

**OPTICAL PHYSICS** 

## Stimulated Raman and Brillouin scattering, nonlinear focusing, thermal blooming, and optical breakdown of a laser beam propagating in water

B. HAFIZI,<sup>1,\*</sup> J. P. PALASTRO,<sup>1</sup> J. R. PEÑANO,<sup>1</sup> T. G. JONES,<sup>1</sup> L. A. JOHNSON,<sup>1</sup> M. H. HELLE,<sup>1</sup> D. KAGANOVICH,<sup>1</sup> Y. H. CHEN,<sup>2</sup> AND A. B. STAMM<sup>1</sup>

<sup>1</sup>Naval Research Laboratory, Washington, DC 20375, USA <sup>2</sup>Research Support Instruments, 4325-B Forbes Blvd., Lanham, Maryland 20706, USA \*Corresponding author: bahman.hafizi@nrl.navy.mil

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The physical processes associated with propagation of a high-power laser beam in a dielectric include selffocusing, stimulated Raman scattering, stimulated Brillouin scattering, thermal blooming, and multiphoton and collisional ionization. The interplay between these processes is analyzed using a reduced model consisting of a few differential equations that can be readily solved, enabling rapid variation of parameters and the development of theoretical results for guiding new experiments. The presentation in this paper is limited to propagation of the pump, the Stokes Raman, and the Brillouin pulses, ignoring the anti-Stokes Raman. Consistent with experimental results in the literature, it is found that self-focusing has a dramatic effect on the propagation of high-power laser beams in water. A significant portion of the pump laser energy is transferred to Stokes Raman forward scatter along with a smaller portion to Brillouin backscatter.

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## **1. INTRODUCTION**

A number of physical processes accompany the propagation of high-power laser beams in dielectrics and, in particular, in water [1–7]. These include self-focusing, stimulated Raman scattering and stimulated Brillouin scattering, thermal blooming, and multiphoton and collisional ionization. Self-focusing can play a critical role in the interplay between these and leads to complex spatio-temporal evolution of the beam. Herein a novel model of all these processes is presented, discussed, and applied to the analysis of experimental results existing in the literature. Self-consistent propagation simulations show behavior that is similar to that in experiments and predict a physical process—gain focusing [8]—that may be observable in future experiments.

The propagation of ~megawatt-class, ~nanosecond-pulse length laser beams in water has been studied in Refs. [5,7]. The self-focusing power  $P_{Lc}$  in water is 1.87 MW (at 532 nm). The experiments in Ref. [5] reported the observation of strong Stokes Raman radiation and no measurable anti-Stokes component. This experiment was dominated by self-focusing. In Ref. [7] (at very similar laser power and energy levels) Stokes Raman radiation was measured along with Brillouin backscatter, while self-focusing is said to be insignificant. In Ref. [7], an analysis of the experiment based on a reduced model is presented. An important finding of Ref. [7]—corroborated by their analysis—is that the efficiency of scattering into the Raman and Brillouin waves is sensitive to water temperature. Interestingly the efficiency of conversion into the Raman Stokes waves was largest at  $\approx 4^{\circ}$ C, the temperature at which water has its maximum density and its thermal-expansion coefficient vanishes. This suggests that thermal blooming is integral to these experiments and must be incorporated in any realistic model. The research presented here was initiated to analyze the results in Ref. [5] in light of these experimental observations. The model includes multiphoton and collisional ionization processes as well as attachment, which are believed to be important in the experiments of Ref. [5], and is therefore more comprehensive than the model in Ref. [7].

One shortcoming of the reduced models (in Ref. [7] and here) is the approximate manner in which temporal dependence and finite pulse length effects are incorporated (see Section 2). However, the simplified models serve as important tools that can complement time-consuming, full-scale, laboratory-frame simulations (3D + time) of ~meter-long water tank experiments or propagation through open water, including the ocean. A further approximation connected with the analysis here is that all radiation fields have a Gaussian transverse profile. This can be a poor approximation if instabilities or dispersion tend to hollow out the pump beam significantly (Section 5).

## 2. PROPAGATION EQUATIONS

The presentation is limited to the case in which the anti-Stokes component is not appreciably excited. Inclusion of the anti-Stokes component is straightforward but was not considered necessary since it was not appreciably excited in the experiments of Ref. [5]. The linearly polarized electric field is expressible as

$$E(z, t) = \frac{1}{2}A_L(z)e^{i\varphi_L(z,t)} + \frac{1}{2}A_S(z)e^{i\varphi_S(z,t)} + \frac{1}{2}A_B(z)e^{i\varphi_B(z,t)} + \text{c.c.},$$
(1)

where the suffixes *L*, *S*, and *B* refer to the pump laser, the Stokes component of Raman forward scatter, and the Brillouin backscatter, respectively;  $k_j$ ,  $\omega_j$  are the wavenumber and frequency, respectively; and  $A_j$  is the amplitude for j = L, *S*, *B*. For j = L, *S* the phase is  $\phi_j \equiv k_j z - \omega_j t$ , while  $\phi_B \equiv -k_B z - \omega_B t$  is the phase of the backscattered Brillouin wave. The Raman scattering process of interest is attributed to the symmetric stretching of O–H bonds and to the hydrogen bond network in liquid water, manifested as a broad band at ~3400 cm<sup>-1</sup> and vibrational frequency ~100 THz [9], driven by a force that is proportional to the time-average of the electric field squared. Making use of Eq. (1), the relevant terms that drive Stokes Raman forward scatter are

$$\langle E(z,t)^2 \rangle_R = \frac{1}{2} A_L A_S^* e^{i(\varphi_L - \varphi_S)} + \text{c.c.};$$
 (2)

the Brillouin backscatter is mediated by an acoustic mode (frequency  $\sim$ 5 GHz) [9] and driven by

$$\langle E(z,t)^2 \rangle_B = \frac{1}{2} A_L A_B^* e^{i(\varphi_L - \varphi_B)} + \frac{1}{2} A_S A_B^* e^{i(\varphi_S - \varphi_B)} + \text{c.c.}$$
 (3)

Brillouin backscatter can be initiated by either the pump laser or the Stokes Raman excitation. The frequency of the Brillouin excitation is different in the two cases; as a first approximation it is permissible to consider the case in which the pump laser is the predominant driver of Brillouin backscatter. Quantitative validation of this is presented in Section 5. The acoustic response due to the beating of the pump laser and the backscattered radiation is obtained via the fluid equations as shown in Appendix A [9–11]. In a coordinate system traveling with laser pulse group velocity  $v_g$  (i.e., independent variables z,  $\tau = t - z/v_g$ ), the propagation equations are

$$\begin{bmatrix} \nabla_{\perp}^{2} + 2ik_{S}\frac{\partial}{\partial z} + i\frac{\omega_{S}^{2}\epsilon''(\omega_{S})}{c^{2}} \end{bmatrix} A_{S}$$

$$= -\frac{\omega_{S}^{2}}{c^{2}}\frac{\partial\epsilon(\omega_{S})}{\partial T}T'A_{S}$$

$$-\frac{6\pi\omega_{S}^{2}}{c^{2}}\left\{ [\chi_{R}(\omega_{S}) + \chi_{NR}]|A_{L}|^{2} + \frac{1}{2}\chi_{NR}|A_{S}|^{2} \right\} A_{S}$$

$$+ \left[ \frac{\omega_{P}^{2}}{c^{2}} \left( 1 - \frac{i\nu_{e}}{\omega_{S}} \right) - \frac{8\pi i\omega_{S}}{c^{2}}\frac{U_{\text{ion}}}{|A_{S}|^{2}} \left( \frac{\partial N_{e}}{\partial \tau} \right)_{S} \right] A_{S}, \quad (4)$$

$$\begin{bmatrix} \nabla_{\perp}^{2} - 2ik_{B}\frac{\partial}{\partial z} + i\frac{\omega_{B}^{2}\epsilon''(\omega_{B})}{c^{2}} \end{bmatrix} A_{B}$$

$$= \frac{\omega_{B}^{2}}{c^{2}}\kappa_{SBS}^{*}|A_{L}|^{2}A_{B} - \frac{6\pi\omega_{B}^{2}}{c^{2}}\chi_{NR}\left(|A_{L}|^{2} + \frac{1}{2}|A_{B}|^{2}\right)A_{B}$$

$$+ \left[\frac{\omega_{p}^{2}}{c^{2}}\left(1 - \frac{i\nu_{e}}{\omega_{B}}\right) - \frac{8\pi i\omega_{B}}{c^{2}}\frac{U_{ion}}{|A_{B}|^{2}}\left(\frac{\partial N_{e}}{\partial \tau}\right)_{B}\right]A_{B}, \quad (5)$$

