

Subpicosecond stimulated Raman scattering in high-pressure hydrogen

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We studied the effect of self-phase modulation and self-focusing on transient stimulated Raman scattering in high-pressure hydrogen by using high-energy, subpicosecond laser pulses. Adding argon to the hydrogen emphasizes the effect of self-phase modulation on stimulated Raman scattering by increasing the former effect without affecting the latter. The behavior of the observed stimulated Raman scattering falls into three regimes depending on input energy: normal stimulated Raman scattering at low energies, suppression by self-phase modulation at medium energies, and a recovery at high energies because strong self-focusing limits self-phase modulation.

INTRODUCTION

Stimulated Raman scattering in high-pressure hydrogen gas is a commonly used means of shifting the wavelength of a laser beam. However, this technique becomes difficult to apply when the pump beam consists of picosecond or subpicosecond laser pulses. In the steady-state stimulated-Raman-scattering regime, when the input pulse width is longer than the steady-state gain G_{SS} times the dephasing time of the Raman excitation level T_2 , shortening the input pulse width increases the total Stokes radiation output because the Raman gain depends on the peak power of the incident beam.¹ But in the transient stimulated-Raman-scattering regime, when the input laser-pulse width is much smaller than $G_{SS}T_2$, the stimulated Raman gain depends only on the integrated energy of the pump laser pulses.²⁻⁵ Therefore shortening the input pulse width will not increase the output Stokes radiation. Moreover, as the pulse width of the pump beam becomes shorter, other nonlinear processes, such as self-phase modulation, self-focusing, harmonic generation, and multiphoton ionization, begin to become important and affect the growth of the Stokes radiation. Investigation of the interaction between stimulated Raman scattering and other nonlinear effects is necessary for obtaining better control of this technique as a frequency shifter for coherent radiation, especially for ultrashort pump laser pulses. Previous studies of transient stimulated Raman scattering with femtosecond laser pulses showed the importance of self-phase modulation in Stokes radiation production.^{6,7} However, these studies focused on the effect of the self-phase modulation on the Raman spectral width, not the Raman gain. Here we investigate the effect of self-phase modulation on transient stimulated-Raman-scattering gain as well as the role played by self-focusing.

EXPERIMENTAL SETUP AND RESULTS

The experimental setup is shown in Fig. 1. A colliding-pulse mode-locked (CPM) dye laser and a 10-Hz high-energy dye amplifier system⁸ without a pulse compressor produce 840-fs, 1-mJ laser pulses at 620 nm that serve as the pump pulses for our experiments. A calibrated phototube detector monitors the input laser-beam energy before the Raman cell. The pump laser pulses are focused into a 1-m-long Raman cell by a 1-m focal-length convex lens, and another convex lens collimates the radiation coming out of the cell. The output at the pump wavelength is detected with a pyroelectric detector with a 10-nm bandpass filter centered at 620 nm. The measured values are corrected to account for reflection and absorption losses in the Raman cell windows and the bandpass filter. The Stokes radiation, at 835 nm for hydrogen gas, is measured by placement of the pyroelectric detector after two prisms and selection of only the Stokes-shifted region of the output spectrum. A spectral measurement of the light in this frequency region, seen in Fig. 2, shows that there is no background of spectrally broadened radiation from the pump beam in the Stokes spectral region. This spectrum confirms that when we select the Stokes spectral region we are measuring only the stimulated-Raman-scattering output.

To measure the strength of self-phase modulation, we define the self-phase-modulation energy as the energy in the radiation that, because of self-phase modulation, lies outside the original pump-pulse spectrum. Then the total output energy is equal to the sum of the output energy at 620 nm, the self-phase-modulation energy, and the Stokes-radiation energy, with small effects such as anti-Stokes radiation and high-order harmonic generation neglected. Since no energy is absorbed in the Raman cell, we calculate the self-phase-modulation energy by subtracting the sum of the 620-nm output energy and the Stokes-radiation energy from the input energy.

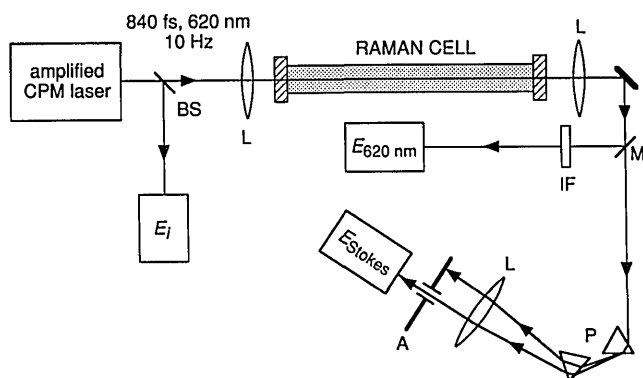


Fig. 1. Experimental setup. BS, beam splitter; L's, lenses; M, removable mirror; IF, interference filter; P, prisms; E_i , input energy; $E_{620\text{ nm}}$, output energy at 620 nm; E_{Stokes} , energy at Stokes-shifted frequency.

