven instabilities, for a given α and a number of power flux densities P_L in MW/cm². In Fig. 5 we exhibit the spatial gain behavior due to the electro-optic effect augmented at the lower speeds by the gain resulting from the body forces. For k a number of values has been chosen in the vicinity of that for the optimum index matched electro-optic mode. As the group velocity approaches the speed of light in the medium, the contributions due to the acoustic processes drop out and we are left with the pure Kerr gain given by Eq. (33).

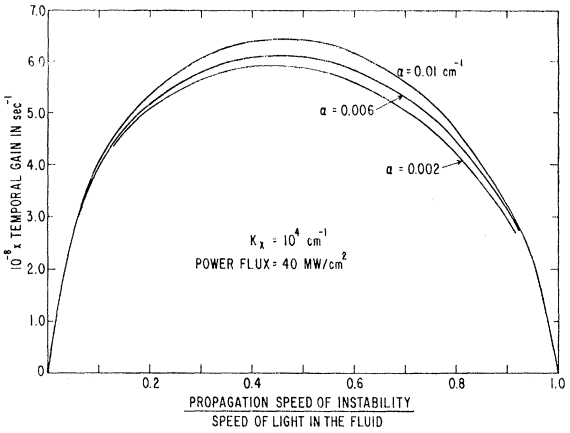

FIG. 3. Temporal gain in carbon disulfide as a function of group velocity for the acoustic regime.

Comparison of Figs. 4 and 5 at $P_L = 40$ MW/cm² shows that the gains for the acoustic and acousto-electro-optic instabilities become equal for $v_g = 1.2\times10^9$ cm/sec. Denoting the cell length by L and the laser pulse time by Δt, we can, therefore, draw the important conclusion that the purely acoustic instability predominates when $\Delta t > 0.8L$ nsec, while the mixed mode yields largest over-all gain when $\Delta t < 0.8L$ nsec. Finally, of course, for $\Delta t = (n_0/c)L$ sec the gain is just that for the index matched mode as given by Eq. (33). Thus, it is clear that the over-all gain will depend strongly on the duration Δt of the laser pulse. More specifically, the total gain for the acoustical mode is from Eq. (44) equal to

$$1.6\times10^3 (P_L L \Delta t)^{1/2},$$

or 5 e foldings for $\Delta t = 25$ nsec and $L = 10$ cm, always assuming that the linear theory continues to hold up to the end of the system.

B. Gases

The growth rates for gases are expected to be appreciably smaller than those for liquids, and to achieve

[28] C. Truesdell, J. Math. Mech. 2, 643 (1953).

a significant spatial gain it is necessary to employ a laser with a comparatively long pulse time ($\approx 100\ \mu$sec). Since high-intensity Q-switched lasers are now precluded, this means not only that the available power flux densities are limited to relatively low levels, but also that coherence lengths as long as 10 km become attainable.

The analysis of the dispersion relation for gases is simplified considerably by the fact that, because of the smallness of ϵ_0-1 and the virtual lack of viscous damping, the electrostrictive and viscous force densities may be neglected in (4). For gases, therefore, it follows from (27) that (26) reduces to

$$\omega(\omega^2-\Omega^2)=-i(k_L/c)\tau\Omega^2 v_s^2 \times[f(\omega,k_z)+f(-\omega,-k_z)], \quad (45)$$

where

$$f(\omega,k_z)=[\Omega^2+(2k_L v_s^2/c)(ck_z-\omega)]^{-1}, \quad (46)$$

and

$$\Omega=v_s k_x,$$

$$\tau=\frac{ck_L(\epsilon_0-1)(\gamma-1)}{\gamma p_{\text{amb}}}10^{13}\alpha P_L, \quad (47)$$

with P_L representing the power flux density in MW/cm^2 in the incident laser beam, and p_{amb} the ambient pressure defined by $\rho_0 C_p(\gamma-1)/\beta\gamma$; a result which is obtained by combining the ideal-gas law with the well-known result from thermodynamics: $C_p=C_v+Nk$, k being the Boltzmann constant. Because the Kerr constant for gases[29] is an exceedingly small quantity, the Kerr-type resonance condition that was adopted for liquids becomes replaced with the vanishing of the real part of the denominator in $f(\omega,k_z)$. It is only necessary to maximize one of the functions f in (45) as the other will then be small. Thus, for maximum temporal gain the independent parameter Ω is to be fixed such that