$$\begin{bmatrix} \nabla_{\perp}^{2} + 2ik_{L}\frac{\partial}{\partial z} + i\frac{\omega_{L}^{2}\epsilon''(\omega_{L})}{c^{2}} \end{bmatrix} A_{L} \\ = -\frac{6\pi\omega_{L}^{2}}{c^{2}} \left\{ [\chi_{R}(\omega_{a}) + \chi_{NR}] |A_{S}|^{2} + \frac{1}{2}\chi_{NR} |A_{L}|^{2} \right\} A_{L} \\ + \frac{\omega_{L}^{2}}{c^{2}} \kappa_{SBS} |A_{B}|^{2} A_{L} - \frac{\omega_{L}^{2}}{c^{2}} \frac{\partial\epsilon(\omega_{L})}{\partial T} T' A_{L} \\ + \left[ \frac{\omega_{P}^{2}}{c^{2}} \left( 1 - \frac{i\nu_{e}}{\omega_{L}} \right) - \frac{8\pi i\omega_{L}}{c^{2}} \frac{U_{ion}}{|A_{L}|^{2}} \left( \frac{\partial N_{e}}{\partial \tau} \right)_{L} \right] A_{L}, \quad (6)$$

where  $\nabla_{\perp}^2$  is the transverse Laplacian operator,

$$\chi_R(\omega_S) = \frac{(N_{\rm H_2O}/6M_r)(\partial p/\partial r_{\rm v})^2}{\omega_{\rm v}^2 - (\omega_L - \omega_S)^2 + 2i\gamma_R(\omega_L - \omega_S)}$$
(7)

is the Raman susceptibility [9],  $\omega_v$  is the vibrational frequency, at resonance  $\omega_v = \omega_L - \omega_S$ ,  $\omega_a = \omega_L + \omega_v$  is the anti-Stokes frequency,  $N_{\rm H_2O}$  is the number density of water molecules,  $M_r$ is the reduced nuclear mass,  $\partial p / \partial r_v$  is the derivative of the molecular polarizability with respect to the O–H bond length, and  $\gamma_R$  is the relevant damping constant. Further,

$$\kappa_{\text{SBS}} = \frac{\text{Re}(\epsilon/\mu)\alpha_1(\alpha_1/2 - i\alpha_2\beta cc_s^2/\Omega c_p)}{8\pi\rho_0(\Omega^2 + i\Gamma_B\Omega - c_s^2q^2)}q^2 \qquad (8)$$

is the Brillouin backscatter susceptibility, measuring the strength of pump-Brillouin coupling in the limit where Rayleigh scattering is unimportant  $(\Omega \gg \gamma \chi q^2); \Omega =$  $\omega_L - \omega_B$  and  $q = k_L + k_B$  are the acoustic frequency and wavenumber, respectively; the Brillouin linewidth is  $\Gamma_B =$  $q^2[\zeta/\rho_0 + \frac{4}{3}\nu + \kappa(\gamma - 1)/c_p]$  [9];  $\chi = \kappa/\rho_0 c_p$  is the thermometric conductivity;  $\kappa$  is the thermal conductivity;  $c_p$  and  $c_v$ are the specific heats at constant pressure and constant volume, respectively;  $\gamma = c_p/c_v$  ( $c_p \approx c_v$ ,  $\gamma \approx 1$  for water);  $\nu$  is the kinematic viscosity;  $\zeta$  is the second (bulk) viscosity;  $\rho_0$  is the ambient density;  $c_s$  is the sound speed;  $\beta$  is the thermalexpansion coefficient;  $\alpha_1 = \rho (\partial \epsilon / \partial \rho)_T / \text{Re}[\sqrt{(\epsilon/\mu)^*}]$  is proportional to the electrostriction coefficient;  $\alpha_2 =$  $(\omega/c)$ Im  $\epsilon/\text{Re}[\sqrt{(\epsilon/\mu)^*}]$  is the absorption coefficient;  $\epsilon(\omega_i) \equiv$  $\epsilon'(\omega_i) + i\epsilon''(\omega_i)$  defines the real and imaginary parts of the permittivity; and  $\mu \approx 1$  is the permeability.

The model for thermal blooming employed here is similar to that in Ref. [7]—justified by the close agreement between their simulations and experimental observations for the efficiency of conversion into Stokes Raman waves. A plausible model can be constructed by considering the various time scales associated with thermal blooming. The decay of elastic stresses in a fluid occurs on the Maxwellian relaxation time scale  $\tau_M \sim \eta_d/G$ , where  $\eta_d \equiv \rho_0 \nu$  is the dynamic viscosity and G is the modulus of rigidity [11]. For water  $\eta_d \approx 10^{-3}$  Pa-s,  $G \approx 2 \times 10^9$  Pa, resulting in a relaxation time  $\tau_M \sim 1/2$  ps. In other words,  $\tau_M \gg 1/\omega_v \ (\omega_v/2\pi \approx 100 \text{ THz})$ , and, thus, with respect to Raman-induced vibrations of molecules, water behaves as a rigid, amorphous solid and is not described by the usual equations of fluid mechanics. Further, vibrations of the moleculesthe cause of the temperature rise in water-equilibrate on the sub-picosecond time scale [12]. Refractive index changes are assumed to occur on the same time scale, as suggested by the results of Ref. [7], although the physical mechanisms for this are not well understood. The terms proportional to the perturbed temperature T' in Eqs. (4) and (6) drive thermal blooming of the Stokes Raman and pump beams, respectively; Brillouin beam heating as a driver of thermal blooming is negligible because of its much smaller amplitude. With the ordering of time scales given above, an expression for the perturbed temperature T' can be obtained as in Appendix B [10,13]; the Lorentz–Lorenz formula may be used to obtain  $\partial \epsilon(\omega)/\partial T =$  $-\beta(\epsilon-1)(\epsilon+2)/3.$ 

The terms proportional to  $\alpha_1^2$  and to  $\alpha_1 \alpha_2$  in Eq. (8) are the electrostrictive and absorptive contributions to the stimulated Brillouin scattering susceptibility, respectively. Employing the expression  $n_j = ck_j/\omega_j [\approx \sqrt{\epsilon'(\omega_j)}]$  for the refractive index of the electromagnetic waves, the frequency  $\Omega_B$  of the sound wave for direct backscattering is given by  $\Omega_B = 2\omega_L/(c/n_Lc_s + 1)$ . One then finds that the electrostrictive contribution in Eq. (8) peaks at  $\Omega = \Omega_B$  and  $q = \Omega_B/c_s$  while the absorptive contribution is maximum for  $\Omega = \Omega_B + \Gamma_B/2$ .

In writing Eqs. (4)–(6), a real-valued susceptibility  $\chi_{NR}$  has been introduced to represent various non-resonant (reactive) effects [14]. These equations include, in addition, the effects of plasma formed by ionization of water, as well as laser energy depletion arising from Joule heating and multiphoton ionization of water molecules. The first of these is represented by the penultimate term on the right-hand side of Eqs. (4)–(6), where the plasma frequency is given by  $\omega_p^2 = 4\pi N_e |e|^2/m$ , e, m are the electronic charge and mass, respectively,  $N_e(r, z, \tau)$ is the free-electron density,  $\nu_e = N_{\rm H_2O} v_{\rm rms} \sigma_c$  is the collision frequency,

$$\mathbf{v}_{\rm rms}(r, z, \tau) = \frac{|e|}{\sqrt{2}m} \sqrt{\frac{|A_L|^2}{\omega_L^2} + \frac{|A_S|^2}{\omega_S^2} + \frac{|A_B|^2}{\omega_B^2}} \qquad (9)$$

is the root-mean-square electron oscillation velocity in the optical field, and  $\sigma_c$  is the collision cross section and it is assumed that  $\nu_e/\omega_j \ll 1$  (j = L, S, B). The terms proportional to  $\nu_e/\omega_j$ represent inverse Bremsstrahlung losses. The loss of photons in the process of multiphoton ionization is represented by the last term on the right-hand side of Eqs. (4)–(6), where  $U_{\rm ion}$  is the ionization potential of water molecules and the suffixes L, S, and B on the time derivative of the density indicate the change in electron density due to absorption of pump, Stokes Raman, and Brillouin photons, respectively.

In deriving Eqs. (4)–(6) from the wave equation, a number of simplifying assumptions have been made. Foremost amongst these is the neglect of several terms involving time derivatives  $\partial^n/\partial \tau^n$ , n = 1, 2, ..., on the left-hand sides. There are several causes for time variation of the amplitudes  $A_j$ . The finite duration of the laser pulse is one source of temporal variation that is imposed on all the amplitudes—the others being the temporal variations due to ionization and to thermal blooming.