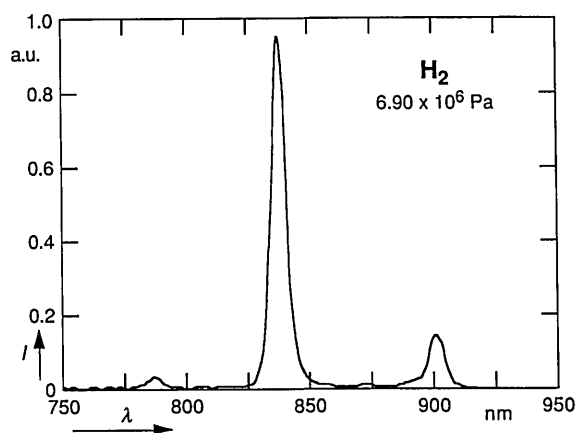


Fig. 2. Stokes part of the Raman spectrum for hydrogen at a pressure of 6.90×10^6 Pa. The input energy into the cell at 620 nm is $530 \mu\text{J}$. The central peak at 835 nm is the first-order vibrational Stokes line; the smaller side peaks are rotational Stokes and anti-Stokes lines produced by the radiation at 835 nm.

Figure 3 shows a plot of the Stokes-radiation energy versus the input pump energy in pure hydrogen gas from 3.45×10^6 to 8.27×10^6 Pa at room temperature, corresponding to a molecular-density range from 1390 to 3340 mol/m^3 . The corresponding plot of self-phase-modulation energy versus input energy appears in Fig. 4. Taking the derivative of the total self-phase-modulation energy with respect to the input pulse energy determines the self-phase-modulation production rate, shown as a function of input energy in Fig. 5. The arrows in Fig. 3 mark the input energies at which the self-phase-modulation production rates in Fig. 5 reach a maximum.

Argon gas has a nonlinear index of refraction 1.2 times that of hydrogen, but because argon is monatomic, it has no Raman-active vibrational modes.⁹ Thus adding argon to hydrogen increases the nonlinear index of refraction without contributing directly to Stokes-radiation production, enhancing the effects of self-phase modulation and self-focusing on the stimulated Raman scattering produced by the hydrogen. Figure 6 shows a plot of the Stokes-radiation energy versus the input pump energy in hydrogen-plus-argon mixtures. The various curves in Fig. 6 correspond to different partial pressures of argon gas added to a fixed partial pressure of hydrogen gas of

4.83×10^6 Pa. As in Fig. 3, the arrows indicate the positions of the maximum self-phase-modulation production rate.

DISCUSSION

We need to make several points to simplify the analysis of our results. First, hydrogen gas has a dispersionless refractive index in the visible wavelength range, permitting straightforward analysis without any complications that are due to walk-off between the pump laser pulse and the Stokes pulse.^{4,5} Second, we used uncompressed 840-fs input pulses, because when we tried using compressed 100-fs pulses we found that self-phase modulation strongly overpowers Stokes radiation at all input energies. Although the 840-fs input laser pulses are severely chirped owing to the group-velocity dispersion in the amplifier stages, the chirp does not strongly affect the total gain of the Stokes radiation. Carman *et al.*³ showed that in the limit of high transient gain the Raman gain coefficient is independent of the phase structure of the input pump pulse [see Eq. (A25)]. Since the

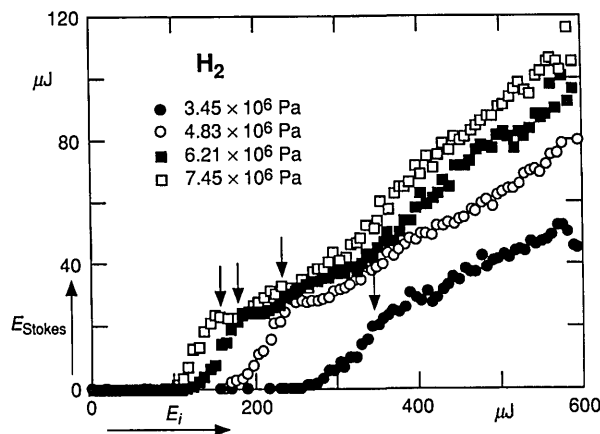


Fig. 3. Stokes output energy versus input energy for hydrogen at pressures of 3.45×10^6 , 4.83×10^6 , 6.21×10^6 , and 7.45×10^6 Pa. The arrows mark the input energies at which the self-phase-modulation production rate reaches a maximum (see Fig. 5).