$$\Omega^2=(2k_L v_s^2/c)(\text{Re}\omega-ck_z), \quad (48)$$

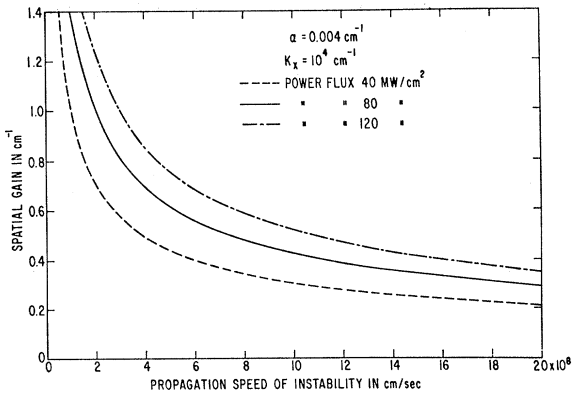

FIG. 4. Spatial gain in carbon disulfide as a function of group velocity for the acoustic regime.

[29] F. W. Quelle (private communication) has listed the optical Kerr constants for a number of gases.

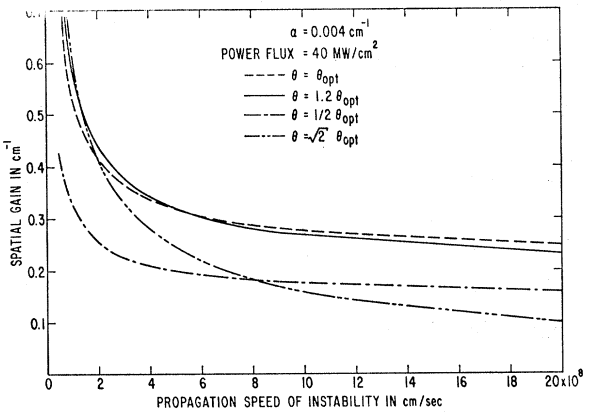

FIG. 5. Dependence of the spatial gain on $\theta=k_z/k_L$ in the vicinity of the Kerr-matched mode.

where Reω stands for the real part of ω. Subject to this restriction on Ω, there is the additional requirement that k_z has to satisfy

$$\frac{\partial}{\partial k_z}\text{Im}\omega=0,$$

where Imω is the imaginary part of ω. This problem can be conveniently solved by setting

$$\omega=\sigma e^{i\theta}, \quad (49)$$

and then optimizing $-$Imω with respect to θ. Substituting for ω in the dispersion relation and imposing condition (48) now yields the relations

$$\sigma^2\cos3\theta-\Omega^2\cos\theta=\tau\Omega^2/\sigma^2\sin\theta, \quad (50)$$

$$\sigma^2=\Omega^2\sin\theta/\sin3\theta, \quad (51)$$

where we have equated real and imaginary parts. The parameter Ω^2 in (50) can be eliminated with the aid of (51), and it follows that

$$\sigma^2=-\tau\sin3\theta/\sin\theta\sin2\theta, \quad (52)$$

and hence that

$$\Omega^2=-\tau(3-4\sin^2\theta)^2/\sin2\theta. \quad (53)$$

The functional dependence of Imω on θ is, therefore, given by

$$\text{Im}\omega=-(\tfrac{1}{2}\tau)^{1/2}(-\sin3\theta/\cos\theta)^{1/2}. \quad (54)$$

The corresponding expression for Reω follows directly from (49). Since Ω is physically restricted to remain finite, $-$Imω attains its maximum value at $\theta=\theta_0$, where θ_0 satisfies the equation

$$\tan3\theta_0\tan\theta_0+3=0, \quad (55)$$

the solutions of which are

$$\theta_0=\pm\tan^{-1}(-3\pm2\sqrt{3})^{1/2}. \quad (56)$$

Evidently, the only solution leading to temporal gain is $\theta_0=-0.60$ rad.

The spatial gain is $-\mathrm{Im}\omega(\theta_0)/v_g$, and v_g—the "group" velocity of the unstable mode—will now be calculated in accordance with the procedure outlined in Sec. III. The preliminary remark should be made here that in differentiating (45) with respect to k_z, Ω is to be regarded as a constant parameter whose optimum value is fixed by the right-hand side of (53) evaluated at $\theta=\theta_0$. Performing the differentiation and invoking (48) yields, after setting $\partial\,\mathrm{Im}\omega/\partial k_z=0$ and equating imaginary parts, that

$$v_g=c/(1+3\sin^2\theta_0). \tag{57}$$

A simplification was achieved here by utilizing (52) and (53). Thus, we deduce the general result that for a given gas the spatial gain varies as the square root of the product of the power flux density and the absorption coefficient.