For nanosecond-long laser pulses in a typical laboratory-scale environment, the pulse length is comparable to the length of the water tank and one can study the development of a single, short temporal slice in the body of the pulse as representative of the pulse as it propagates along the tank. Since the pump laser and the Stokes Raman waves travel with nearly the same velocity, the same applies to any co-propagating temporal slice of the Stokes Raman pulse. For these waves the terms on the left-hand side of Eqs. (4) and (6) associated with finite pulse length and group velocity dispersion [15] can legitimately be neglected at the lowest order. Since the Brillouin backscatter is counter-propagating (i.e., traveling upstream) it cannot be treated in the same manner. Its temporal variation can be approximately accommodated if the explicitly time-dependent terms on the right-hand sides of Eqs. (4)–(6) can be shown to be relatively small. This must be verified in each particular case.

# 3. MULTIPHOTON AND COLLISIONAL IONIZATION

The free-electron density in water can change because of ionization, recombination, and attachment processes. The rate equation for electron density is

$$\frac{\partial N_e}{\partial \tau} = W_{\rm MPI} N_{\rm H_2O} + \nu_i N_e - \eta N_e - \beta N_e^2, \qquad (10)$$

where  $W_{\rm MPI}$  is the multiphoton ionization rate,  $\nu_i$  is the collisional ionization rate,  $\eta$  is the electron attachment rate, and  $\beta$  is the radiative-recombination rate. Equation (10) assumes that the degree of ionization is small (i.e.,  $N_e \ll N_{\rm H_2O}$ ).

The multiphoton ionization rate is

$$W_{\rm MPI} = \frac{2\pi}{(\ell-1)!} \sum_{j=L,S,B} \omega_j \left[ \frac{I_j(r,z,\tau)}{I_{\rm MPI}} \right]^{\ell}, \qquad (11)$$

where  $\ell$  is an integer denoting the number of photons required for ionization, i.e.,  $\ell = \text{Int}[U_{\text{ion}}/\hbar\omega + 1]$ ,  $\hbar\omega$  is the photon energy,  $I_j(r, z, \tau)$  is the intensity (j = L, S, B), and  $I_{\text{MPI}}$  is the characteristic ionization intensity. It is assumed that the same number of photons is required to ionize a water molecule irrespective of whether it is a pump wave, Stokes Raman wave, or Brillouin backscatter wave. Additionally it is assumed that any given multiphoton ionization event is affected entirely by laser photons, by Stokes Raman photons, or by Brillouin backscatter photons independently.

The avalanche ionization rate  $\nu_i$  is

$$\nu_i(r, z, \tau) = \frac{\nu_e}{U_{\rm ion}} \frac{|e|^2}{2m} \left( \frac{|A_L|^2}{\omega_L^2} + \frac{|A_S|^2}{\omega_S^2} + \frac{|A_B|^2}{\omega_B^2} \right).$$
(12)

When the degree of ionization is relatively small  $(\beta N_e \ll \nu_i)$  recombination can be ignored [16] and the solution of Eq. (10) is

$$N_{e}(r, z, \tau) = N_{\mathrm{H}_{2}\mathrm{O}} \int_{0}^{\tau} \mathrm{d}\tau' W_{\mathrm{MPI}}(r, z, \tau')$$
$$\times \exp\left[-\eta(\tau - \tau') + \int_{\tau'}^{\tau} \mathrm{d}\tau'' \nu_{i}(r, z, \tau'')\right].$$
(13)

Additionally it is assumed that hydrated electrons [17] do not fundamentally alter the physics because the pump wavelength is not strongly resonant.

## 4. ANALYSIS OF PROPAGATION EQUATIONS

Analysis of optical propagation, stimulated Raman scattering, stimulated Brillouin scattering, thermal blooming, and ionization proceeds by substituting self-similar forms for the complex-valued amplitudes into the propagation equations (4)–(6) and obtaining equations for the spot radius and power. The approach employed here is to make use of the source-dependent expansion (SDE) method [18], assuming that the field amplitudes are peaked on axis and have Gaussian forms

$$A_{i}(r,z) = B_{i}(z)e^{i\theta_{j}(z)}e^{-[1+i\alpha_{j}(z)]r^{2}/R_{j}^{2}(z)},$$
(14)

where  $B_j$  is the amplitude,  $\theta_j$  is the phase,  $R_j$  is the spot radius, and  $\alpha_j$  is related to the curvature of the wavefronts for the *j*th wave (*j* = *L*, *S*, *B*).

Electrons are initially  $(\nu_i \tau \ll 1)$  generated by multiphoton ionization. From Eqs. (9)–(13) it follows that the radial dependence of plasma density is  $N_e(r, z, \tau) \propto \exp(-2\ell r^2/R^2)$ since the multiphoton ionization rate is proportional to  $I^{\ell}$ . Thus for  $\ell \gg 1$  the electron density is highly peaked compared to the radial variation of the optical field. In performing the radial integrals in SDE it is permissible to evaluate Eq. (12) at r = 0 since the electrons generated by multiphoton ionization are the seed from which avalanche ionization proceeds. Validity of this approximation requires that  $R_L \sim R_S \sim R_B$ . Following the SDE method, making use of the definition of refractive index  $n_j = ck_j/\omega_j$ , the spatial variation of power  $P_j = cn_j R_j^2 B_j^2/16$  in the beams is given by

$$\frac{1}{P_j}\frac{\partial P_j}{\partial z} = 2F_j'', \qquad (j = L, S, B),$$
(15)

and that of the spot radius is given by

$$\frac{\partial^2 R_j}{\partial z^2} + \frac{1}{R_j} \frac{\partial}{\partial z} (R_j^2 G_j'') + \frac{4}{k_j^2 R_j^3} \left[ -1 - k_j R_j^2 G_j' + \frac{1}{4} (k_j R_j^2 G_j'')^2 \right] = 0, \quad (j = L, S),$$
(16a)

$$\frac{\partial^2 R_B}{\partial z^2} + \frac{1}{R_B} \frac{\partial}{\partial z} (R_B^2 G_B^{\prime\prime}) + \frac{4}{k_B^2 R_B^3} \left[ -1 + k_B R_B^2 G_B^{\prime} + \frac{1}{4} (k_B R_B^2 G_B^{\prime\prime})^2 \right] = 0.$$
 (16b)

Explicit forms for the functions F'', G', and G'' are given in Appendix C.

Equation (16) has the form of an envelope equation for the spot radius. Equations of this form are encountered in numerous applications describing electrons beams, radiation beams in free-electron lasers, etc. The -1 in square brackets is associated with vacuum diffraction. Referring to Eqs. (C12)–(C14), the term  $-k_j R_j^2 G'_j$  is associated with self- and cross-focusing as well as thermal blooming and refraction from plasma. These are all refractive effects. The term  $+\frac{1}{4} (k_j R_j^2 G'_j)^2$ , which is associated with the transfer of energy from the pump to the Stokes forward Raman wave and to the Brillouin backscatter, is positive definite and is always focusing. Gain guiding in lasing media—wherein the optical field is replenished by emission from the

gain medium—is well known. In the present context, however, the pump laser beam, for example, can experience guiding even as it transfers energy to the Stokes Raman wave (see Section 5).

Noting that  $\chi_R(\omega_S) = \chi_R^*(\omega_a)$  and making use of Eq. (15) and the definitions in Eqs. (C9)–(C11), the following Manley–Rowe-type relation is satisfied:

$$\frac{1}{\omega_L}\frac{\partial P_L}{\partial z} + \frac{1}{\omega_S}\frac{\partial P_S}{\partial z} - \frac{1}{\omega_B}\frac{\partial P_B}{\partial z} = 0,$$
 (17)

provided linear absorption losses and ionization are negligible. More generally pump laser power is transferred to the Stokes Raman wave and the Brillouin backscatter along with an additional power loss in water appearing in the form of vibrational energy of water molecules and of sound waves. If *Q* denotes the power dissipated in water, it follows that

$$\frac{\partial Q}{\partial z} = \frac{96\pi}{n_S n_L c^2} \frac{\mathrm{Im} \chi_R(\omega_a)}{R_L^2 + R_S^2} P_S P_L(\omega_L - \omega_S) + \frac{16}{n_B n_L c^2} \frac{(-\mathrm{Im} \kappa_{\mathrm{SBS}})}{R_L^2 + R_B^2} P_B P_L(\omega_L - \omega_B).$$
(18)

The second term on the right-hand side is due to the transformation of energy into sound waves by the Brillouin mechanism. This relatively small energy is eventually dissipated into heat; for parameters of interest here this happens on a time scale  $2\pi/\Gamma_B \ge 1$  ns.

### 5. COMPARISON WITH EXPERIMENT

The motivation for the foregoing analysis is to gain understanding of the experimental results in Ref. [5]. The measurements in Ref. [5] covered a range of pump laser powers,  $P_L = (10^{-4} - 5)P_{Lc}$ , where the self-focusing laser power  $P_{Lc}$  [defined after Eq. (C11)] in water is 1.87 MW (at 532 nm). The pump laser was focused at f-number = 310 in water. These experiments showed significant self-focusing and  $\geq 50\%$  power conversion into Raman forward scatter. In contrast self-focusing was stated to be negligible in Ref. [7] although the pump power reaches up to  $\approx 14P_{Lc}$ . Strong Raman scattering was observed, along with up to 30% (depending on water temperature) of the pump laser energy appearing as Brillouin backscatter. There were no diagnostics to detect Brillouin backscatter in Ref. [5].

