Self-induced gain and loss modulation in coherent, transient Raman pulse propagation*

G. I. Kachen† and W. H. Lowdermilk
Lawrence Livermore Laboratory, University of California, Livermore, California 94550
(Received 19 April 1976)

Self-induced modulation of Raman-Stokes pulse amplification and laser pulse attenuation was observed in coherent Raman pulse propagation in H_2 gas. This modulation is due to laser pulse depletion and transient response of the material polarization. Theoretical analysis of the propagation equations agrees well with experimental observations.

Coherent, transient response of material polarization plays an important role in the propagation of high-intensity, short-duration laser pulses through Raman active media. In particular, the transient material response can result in a reversal of the normal direction of energy transfer from the incident laser pulse to the Raman-Stokes pulse. This effect is observed as a temporal modulation of the laser and Stokes pulse intensities transmitted through the medium. We have made direct observation of this self-induced gain and loss modulation in hydrogen gas using a high-speed streak camera. In addition, we have performed a theoretical and numerical analysis of coherent, transient Raman pulse propagation in the limit of negligible material excitation. The calculations are found to be in good agreement with experimental results.

Coherence effects are observed for times shorter than the dephasing time T_2 of a coherent molecular polarization. To make this time long, hydrogen gas, with its unusually narrow vibrational linewidth T_2^{-1}, was used at low pressure. With the available laser power, consistent stimulated scattering was observed at 30 atm. At this pressure, $T_2 \simeq 0.24$ nsec.[1] The laser pulse was generated by a Nd:YAlG/glass laser, which has been previously described.[2] The laser produced a 200-mJ single pulse of 0.8 nsec duration. This pulse was converted to the second-harmonic wavelength in KDP. The spatial and temporal intensity distribution of the second-harmonic pulse was approximately Gaussian down to the e^{-4} intensity with random fluctuations less than 4%. Furthermore, the laser pulse bandwidth of 0.02 cm^{-1} was the transform limit of a 0.8-nsec Gaussian pulse, which indicates that the pulse did not contain faster time structure. The Raman-Stokes pulse was generated in a 100-cm-long gas cell by the collimated laser beam with a peak intensity of 10^8 W/cm^2. The gas cell was long compared to the spatial extent of the laser pulse so that complications due to feedback from the cell windows were avoided. In addition, the laser pulse intensity could be adjusted so that neither stimulated Raman scattering in the backward direction nor higher-order Stokes pulses were observable. After generation in the gas cell, the laser and Stokes pulses were spatially separated by a prism. Both these pulses and a portion of the incident laser pulse which did not pass through the gas cell were simultaneously recorded by a streak camera with 10 psec resolution. To reduce spatial averaging of the pulse intensities, apertures were placed to allow only a small cross-sectional area of the beam to enter the streak camera. A typical streak camera photograph, which displays the time dependence of the various pulse intensities, is shown in Fig. 1. The laser pulse is severely depleted near its peak by conversion to the Stokes pulse, whose intensity increases sharply with time. A modulation then occurs, with the laser and Stokes pulse intensities alternately rising and falling.

Theoretical analyses of coherent Raman propagation have been given by several authors.[3] These analyses consist of a semiclassical calculation of the three-level system under simultaneous perturbation of the Stokes and laser fields $E_S(z,t)$ and $E_L(z,t)$. The fields are represented as monochromatic plane waves $E_i(z,t) = \mathcal{E}_i(z,t)\cos[\omega_i t - k_i z + \phi_i(z,t)]$, $i = L, S$. The important dynamic variables of the material system are the normalized population inversion w between the ground and excited vibrational or electronic level, and the out-of-phase component of the second-order induced polarizability v, which couples these levels through an off-resonance virtual level. The material equations of motion, derived by a perturbation expansion of the Schrödinger equation, are given in a form analogous to the Bloch equations,

$$\dot{w} = \alpha \mathcal{E}_L \mathcal{E}_S v - (w - w_{eq})/T_1 , \tag{1a}$$

$$\dot{v} = -\alpha \mathcal{E}_L \mathcal{E}_S w - v/T_2 , \tag{1b}$$

where α is a constant of the material. The set of equations is closed by addition of Maxwell's equa-

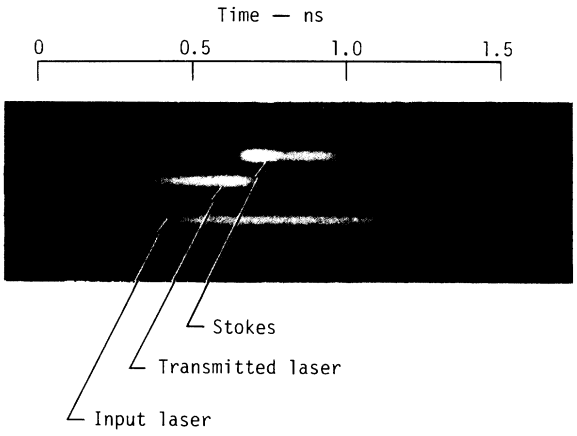

FIG. 1. Streak camera photograph showing time dependence of incident laser pulse and transmitted Stokes and laser pulses.