To illustrate, consider the case of air at 1 atm for which[28] $\epsilon_2\approx 2\times10^{-16}$ esu, $\epsilon_0-1=5.8\times10^{-4}$, $v_s=3.3\times10^4$ cm/sec, $\gamma=1.4$, and $p_{\mathrm{amb}}=10^6$ dyn/cm^2. Equations (49), (53), and (54) predict that for the mode growing fastest in time[30]

$$\omega=(2.8-i1.9)10^9(\alpha P_L)^{1/2}, \tag{58}$$

corresponding to the wave numbers $k_x=13$ cm^{-1} and $k_z=-9\times10^{-4}$ cm^{-1}. The velocity with which the peak of the instability convects through the medium is from (57) given by $v_g\approx\frac{1}{2}c$. The maximum spatial gain is, therefore, approximately equal to 10^{-5} cm^{-1} for a power flux density, absorption coefficient product of 10^{-8} MW/cm^3, provided the interaction length L, say, is infinite as far as this unstable mode is concerned, i.e., provided the laser pulse time $\Delta t<2L/c$. If this inequality is not satisfied, the peak of the temporal instability moves beyond the limits of the system during the interaction time, and a larger over-all gain may be realized by an unstable mode whose propagation speed is such that it reaches the end of the system in time Δt. We will now calculate the maximum gain for this case by the procedure set forth in Sec. III. Since the Kerr constant for gases is exceedingly small, we ignore it and, retaining one function f, as before, write Eq. (45) as

$$rk_z=-q+s\omega-i/\omega(\omega^2-\Omega^2), \tag{59}$$

where

$$\begin{aligned} q&=c/2k_Lv_s^2\tau,\\ r&=(c/\Omega^2\tau)(1-i\alpha/2k_L),\\ s&=(\Omega^2\tau)^{-1}. \end{aligned} \tag{60}$$

To derive the dependence of the growth rate on the group velocity, we make the transformation

$$\omega=\omega'+vk_z, \tag{61}$$

and impose the saddle-point condition $(\partial\omega'/\partial k_z)=0$. Thus, ω must satisfy

$$y^3+y^2-3i\lambda y-2i\lambda=0, \tag{62}$$

with

$$\omega^2=\Omega^2(1+y),$$

$$\lambda=\frac{\tau}{\Omega^2}\left[\frac{c}{v}\left(1-i\frac{\alpha}{2k_L}\right)-1\right]^{-1}. \tag{63}$$

The damping term in λ has been retained to avoid a singularity at $v=c$. Once (62) has been solved for y (and hence for ω) over a range of v, the corresponding values of the saddle points ω_s' are determined by (61), k_z being given by (59). For small λ, Eq. (62) can be solved by substituting for y a power series in $i\lambda$ and equating the coefficients of equal powers in λ. Selecting the root predicting maximum gain, we obtain to $O(\lambda^3)$ that

$$y=-1+\lambda^2-i\lambda. \tag{64}$$

The corresponding values for ω and k_z are given by (63) and (59), respectively. Carrying out the algebra, we find for the spatial gain

$$\frac{-\mathrm{Im}\omega_s'}{v}=\frac{1}{c}\left[2\left(\frac{c}{v}-1\right)\tau\right]^{1/2}[1+\tfrac{1}{2}\lambda-\tfrac{3}{8}\lambda^2+O(\lambda^3)]. \tag{65}$$

This expression is valid for $\lambda\ll1$, which by (63) corresponds to the inequality

$$k_x^2(c/v-1)\gg5.51\times10^9\alpha P_L, \tag{66}$$

for air at 1 atm. Thus, for $\alpha P_L=10^{-8}$ MW/cm^3 and $k_x>10$ cm^{-1}, (65) is a good approximation to the spatial gain up to $v\simeq\frac{2}{3}c$ as is verified by Figs. 6 and 7, which exhibit the behavior of the exact solutions for $\alpha P_L=10^{-8}$ MW/cm^3. In particular, Fig. 7 substantiates the conclusion based on (65) that for k_x sufficiently large or v small the gain approaches a limiting value which is independent of k_x. For the slowly moving modes, i.e., v small, we deduce that the number of e foldings in a