Table 1 lists the parameters used to model an experiment similar to those in Refs. [5,7], noting that Eqs. (15) and (16) provide a highly reduced description. Linear absorption of electromagnetic energy is determined by the damping rate  $\alpha_2 \approx (\omega/nc) \text{Im} \epsilon$ . The discussion is limited to the case in which Brillouin backscatter is driven by the electrostrictive mechanism; i.e., it assumes that  $\alpha_1 \gg 2\alpha_2\beta cc_s^2/\Omega c_p$  in Eq. (8) [9]. In this case, i.e., when the Brillouin backscatter is dominated by the electrostrictive contribution,  $\kappa_{\text{SBS}}$  is a pure imaginary number.

The initial values for the field amplitudes—given in Table 1—correspond to the spontaneous emission levels given in the experiments in Ref. [1]. The spot radius for the pump is taken from Ref. [5]—and is assumed to equal the spot radii for the Stokes Raman and Brillouin backscatter.

Equations (15) and (16) apply to an experiment that is run in a steady-state regime wherein each slice of the laser pulse

Self-focusing power (at 532 nm) $P_{Lc}$	1.87 MW (Ref. [19])
Laser power at input $P_L$	$(\frac{1}{2}, 5) \times P_{Ic}$
Initial Stokes Raman, Brillouin power $P_S$ , $P_B$	$2.21 \times 10^{-5}$ W (Ref. [1])
Initial laser, Stokes Raman, Brillouin spot radius $R_L$ , $R_S$ , $R_B$	0.375 mm
Initial pump laser beam divergence	-0.6 mrad
Laser, Stokes Raman wavelength $\lambda_L$ , $\lambda_S$	532, 652 nm
Acoustic speed $c_s$	$1.5 \times 10^3$ m/s
Acoustic frequency $\Omega_B/2\pi$	7.4 GHz
Laser, Stokes Raman refractive index $n_L$ , $n_S$	1.334, 1.331
Stokes Raman susceptibility $\chi_R(\omega_S)$	$-i3.42 \times 10^{-14} \text{ cm}^3/\text{erg}$ (Refs. [7,9])
Brillouin susceptibility $\kappa_{SBS}$	$-i4.5 \times 10^{-13} \text{ cm}^3/\text{erg}$ (Refs. [7,9])
Brillouin linewidth $\Gamma_B$	$3.39 \times 10^9 \text{ s}^{-1}$ (Refs. [7,9])
Ionization potential of water $U_{\rm ion}$	6.5 eV (Refs. [20,21])
Characteristic multiphoton ionization intensity I <sub>MPI</sub>	$10^{14} \text{ W/cm}^2$ (Ref. [18])
Collision cross section $\sigma_c$	$10^{-15} \text{ cm}^2$ (Ref. [18])
Electron attachment rate $\eta$	$1.7 \times 10^{11} \text{ s}^{-1}$ (For reaction $e + \text{H}_2\text{O} \rightarrow \text{H} + \text{OH}^-$ in Ref. [16])
Thermal-expansion coefficient $\beta$ (at 20°C)	$2.07 \times 10^{-4} \text{ K}^{-1}$
Linear absorption coefficient $\alpha_2$	$2 \times 10^{-4} \text{ cm}^{-1}$ (Ref. [22])
Length of water tank	120 cm

 Table 1. Parameters for Simulations of Stimulated, Forward Stokes Raman Scattering and Brillouin Backscattering in

 Water

develops in an identical fashion as it propagates through the tank of water. Note that the laser used in Ref. [5] had a macro-pulse length ~6 ns with a temporal structure composed of micro-pulses of duration ~50 ps. Figures 1(a) and 1(b) show the fractional laser energy lost due to ionization of water molecules for  $P_L/P_{Lc} = 0.51$  and 5, respectively. [As shown in Fig. 2(b), the choice  $P_L/P_{Lc} = 0.51$  is of interest because it happens to be where Brillouin backscatter efficiency peaks.] For times  $\tau \ge 100$  ps [upper limit of integration in Eq. (13)] the electron density and the energy expended in ionization results hereafter pertain to a slice at  $\tau = 100$  ps, at which time single-slice, steady-state simulations are valid for both the



**Fig. 1.** Fractional laser energy used to ionize water as a function time in the laser pulse for (a) weak self-focusing  $P_L/P_{Lc} = 0.51$  and (b) strong self-focusing  $P_L/P_{Lc} = 5$ . Other parameters are listed in Table 1.

forward propagating Stokes Raman and the backward propagating Brillouin excitations. Simulations use an alternating direction scheme whereby the pump laser and Stokes Raman are alternatively advanced with Brillouin backscatter to their respective final states.

Care must be exercised in comparing the simulation results with Figs. 2–4 in Ref. [5] since the experimental observations are integrated over the duration of the laser pulse. This may be a possible explanation of the differing behavior of the pump and Stokes Raman powers observed in experiments versus that observed in the simulations. In particular, in the simulation there is always an initial distance wherein the Stokes Raman power is very small while growing exponentially. Experimental data, in which scattering from all time slices of the few-nanosecond pump pulse is integrated, does not exhibit such rapid exponential growth and conversion to Stokes Raman (Fig. 4 in Ref. [5]).

Figures 2(a)-2(d) show, in percentages, the conversion efficiency into Stokes Raman and Brillouin excitations, and



**Fig. 2.** Fractional laser energy converted into (a) Stokes Raman forward scatter, (b) Brillouin backscatter, (c) ionization, and (d) heating of water as a function of normalized pump power  $P_L/P_{Lc}$  after 100 ps into the pump pulse. Other parameters are listed in Table 1.



**Fig. 3.** Simulation of laser beam propagation with normalized power  $P_L/P_{Lc} = 0.51$ . (a) Normalized pump (blue) and Stokes Raman power (red), (b) spot radii of pump (blue) and Stokes Raman (red), (c) Brillouin backscatter power normalized to pump power, (d) spot radius of Brillouin backscatter, (e) electron density, and (f) pump (blue) and Stokes Raman (red) intensities, shown for slice at 100 ps in the pulse.

the energy expended in ionization and in heating of water, respectively, as functions of  $P_L/P_{Lc}$ . These figures show that the variation with  $P_L/P_{Lc}$  for all the plotted quantities is dominated by self-focusing, which begins to become important for  $P_L/P_{Lc} \ge 1/2$ . The peak in the Brillouin backscatter efficiency comes about for the following reasons. For  $P_L/P_{Lc} \leq$ 1/2 the laser intensity is too small to cause substantial excitation of the Brillouin mode. On the other hand, for  $P_L/P_{Lc} \gg 1/2$ , the conversion to Stokes Raman scattering is so strong that there is very little pump energy at the far end of the water tank from which Brillouin backscatter can be stimulated. This explanation is substantiated in Figs. 3 and 4, which show the variation of the pump laser, the Stokes Raman, and the Brillouin excitations along the length of the tank. Figures 3 and 4 also show the variation of the spot radii for the three electromagnetic waves, the intensities, and the corresponding electron densities along the tank.

The pump spot radius in Fig. 4(b) shows two locations where focusing takes place. The first focusing event at  $z \approx 16.5$  cm is due to the usual self-focusing phenomenon since the laser power  $P_L$  exceeds the critical power  $P_{Lc}$  for self-focusing. The pump laser pinches down to  $R_L \approx$ 221 µm at this location. The secondary focusing further downstream, at  $z \approx 43.5$  cm, is caused by the gain associated with the Raman process modifying the refractive index for the pump. Here the pump laser pinches down to  $R_L \approx 26$  µm. The secondary focusing, referred to as "gain focusing," has been discussed in Ref. [8] (see also Ref. [23]).