tion for the fields. Assuming slowly varying amplitudes and constant phases, the reduced field equations are

$$\frac{\partial \mathcal{E}_L}{\partial z} + \frac{n_L}{c}\frac{\partial \mathcal{E}_L}{\partial t} = -\kappa\omega_L \mathcal{E}_S v \ , \tag{2a}$$

$$\frac{\partial \mathcal{E}_S}{\partial z} + \frac{n_S}{c}\frac{\partial \mathcal{E}_S}{\partial t} = \kappa\omega_S \mathcal{E}_L v \ , \tag{2b}$$

where κ is a material constant related to the steady-state Raman gain coefficient g. The fields couple to the material system through the induced nonlinear macroscopic polarizations which act as sources for the fields. Thus Eqs. (1) and (2) characterize the Raman pulse propagation dynamics in a self-consistent form. Tan-no *et al.*[3] give solutions to these equations which exhibit gain and loss modulation due to material saturation. This source of modulation, which has not been observed, is particularly evident in the undamped limit $T_1 = T_2 = \infty$. Then solution of Eq. (1) is $v = \sin\phi(z,t)$, $w = -\cos\phi(z,t)$, with

$$\phi(z,t) = \alpha \int_{-\infty}^{t} \mathcal{E}_L(z,t')\mathcal{E}_S(z,t')\,dt' .$$

Thus for pulses having a large value of $\phi(0,\infty)$, the source polarization terms in Eq. (2) vary as $\sin\phi$, resulting in periodic gain and loss. Modulation of gain and loss in coherent propagation also occurs due to depletion of the laser field. This effect is the source of the observed modulation.

Under the experimental conditions, less than one molecule in 10^4 is excited; thus w is approximately constant. Assuming both pulses travel at velocity c/n, and transforming to the moving-coordinate frame ($\tau = t - zn/c$), the Stokes field amplitude is given by

$$\frac{\partial \mathcal{E}_S(z,\tau)}{\partial z} = gT_2^{-1}\mathcal{E}_L(z,\tau) \times \int_{-\infty}^{\tau} \exp[(t-\tau)T_2^{-1}]\,\mathcal{E}_L(z,t)\mathcal{E}_S(z,t)\,dt \ . \tag{3}$$

In this case, the steady-state gain is related to previously introduced material constants by the relation $g = -\kappa\alpha\omega_S w_{eq} T_2$. Equation (2) leads to the Manley-Rowe relation

$$\frac{\mathcal{E}_L^2(z,\tau)}{\omega_L} + \frac{\mathcal{E}_S^2(z,\tau)}{\omega_S} = \frac{\mathcal{E}_L^2(0,\tau)}{\omega_L} \ . \tag{4}$$

A study of the laser and Stokes pulse evolution was performed by numerical solution of Eqs. (3) and (4). An example of the results obtained is shown in Fig. 2. In this set of calculations, the incident laser pulse was taken to be Gaussian in time, and a Stokes noise field $\mathcal{E}_S(0,\tau) = \mathcal{E}_L(0,\tau)e^{-18}$ was introduced. The calculated laser and Stokes field amplitudes and intensities in Fig. 2 show the evolution of the pulses for increasing distance of propagation into the Raman-active medium. Initially, the laser pulse is attenuated, and the Stokes pulse grows with the characteristic pulse shortening and delay relative to the laser pulse.[2,4] This process continues until at some point in the pulse the laser field becomes totally depleted. Then, since the coherent molecular polarization which has been

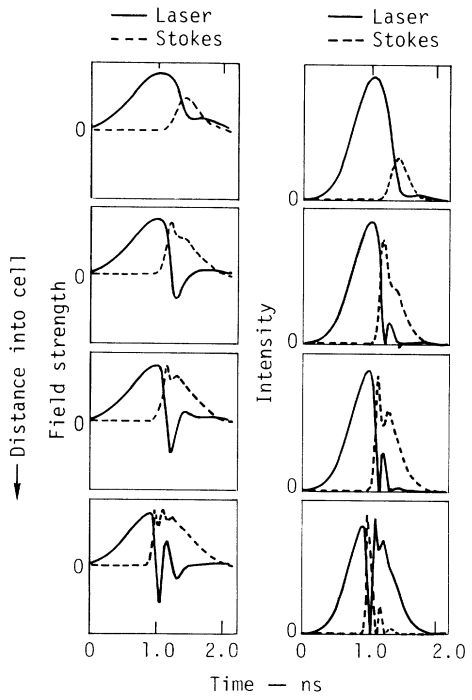

FIG. 2. Numerical solution of pulse propagation equations showing the evolution of laser and Stokes pulse field amplitudes and intensities for increasing distance of propagation.