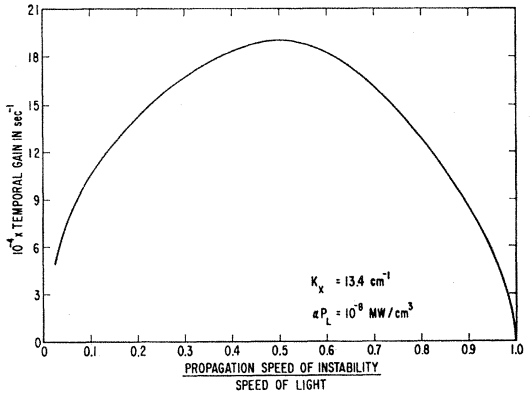

FIG. 6. Temporal gain in gases as a function of group velocity.

[30] These results have been verified by the detailed numerical computations of S. S. Rangnekar (private communication).

distance L is approximately

$$\frac{\mathrm{Im}\omega_s'}{v}L \simeq L\left(\frac{2\tau}{cv}\right)^{1/2} = 4.47\times 10^6\left[\frac{(\epsilon_0-1)(\gamma-1)}{\gamma p_{\mathrm{amb}}}k_L L\alpha P_L \Delta t\right]^{1/2}, \quad (67)$$

where Δt is the pulse time of the incident laser beam.

As an example, let the cell length be 10 km. Since the propagation speed of the fastest growing temporal mode is $\frac{1}{2}c$ with $k_x=13.4$ cm^{-1}, a disturbance growing according to this mode spends 70 μsec in the cell. If Δt is less than this critical value, the cell is essentially infinite and the gain is, from (58) or Fig. 6, given by $(1.9\Delta t)$ 10^5 e foldings. For $\Delta t>70$ μsec the maximum gain is $\exp(g\times 10^6)$, where g is the spatial gain determined from (65) or Fig. 7 with $v=10^6/\Delta t$. For instance, if $\Delta t=200$ μsec the optimum speed $v=0.16c$ and the total gain is 27 e foldings. We note from inequality (66) that this result is relatively insensitive to changes in k_x, provided $k_x>5$ cm^{-1}.

To sum up, the foregoing indicates that as a result of the laser-induced instabilities, breakdown in the mode structure of the laser beam can occur in distances of the order of a few kilometers if we adopt the criterion that a few e foldings are necessary for the perturbations to attain significant levels.

V. CONCLUSIONS

The linearized macroscopic theory developed in this paper shows that the induced waves generated by the coupling of a laser beam to the free modes of a fluid can, in the presence of fluctuations in the laser intensity or in the thermodynamical properties of the medium, lead to the establishment of rapidly growing spatial and temporal instabilities. The results indicate that stimulated Raman scattering plays a relatively minor role in the initial stages of the instability growth, even for a Stokes conversion rate as high as a few percent, and that the main contributions to such growth arise from thermal-energy deposition, electrostriction, and the optical Kerr effect. We would also suggest that it is the breakup of the mode structure caused by the instability development that is ultimately responsible for the random distribution of the optical filaments observed by Garmire, Chiao, and Townes.[3] In gases, the effect of electrostriction is negligible and the strength of the instability is governed mainly by thermal-energy deposition. While the growth rates are much smaller than those in liquids, mode breakup should also occur in these media, albeit at a considerable distance down the laser beam.

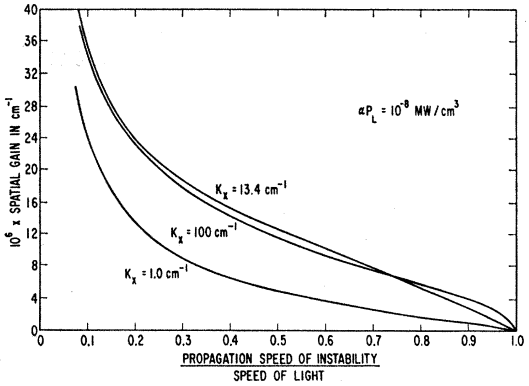

FIG. 7. Spatial gain in gases as a function of group velocity.

The actual distance at which the perturbations attain significant values depends, of course, on the magnitude and dispersive character of the inhomogeneities acting as sources for the instability development. The detailed treatment of this problem, which would entail replacing the δ function assumed for s_u in (19) by a more realistic function and actually evaluating the residue of the integrand in (20) at the saddle point, lies outside the scope of this paper. The subject of our inquiry has instead been the more general question of determining the nature of the instabilities induced by the laser in the fluid, and their initial growth rates.

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