Refraction and defocusing by ionization, plasma formation, and thermal blooming are important processes that prevent



**Fig. 4.** Simulation of laser beam propagation with normalized power  $P_L/P_{Lc} = 5$ . (a) Normalized pump (blue) and Stokes Raman power (red), (b) spot radii of pump (blue) and Stokes Raman (red), (c) Brillouin backscatter power normalized to pump power, (d) spot radius of Brillouin backscatter, (e) electron density, and (f) pump (blue) and Stokes Raman (red) intensities, shown for slice at 100 ps in the pulse.

complete collapse of the laser beam for  $P_L/P_{Lc} \gg 1$ . To gain understanding of these effects, consider the envelope equation for the pump laser spot radius,

$$\frac{\partial^2 R_L}{\partial z^2} - \frac{4}{k_L^2 R_L^3} (1 + k_L R_L^2 G'_L) \approx 0$$
 (19a)

[cf. Eq. (16a)], where

$$k_{L}R_{L}^{2}G_{L}' = -\frac{P_{L}}{P_{Lc}} + 32\left(\frac{\omega_{L}}{c}\right)^{2} [\epsilon(\omega_{L}) - 1][\epsilon(\omega_{L}) + 2]$$

$$\times \frac{\beta}{\rho_{0}c_{p}} \frac{\Omega \operatorname{Im}\chi_{R}(\omega_{a})}{c^{2}} \frac{P_{S}P_{L}}{n_{L}n_{S}} \frac{R_{L}^{2} + R_{S}^{2}}{(R_{L}^{2} + 2R_{S}^{2})^{2}} \tau$$

$$+ \frac{\pi\ell}{(\ell-1)!(\ell+1)^{2}} \frac{\omega_{H_{2}O}^{2}R_{L}^{2}}{c^{2}} \left(\frac{I_{L}}{I_{MPI}}\right)^{\ell} \frac{\omega_{L}[e^{(\nu_{A0}-\eta)\tau} - 1]}{\nu_{i0} - \eta}$$
(19b)

is obtained from Eq. (C13) in Appendix C. Equations (19a) and (19b) are highly simplified versions of the equations that are used in the simulations; they are approximated here to illustrate some of the physical processes involved. The term  $-4/k_L^2 R_L^3$  in Eq. (19a) represents vacuum diffraction. The first term in the expression for  $k_L R_L^2 G'_L$  in Eq. (19b) is proportional to  $P_L$  and represents the self-focusing effect of the pump laser beam. The second term in Eq. (19b) drives thermal defocusing. It is proportional to the Raman susceptibility since it is the conversion of pump into Stokes Raman photons that heats the water. This term is also proportional to the time  $\tau$  elapsed since the defocusing effect accumulates with increasing water temperature. The last term in Eq. (19b) represents the refractive defocusing effect of the plasma formed by multiphoton and collisional ionization processes ( $\nu_{i0}$  is the ionization rate and  $\eta$  is the attachment rate-representing the loss of free electrons). Like thermal blooming, this effect is also proportional to the elapsed time  $\tau$  for early times (or for short laser pulses), i.e., for  $(\nu_{i0} - \eta)\tau \ll 1$ . For a long pulse, however,  $(\nu_{i0} - \eta)\tau \gg 1$ and the effect of plasma defocusing can become overwhelming, rising exponentially with time. In the closing paragraph of Section 2, it was noted that the present analysis is valid strictly in the parameter regime where any time dependences (due to temporal variation of the laser pulse, ionization, thermal blooming, etc.) are relatively small. This requirement is marginally satisfied for the case  $P_L/P_{Lc} = 5$ —and requires full-scale, 3D + time simulations for verification.

From Eq. (19a) a matched laser beam is obtained when  $1 + k_L R_L^2 G'_L \approx 0$ . Based on the complex spatio-temporal dependence of the function  $k_L R_L^2 G'_L$  noted in the previous paragraph, it is hard to envisage the laser beam maintaining a near-uniform profile for an extended length of time.

A key experimental observation is related to the appearance of an extended light "filament" for  $P_L/P_{Lc} \ge 1$  with a nearly uniform characteristic radius of  $\simeq 50 \ \mu m$  extending over the range 20 cm  $\leq z \leq 80$  cm along the tank (Fig. 2, Ref. [5]). Bearing in mind that the experimental data are integrated over the duration of the laser pulse, a possible interpretation of Fig. 2 therein is that the plasma plays an important role in light filament propagation. Early-on in the laser pulse, i.e., near the front of the pulse,  $P_L/P_{Lc} \ll 1$  and based on Fig. 3 appreciable focusing and plasma formation occur near the geometric focus ( $z \sim 60$  cm), while near the middle of the laser pulse  $(P_L/P_{Lc} \ge 1)$  focusing and plasma formation occur near z =20 cm based on Fig. 4. For comparison the minimum laser spot radius in Fig. 3(b) is  $R_L \approx 126 \,\mu\text{m}$ , while the minimum laser spot radius in Fig. 4(b) is  $R_L \approx 221 \ \mu\text{m}$ . Based on the argument advanced in the second paragraph of Section 4, the corresponding 1/e radii of the plasma are  $126/\sqrt{2\ell} \approx 51 \ \mu\text{m}$  and  $221/\sqrt{2\ell} \approx 90 \ \mu\text{m}$ , respectively, where  $\ell = 3$  is the number of photons required to ionize a water molecule.

The susceptibilities listed in Table 1 are obtained from various sources (cited in the caption) and are in the range reported in Refs. [5,7]; however, they are not known to high accuracy. It was verified that the Raman susceptibility employed gave a growth rate in the linear regime that was comparable to the experiments of Sceats *et al.* [1]. The proportion of pump power transferred to Stokes Raman power was taken as a measure of the accuracy of the susceptibility. Thus, in Fig. 4 of Ref. [5] the Stokes Raman is seen to peak at around ~60% of the pump power for  $P_L/P_{Lc} = 5$ , while the reduced analysis predicts that a little more than 60% of the pump power ends up in the Stokes Raman pulse [Fig. 4(a)].

It is pertinent to note that the details of simulation results are relatively sensitive to variations in the parameters listed in Table 1, some of which (e.g., attachment rate  $\eta$ ) are only known to within orders of magnitude. As an example Fig. 5



**Fig. 5.** Simulation of laser beam propagation with normalized power  $P_L/P_{Lc} = 5$ . The Raman susceptibility is  $-i4 \times 10^{-14}$  cm<sup>3</sup>/ erg, and other parameters are listed in Table 1. (a) Normalized pump (blue) and Stokes Raman power (red), (b) spot radii of pump (blue) and Stokes Raman (red), (c) Brillouin backscatter power normalized to pump power, (d) spot radius of Brillouin backscatter, (e) electron density, and (f) pump (blue) and Stokes Raman (red) intensities, shown for slice at 100 ps in the pulse.

shows the variation of the pump laser, the Stokes Raman, and the Brillouin excitations along the length of the tank for the case in which the Raman scattering susceptibility is taken to be  $-i4 \times 10^{-14}$  cm<sup>3</sup>/erg (rather than  $-i3.42 \times 10^{-14}$  cm<sup>3</sup>/erg listed in Table 1). Comparing Figs. 4 and 5 one observes the following differences: (i) the efficiency of Stokes Raman generation is higher [cf. Figs. 4(a) and 5(a), 68.38% versus 71.26%], (ii) secondary pump focusing occurs further downstream [cf. Figs. 4(b) and 5(b)], (iii) the efficiency of Brillouin backscatter generation is nearly an order of magnitude smaller [cf. Figs. 4(c) and 5(c)], and (iv) the peak electron density is lower [cf. Figs. 4(e) and 5(e),  $1.123 \times 10^{16}$  cm<sup>-3</sup> versus  $8.303 \times 10^{15}$  cm<sup>-3</sup>]. Observe that (i) and (iii) are consistent with the earlier assertion that increased efficiency of Stokes Raman generation occurs at the expense of pump power, which then reduces the growth of the Brillouin backscatter.

In closing this section the following remarks must be made. The reduced model can be an accurate representation in an experiment where propagation of the pump and of the Raman Stokes dominates, while the temporal variations introduced by ionization, thermal blooming, and Brillouin backscatter are relatively slow. Assuming the temporal form of the pump pulse to be a step function, the results of the reduced model for later slices in the pulse (e.g., at 1 ns) have also been examined. It is generally found that plots for the pump and Raman Stokes power [shown in Figs. 3(a), 4(a), and 5(a)] do not vary much between different slices. For some other characteristics, e.g., spot radius [shown in Figs. 3(b), 4(b), and 5(b)], there can be a qualitative difference. For example, for the slice at 1 ns

and  $P_L/P_{Lc} = 5$  the second focusing of the pump does not occur as a result of thermal blooming. In other words the time integrated (fluence) plots from the experiment can be expected to look similar to those from any given slice in the reduced model. This may explain why the time integrated spot radius in the experiments does not show the second focus obtained by analysis.

The propagation equations in the reduced model assume that the radiation beams maintain self-similar Gaussian transverse profiles throughout the propagation. This can be a poor approximation if the instabilities tend to hollow out the pump beam significantly. To validate this a (multimode) 2D + time pulse-frame simulation code has been employed to study the propagation of the pump and the Raman Stokes pulses. When one follows a constant energy surface of the pump beam, these simulations show refocusing behavior as a function of propagation distance; on the whole its variation is comparable to the single-mode, fundamental Gaussian approximation.