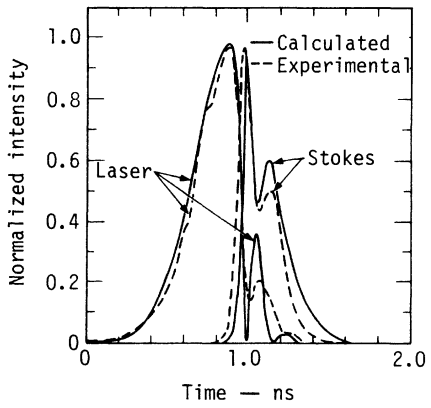

FIG. 3. Comparison of calculated and experimentally observed time dependence of transmitted laser and Stokes pulses.

created at that point in the medium does not dephase instantly, the nonlinear source polarization at the laser frequency remains finite. Thus the laser field is regenerated by this source polarization with a phase shift of π rad from the initial field, while the Stokes field traveling with this portion of the laser pulse experiences loss. The magnitude of the induced polarizability v, which had been increasing through the action of the laser and Stokes fields at earlier times, begins to decrease. This process continues until the polarizability vanishes. Then the original conditions exist and the process begins again, leading to modulation of the Stokes and laser pulses. This modulation does not occur in the steady-state limit ($T_2 \to 0$), because in this case the amplitude of the molecular polarization responds instantly to the driving fields and vanishes when the laser field is depleted.

To compare these predictions with the experimental results, calculations were performed using the incident laser field $\mathcal{E}_L(0,\tau)$ obtained from a microdensitometer trace of the streak camera photograph. The theoretical and experimental results are compared in Fig. 3. The theoretical result, chosen from a series of calculations at increasing propagation distances, had the best fit to the experimental results. The calculated pulse shapes and locations of the modulation peaks are in good qualitative agreement with the experimental results. Several factors contribute to the lack of exact agreement. The laser pulse modulation observed by the streak camera is not 100%, since the modulation depends on the field amplitudes, and some spatial averaging over the beam profile is unavoidable due to the finite aperture of the streak camera. In addition, because of scattering and diffraction losses, the Manley-Rowe condition [Eq. (4)] is not rigorously obeyed in the experiment.

In conclusion, the data presented show a new aspect of coherent, transient Raman pulse propagation. When the Raman gain is sufficiently large, interplay of laser pulse depletion and the transient material response result in a self-induced gain and loss modulation of the Stokes and laser pulses. Numerical solutions of the pulse propagation equations were obtained and are found to be in good agreement with the experimental results.

---

*Work performed under the auspices of the U. S. Energy Research and Development Administration under Contract No. W-7405-Eng-48.

†Present address: Dept. of Electronics, University of Southampton and Culham Laboratory, Abingdon, OXON, OX14-3DB, England.

[1]J. R. Murray and A. Javan, J. Mol. Spectrosc. 42, 1 (1972).

[2]W. H. Lowdermilk and G. I. Kachen, Appl. Phys. Lett. 27, 133 (1975).

[3]For references see E. Courtens, in *Laser Handbook*, edited by F. T. Arecchi and E. O. Schulz-Dubois (North-Holland, Amsterdam, 1972), p. 1261. See also N. Tanno, T. Shirahata, and K. Yokoto, Phys. Rev. A 12, 159 (1975).

[4]R. L. Carman, F. Shimuzu, C. S. Wang, and N. Bloembergen, Phys. Rev. A 2, 60 (1970).

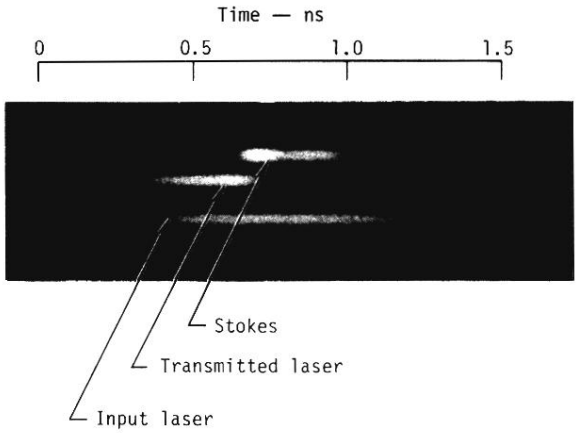

FIG. 1. Streak camera photograph showing time dependence of incident laser pulse and transmitted Stokes and laser pulses.