Finally it should be noted that the results in Figs. 1–5 are based on the assumption that the Brillouin backscatter is driven solely by the pump—as reflected by the terms on the righthand side of Eqs. (5) and (6). Examination of Fig. 4(a) may give the impression that there is sufficient energy in the Stokes Raman, spread over a fairly large fraction of the water tank, to drive Brillouin backscatter to a significant level. Similar calculations, where Brillouin backscatter is driven by the Stokes Raman wave, have been made. These show that the efficiency of conversion into Brillouin backscatter is much smaller than that shown in Fig. 2(b)—where the pump wave drives Brillouin backscatter. The physical reason for this is that, while there can be significant power in the Stokes Raman wave, its spot radius is quite large and therefore it is not sufficiently intense to excite much Brillouin backscatter.

#### 6. CONCLUSIONS

The physical processes associated with propagation of a highpower laser beam in a dielectric include self-focusing, stimulated Raman and stimulated Brillouin scattering, thermal blooming, multiphoton and collisional ionization, and plasma formation. In this paper analysis of these is presented based on the assumption that the radiation beams maintain self-similar Gaussian transverse profiles throughout the propagation. The analysis is used to perform self-consistent simulations of experimental results in the literature on propagation in water. Short temporal slices of the radiation pulses are followed as they progress along a water tank, assuming that the temporal evolution imposed by ionization and thermal blooming is relatively slow. Self-focusing of the pump is shown to have a significant impact on the stimulated processes. For relatively early times (~100's ps) simulations of the water tank experiments in Ref. [5] show that up to  $\approx 70\%$  of the laser energy can be channeled into stimulated Stokes Raman scattering and up to ≈10% can be channeled into stimulated Brillouin backscattering. In conjunction with the scattering processes, a relatively short and thin plasma filament forms along the propagation path that, in addition to thermal blooming, prevents catastrophic collapse of the laser beam.

#### **APPENDIX A**

Analysis of Brillouin backscattering makes use of the fluid equations for density  $\rho$ , velocity **u**, and entropy *s* to obtain a pair of equations for perturbed density  $\rho'$  and temperature T'[9-11],

$$\begin{split} \ddot{\rho}' &= \frac{c_s^2}{\gamma} \nabla^2 (\rho' + \beta \rho_0 T') + \left(\frac{\zeta}{\rho_0} + \frac{4}{3}\nu\right) \nabla^2 \dot{\rho}' \\ &- \frac{\alpha_1}{2c} \nabla^2 I_B \rho_0 c_p \dot{T}' - \frac{(\gamma - 1)c_v}{\beta} \dot{\rho}' - (c_p - c_v) \rho_0 \dot{T}' \\ &= \nabla \cdot (\kappa \nabla T') + \alpha_2 I_B, \end{split}$$

where an over dot denotes  $\partial/\partial t$ ,  $\rho_0$  is the ambient mass density,  $c_p = T(\partial s/\partial T)_p$  and  $c_v = T(\partial s/\partial T)_v$  are the specific heats,  $\gamma = c_p/c_v$ ,  $c_s = \sqrt{(\partial p/\partial \rho)_s}$  is the sound speed, p is the pressure,  $\beta = -(\partial \rho/\partial T)_p/\rho$  is the thermal-expansion coefficient,  $\zeta$ is the second (bulk) viscosity,  $\nu$  is the kinematic viscosity,  $\alpha_1 = \rho(\partial e/\partial \rho)_T/\text{Re}[\sqrt{(e/\mu)^*}]$  is proportional to the electrostriction coefficient,  $\epsilon$  is the permittivity,  $\mu$  is the permeability,  $\kappa$  is the thermal conductivity, and  $\alpha_2 = (\omega/c)\text{Im } e/\text{Re}[\sqrt{(e/\mu)^*}]$ is the absorption coefficient. As is well known,  $c_p \approx c_v$ ,  $\gamma \approx 1$  for water. In addition,  $I_B = (c/4\pi)\text{Re}[\sqrt{(e/\mu)^*}]\langle E^2 \rangle_B$ , where the suffix *B* attached to  $\langle E^2 \rangle_B$  identifies the contribution that drives Brillouin backscatter. These equations include the effects of both Brillouin and Rayleigh scattering.

## **APPENDIX B**

Analysis of thermal blooming in a medium makes use of the linearized equations for density  $\rho$ , velocity **u**, and entropy *s* to obtain an equation for perturbed temperature T' [9,10,13]. Using the same notation as in Appendix A and noting the absence of convection, one obtains

$$\rho_0 c_p T' = \nabla \cdot (\kappa \nabla T') + Q,$$

where Q is the quantity of heat generated by external sources (e.g., electromagnetic waves) in unit volume of the medium per unit time. The irreversible heat-generation terms are obtained by making use of Eqs. (4)–(6), resulting in

$$\rho_0 c_p \dot{T}' = \frac{3}{4} \{ \omega_L [\chi_R''(\omega_S)]^* + \omega_S \chi_R''(\omega_S) \} |A_L|^2 |A_S|^2 + \nabla$$
$$\cdot (\kappa \nabla T') + \frac{\omega_L}{8\pi} \epsilon''(\omega_L) |A_L|^2 + \frac{\omega_S}{8\pi} \epsilon''(\omega_S) |A_S|^2.$$
(B1)

The first term on the right-hand side is due to pump-Raman excitation of molecular vibrations, the next term describes thermal conduction, and the last two terms arise from linear absorption of electromagnetic radiation. Brillouin backscatter is mediated by sound waves, which are eventually damped and contribute to heating; however, this contribution is much smaller than that due to molecular vibrations.

For times  $\tau \ll R^2/8\chi$ , where *R* is a typical spot radius, thermal conduction is negligible [13], and then Eq. (B1) is simply integrated in time to obtain *T'*. For the parameters of interest here, this is a good approximation.

## **APPENDIX C**

An outline of the derivation of the explicit forms for the functions F'', G', and G'' for Eqs. (4)–(6) is given in this appendix.

In the SDE formulation a reduced wave equation of the general form

$$\left(\nabla_{\perp}^{2} + 2ik\frac{\partial}{\partial z}\right)A(r, z, \tau) = \mathbf{M}(r, z, \tau)A(r, z, \tau) \qquad (C1)$$

is solved, where  $M(r, z, \tau)$  is a nonlinear complex-valued function of  $A(r, z, \tau)$ . The electric field is expressed in the form

$$A(r, z, \tau) = B(z, \tau) \exp\left\{i\theta(z, \tau) - \frac{[1 + i\alpha(z, \tau)]r^2}{R^2(z, \tau)}\right\},$$
 (C2)

where *B* is the amplitude,  $\theta$  is the phase, *R* is the spot radius, and  $\alpha$  is related to the curvature of the wavefronts. The quantities *B*,  $\theta$ , *R*,  $\alpha$  are real-valued functions of *z*,  $\tau$ . Using the SDE method, the following set of coupled equations can be derived:

$$\frac{1}{BR}\frac{\partial(BR)}{\partial z} = F'',$$
 (C3)

$$\frac{\partial\theta}{\partial z} + \frac{(1+\alpha^2)}{kR^2} + \frac{\alpha}{R}\frac{\partial R}{\partial z} - \frac{1}{2}\frac{\partial\alpha}{\partial z} = -F',$$
 (C4)

$$\frac{1}{R}\frac{\partial R}{\partial z} + \frac{2\alpha}{kR^2} = -G^{\prime\prime},$$
(C5)

$$\frac{1}{2}\frac{\partial\alpha}{\partial z} + \frac{(1+\alpha^2)}{kR^2} = -G' - \alpha G'',$$
 (C6)

where  $F \equiv F' + iF''$  and  $G \equiv G' + iG''$  are given by

$$F(z,\tau) = \frac{1}{2k} \int_0^\infty d(\xi) M(r,z,\tau) e^{-\xi},$$
 (C7)

$$G(z,\tau) = \frac{1}{2k} \int_0^\infty d(\xi) M(r,z,\tau) (1-\xi) e^{-\xi},$$
 (C8)

where  $\xi \equiv 2r^2/R^2$ .

Calculation leads to the following explicit forms:

$$F_{S}^{\prime\prime} = -\frac{8(\omega_{S}/\omega_{L})^{2}}{k_{S}(R_{L}^{2} + R_{S}^{2})} \frac{\mathrm{Im}\chi_{R}(\omega_{S})}{\chi_{NR}} \frac{P_{L}}{P_{Lc}} - \frac{(\omega_{S}/c)^{2}\epsilon^{\prime\prime}(\omega_{S})}{2k_{S}} - \frac{\pi^{2}N_{\mathrm{H}_{2}\mathrm{O}}}{2\ell(\ell-1)!} R_{S}^{2} \frac{U_{\mathrm{ion}}\omega_{S}}{P_{S}} \left(\frac{I_{S}(z,\tau)}{I_{\mathrm{MPI}}}\right)^{\ell} - \frac{\omega_{\mathrm{H}_{2}\mathrm{O}}^{2}/c^{2}}{k_{S}} \frac{\pi\nu_{\epsilon0}}{(\ell-1)!} \int_{0}^{\tau} \mathrm{d}\tau^{\prime} \exp\left\{\int_{\tau^{\prime}}^{\tau} \mathrm{d}\tau^{\prime\prime}[\nu_{i0}(z,\tau^{\prime\prime}) - \eta]\right\} \times \left\{\frac{1}{\ell+1} \left(\frac{I_{S}(z,\tau^{\prime})}{I_{\mathrm{MPI}}}\right)^{\ell} + \frac{\omega_{L}/\omega_{S}}{\ell R_{S}^{2}(z,\tau^{\prime})/R_{L}^{2}(z,\tau^{\prime}) + 1} \left(\frac{I_{L}(z,\tau^{\prime})}{I_{\mathrm{MPI}}}\right)^{\ell} + \frac{\omega_{B}/\omega_{S}}{\ell R_{S}^{2}(z,\tau^{\prime})/R_{B}^{2}(z,\tau^{\prime}) + 1} \left(\frac{I_{B}(z,\tau^{\prime})}{I_{\mathrm{MPI}}}\right)^{\ell}\right\},$$
(C9)

$$F_{L}^{\prime\prime} = -\frac{8(n_{L}/n_{S})}{k_{L}(R_{L}^{2} + R_{S}^{2})} \frac{\mathrm{Im}\chi_{R}(\omega_{a})}{\chi_{NR}} \frac{P_{S}}{P_{Lc}} + \frac{(\omega_{L}/c)^{2}}{2k_{L}}$$

$$\times \left[\frac{16\,\mathrm{Im}\kappa_{\mathrm{SBS}}}{cn_{B}(R_{L}^{2} + R_{B}^{2})}P_{B} - \epsilon^{\prime\prime}(\omega_{L})\right] - \frac{\pi^{2}N_{\mathrm{H}_{2}\mathrm{O}}}{2\ell(\ell - 1)!}R_{L}^{2}\frac{U_{\mathrm{ion}}\omega_{L}}{P_{L}}$$

$$\times \left(\frac{I_{L}(z,\tau)}{I_{\mathrm{MPI}}}\right)^{\ell} - \frac{\omega_{\mathrm{H}_{2}\mathrm{O}}^{2}/c^{2}}{k_{L}}\frac{\pi\nu_{e0}}{(\ell - 1)!}\int_{0}^{\tau}\mathrm{d}\tau'$$

$$\times \exp\left\{\int_{\tau'}^{\tau}\mathrm{d}\tau''[\nu_{i0}(z,\tau'') - \eta]\right\}\left\{\frac{1}{\ell + 1}\left(\frac{I_{L}(z,\tau')}{I_{\mathrm{MPI}}}\right)^{\ell}$$

$$+ \frac{\omega_{S}/\omega_{L}}{\ell R_{L}^{2}(z,\tau')/R_{S}^{2}(z,\tau') + 1}\left(\frac{I_{S}(z,\tau')}{I_{\mathrm{MPI}}}\right)^{\ell}\right\}, \quad (C10)$$

$$F_{B}^{\prime\prime} = -\frac{(\omega_{B}/c)^{2}}{2k_{B}} \left[ \frac{16 \text{Im}\kappa_{\text{SBS}}^{*}}{cn_{L}(R_{L}^{2} + R_{B}^{2})} P_{L} - \epsilon^{\prime\prime}(\omega_{B}) \right] \\ - \frac{\pi^{2}N_{\text{H}_{2}\text{O}}}{2\ell(\ell-1)!} R_{B}^{2} \frac{U_{\text{ion}}\omega_{B}}{P_{B}} \left( \frac{I_{B}(z,\tau)}{I_{\text{MPI}}} \right)^{\ell} - \frac{\omega_{\text{H}_{2}\text{O}}^{2}/c^{2}}{k_{B}} \frac{\pi\nu_{\epsilon0}}{(\ell-1)!} \\ \times \int_{0}^{\tau} d\tau' \exp\left\{ \int_{\tau'}^{\tau} d\tau'' [\nu_{i0}(z,\tau'') - \eta] \right\} \left\{ \frac{1}{\ell+1} \left( \frac{I_{B}(z,\tau')}{I_{\text{MPI}}} \right)^{\ell} \\ + \frac{\omega_{L}/\omega_{B}}{\ell' R_{B}^{2}(z,\tau')/R_{L}^{2}(z,\tau') + 1} \left( \frac{I_{L}(z,\tau')}{I_{\text{MPI}}} \right)^{\ell} \right\} \\ + \frac{\omega_{S}/\omega_{B}}{\ell' R_{B}^{2}(z,\tau')/R_{S}^{2}(z,\tau') + 1} \left( \frac{I_{S}(z,\tau')}{I_{\text{MPI}}} \right)^{\ell} \right\}.$$
(C11)

In these equations  $P_j = cn_j R_j^2 B_j^2/16$  is the power,  $I_j = cn_j B_j^2/8\pi$  is the on-axis intensity,  $n_j = ck_j/\omega_j$  is the refractive index,  $P_{jc} = n_j/6\pi(\omega_j^2/c^3)\chi_{NR}$  is the critical power for self-focusing  $(j = L, S, B), \omega_{H_2O}^2 = 4\pi N_{H_2O}|e|^2/m, \nu_{e0} = \nu_e(r = 0, z, \tau)$ , and  $\nu_{i0} = \nu_i(r = 0, z, \tau)$ .

Terms proportional to  $\text{Im} \chi_R$  and  $\text{Im} \kappa_{\text{SBS}}$  in Eqs. (C9)– (C11) represent energy transfer from the pump laser due to Raman scattering and Brillouin scattering, and terms proportional to Im  $\epsilon$  represent power loss due to dielectric dissipation. The last two terms in Eqs. (C9)–(C11) represent energy loss due to multiphoton ionization and Joule heating, respectively:

$$k_{s}R_{S}^{2}G_{S}^{\prime} = -\frac{P_{S}}{P_{Sc}} - \frac{8(\omega_{S}/\omega_{L})^{2}}{(R_{L}^{2}/R_{S}^{2}+1)^{2}} [1 + \operatorname{Re}\chi_{R}(\omega_{S})/\chi_{NR}] \frac{P_{L}}{P_{Lc}} -32\left(\frac{\omega_{S}}{c}\right)^{2} [\epsilon(\omega_{S})-1][\epsilon(\omega_{S})+2] \times \frac{\beta\tau}{\rho_{0}c_{p}} \frac{\Omega\operatorname{Im}\chi_{R}(\omega_{S})P_{S}P_{L}}{c^{2}} \frac{R_{L}^{2}+R_{S}^{2}}{n_{L}n_{S}} \frac{R_{L}^{2}+R_{S}^{2}}{(2R_{L}^{2}+R_{S}^{2})^{2}} + \frac{\pi}{(\ell-1)!} \frac{\omega_{H}^{2}}{c^{2}} R_{S}^{2} \int_{0}^{\tau} d\tau' \exp\left\{\int_{\tau'}^{\tau} d\tau''[\nu_{i0}(z,\tau'')-\eta]\right\} \times \left\{\frac{\omega_{S}\ell}{(\ell+1)^{2}} \left(\frac{I_{S}(z,\tau')}{I_{MPI}}\right)^{\ell} + \frac{\omega_{L}\ell R_{S}^{2}(z,\tau')/R_{L}^{2}(z,\tau')}{[\ell R_{S}^{2}(z,\tau')/R_{L}^{2}(z,\tau')+1]^{2}} \left(\frac{I_{L}(z,\tau')}{I_{MPI}}\right)^{\ell} \right\}, \quad (C12)$$

$$k_{L}R_{L}^{2}G_{L}' = -\frac{P_{L}}{P_{Lc}} - \frac{8(n_{L}/n_{S})}{(R_{S}^{2}/R_{L}^{2}+1)^{2}} [1 + \operatorname{Re}\chi_{R}(\omega_{a})/\chi_{NR}] \frac{P_{S}}{P_{Lc}} + 32\left(\frac{\omega_{L}}{c}\right)^{2} [\epsilon(\omega_{L})-1] [\epsilon(\omega_{L})+2] \times \frac{\beta\tau}{\rho_{0}c_{p}} \frac{\Omega \operatorname{Im}\chi_{R}(\omega_{a})P_{S}P_{L}}{c^{2}} \frac{R_{L}^{2}+R_{S}^{2}}{n_{L}n_{S}} \frac{R_{L}^{2}+R_{S}^{2}}{(R_{L}^{2}+2R_{S}^{2})^{2}} + \frac{\pi}{(\ell-1)!} \frac{\omega_{H_{2}O}^{2}}{c^{2}} R_{L}^{2} \int_{0}^{\tau} d\tau' \exp\left\{\int_{\tau'}^{\tau} d\tau'' [\nu_{i0}(z,\tau'')-\eta]\right\} \\\times \left\{\frac{\omega_{L}\ell'}{(\ell+1)^{2}} \left(\frac{I_{L}(z,\tau')}{I_{MPI}}\right)^{\ell} \\+ \frac{\omega_{S}\ell R_{L}^{2}(z,\tau')/R_{S}^{2}(z,\tau')}{[\ell R_{L}^{2}(z,\tau')/R_{S}^{2}(z,\tau')+1]^{2}} \left(\frac{I_{S}(z,\tau')}{I_{MPI}}\right)^{\ell}\right\}, \quad (C13)$$

$$k_{B}R_{B}^{2}G_{B}^{\prime} = -\frac{P_{B}}{P_{Bc}} - \frac{8(\omega_{B}/\omega_{L})^{2}}{(R_{L}^{2}/R_{B}^{2}+1)^{2}} \frac{P_{L}}{P_{Lc}} + \frac{\pi}{(\ell-1)!} \frac{\omega_{H_{2}O}^{2}}{c^{2}} R_{B}^{2}$$

$$\times \int_{0}^{\tau} d\tau^{\prime} \exp\left\{\int_{\tau^{\prime}}^{\tau} d\tau^{\prime\prime} [\nu_{i0}(z,\tau^{\prime\prime}) - \eta]\right\}$$

$$\times \left\{\frac{\omega_{B}\ell^{\prime}}{(\ell+1)^{2}} \left(\frac{I_{B}(z,\tau^{\prime})}{I_{MPI}}\right)^{\ell}$$

$$+ \frac{\omega_{L}\ell R_{B}^{2}(z,\tau^{\prime})/R_{L}^{2}(z,\tau^{\prime})}{[\ell R_{B}^{2}(z,\tau^{\prime})/R_{L}^{2}(z,\tau^{\prime}) + 1]^{2}} \left(\frac{I_{L}(z,\tau^{\prime})}{I_{MPI}}\right)^{\ell}$$

$$+ \frac{\omega_{S}\ell R_{B}^{2}(z,\tau^{\prime})/R_{S}^{2}(z,\tau^{\prime}) + 1]^{2}}{[\ell R_{B}^{2}(z,\tau^{\prime})/R_{S}^{2}(z,\tau^{\prime}) + 1]^{2}} \left(\frac{I_{S}(z,\tau^{\prime})}{I_{MPI}}\right)^{\ell}\right\}.$$
 (C14)

Equations (C12)–(C14) describe modification of the reactive (real) part of the refractive index of the Stokes Raman, the pump, and the Brillouin backscatter waves, respectively. The first terms in Eqs. (C12)–(C14) represent self-focusing, the second term represents cross-beam focusing [9], and the third term represents plasma defocusing. Terms proportional to  $(\epsilon - 1)(\epsilon + 2) \text{Im} \chi_R$  represent the defocusing effect of thermal blooming arising from pump-Raman excitation of molecular vibrations. Thermal blooming arising from linear absorption of electromagnetic waves [proportional to  $\epsilon''(\omega_{L,S})$ ] is comparatively small.

$$k_{s}R_{s}^{2}G_{s}^{\prime\prime} = -\frac{8(\omega_{s}/\omega_{L})^{2}}{(1+R_{L}^{2}/R_{s}^{2})^{2}}\frac{\mathrm{Im}\chi_{R}(\omega_{s})}{\chi_{NR}}P_{Lc}$$

$$-\frac{\pi^{2}N_{\mathrm{H}_{2}\mathrm{O}}}{2\ell^{2}(\ell-2)!}R_{s}^{4}\frac{U_{\mathrm{ion}}k_{s}\omega_{s}}{P_{s}}\left(\frac{I_{s}(z,\tau)}{I_{\mathrm{MPI}}}\right)^{\ell}$$

$$-\frac{\pi}{(\ell-1)!}\frac{\omega_{\mathrm{H}_{2}\mathrm{O}}^{2}}{c^{2}}R_{s}^{2}\frac{\nu_{e0}}{\omega_{s}}\int_{0}^{\tau}\mathrm{d}\tau'$$

$$\times\exp\left\{\int_{\tau'}^{\tau}\mathrm{d}\tau''[\nu_{i0}(z,\tau'')-\eta]\right\}\left\{\frac{\omega_{s}\ell'}{(\ell+1)^{2}}\left(\frac{I_{s}(z,\tau')}{I_{\mathrm{MPI}}}\right)^{\ell}$$

$$+\frac{\omega_{L}\ell R_{s}^{2}(z,\tau')/R_{L}^{2}(z,\tau')}{[\ell R_{s}^{2}(z,\tau')/R_{L}^{2}(z,\tau')+1]^{2}}\left(\frac{I_{L}(z,\tau')}{I_{\mathrm{MPI}}}\right)^{\ell}$$

$$+\frac{\omega_{B}\ell R_{s}^{2}(z,\tau')/R_{B}^{2}(z,\tau')+1]^{2}}{[\ell R_{s}^{2}(z,\tau')/R_{B}^{2}(z,\tau')+1]^{2}}\left(\frac{I_{B}(z,\tau')}{I_{\mathrm{MPI}}}\right)^{\ell}\right\}, \quad (C15)$$

$$k_{L}R_{L}^{2}G_{L}^{\prime\prime} = -\frac{8(n_{L}/n_{S})}{(1+R_{S}^{2}/R_{L}^{2})^{2}} \frac{\mathrm{Im}\chi_{R}(\omega_{a})}{\chi_{NR}} \frac{P_{S}}{P_{Lc}} \\ + \left(\frac{\omega_{L}}{c}\right)^{2} \frac{8\mathrm{Im}\kappa_{\mathrm{SBS}}}{cn_{B}(1+R_{B}^{2}/R_{L}^{2})^{2}} P_{B} \\ - \frac{\pi^{2}N_{\mathrm{H_{2}O}}}{2\ell^{2}(\ell-2)!} R_{L}^{4} \frac{U_{\mathrm{ion}}k_{L}\omega_{L}}{P_{L}} \left(\frac{I_{L}(z,\tau)}{I_{\mathrm{MPI}}}\right)^{\ell} \\ - \frac{\pi}{(\ell-1)!} \frac{\omega_{\mathrm{H_{2}O}}^{2}}{c^{2}} R_{L}^{2} \frac{\nu_{e0}}{\omega_{L}} \int_{0}^{\tau} \mathrm{d}\tau' \\ \times \exp\left\{\int_{\tau'}^{\tau} \mathrm{d}\tau''[\nu_{i0}(z,\tau'')-\eta]\right\} \\ \times \left\{\frac{\omega_{L}\ell}{(\ell+1)^{2}} \left(\frac{I_{L}(z,\tau')}{I_{\mathrm{MPI}}}\right)^{\ell} \\ + \frac{\omega_{S}\ell R_{L}^{2}(z,\tau')/R_{S}^{2}(z,\tau')}{[\ell R_{L}^{2}(z,\tau')/R_{S}^{2}(z,\tau')+1]^{2}} \left(\frac{I_{S}(z,\tau')}{I_{\mathrm{MPI}}}\right)^{\ell} \right\},$$
(C16)

$$\begin{aligned} k_{B}R_{B}^{2}G_{B}^{"} &= -\left(\frac{\omega_{B}}{c}\right)^{2} \frac{8\,\mathrm{Im}\,\kappa_{\mathrm{SBS}}^{*}}{cn_{L}(1+R_{L}^{2}/R_{B}^{2})^{2}}P_{L} \\ &- \frac{\pi^{2}N_{\mathrm{H_{2}O}}}{2\ell^{2}(\ell-2)!}R_{L}^{4}\frac{U_{\mathrm{ion}}k_{L}\omega_{L}}{P_{L}}\left(\frac{I_{L}(z,\tau)}{I_{\mathrm{MPI}}}\right)^{\ell} \\ &- \frac{\pi}{(\ell-1)!}\frac{\omega_{\mathrm{H_{2}O}}^{2}}{c^{2}}R_{L}^{2}\frac{\nu_{e0}}{\omega_{L}}\int_{0}^{\tau}\mathrm{d}\tau' \\ &\times \exp\left\{\int_{\tau'}^{\tau}\mathrm{d}\tau''[\nu_{i0}(z,\tau'')-\eta]\right\} \\ &\times \left\{\frac{\omega_{B}\ell}{(\ell+1)^{2}}\left(\frac{I_{B}(z,\tau')}{I_{\mathrm{MPI}}}\right)^{\ell} \\ &+ \frac{\omega_{S}\ell R_{L}^{2}(z,\tau')/R_{S}^{2}(z,\tau')}{[\ell R_{L}^{2}(z,\tau')/R_{B}^{2}(z,\tau')+1]^{2}}\left(\frac{I_{S}(z,\tau')}{I_{\mathrm{MPI}}}\right)^{\ell} \right\}. \text{ (C17)} \end{aligned}$$

Equations (C15)–(C17) represent the contribution of energy exchange between the waves, ionization losses, and Joule heating to the variation of the spot radius of the Stokes Raman, the pump, and the Brillouin backscattered waves, respectively.

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