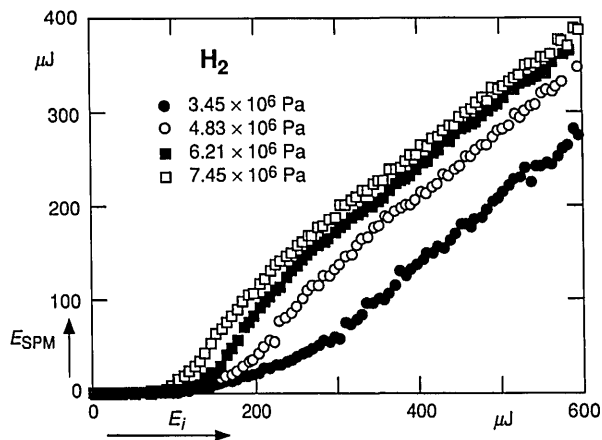


Fig. 4. Self-phase-modulation output energy versus input energy for hydrogen at the same pressures as in Fig. 3. The self-phase-modulation energy is calculated by subtracting the output energy at the input wavelength (620 nm) and at the Stokes-shifted wavelength (835 nm) from the total input energy, $E_{\text{SPM}} = E_i - E_{620} - E_{\text{Stokes}}$.

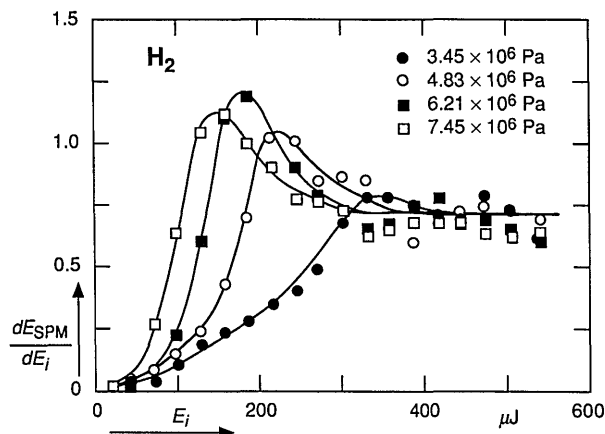


Fig. 5. Self-phase-modulation production rate versus input energy for hydrogen at the same pressures as in Figs. 3 and 4. The data points in this figure are the derivative with respect to the input energy of the self-phase-modulation energy in Fig. 4. The curves through the data sets are drawn to guide the eye.

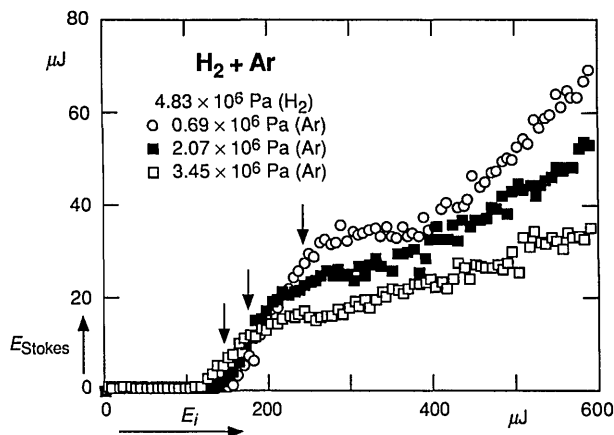


Fig. 6. Stokes output energy versus input energy for a hydrogen partial pressure of 4.83×10^6 Pa with argon partial pressures of 0.69×10^6 , 2.07×10^6 , and 3.45×10^6 Pa. The arrows again mark the input energies at which the self-phase-modulation production rate reaches a maximum.

dephasing time for hydrogen in the pressure range used is 40–100 ps,¹⁰ we are well within the transient stimulated-Raman-scattering regime with 840-fs input pulses. Furthermore, the spectral broadening that is due to self-phase modulation in the hydrogen gas is not highly sensitive to the chirp.¹¹ Finally, our power density is of the order of 10^{15} W/m², which is at least an order of magnitude too low for multiphoton ionization to play an important role in our experiment.¹² This absence of multiphoton ionization was confirmed experimentally by the linear relation between the total output energy and the input laser-pulse energy in our input energy range.

Low-Energy Regime

Figure 3 suggests three regimes for transient stimulated Raman scattering: low, medium, and high input energy. In the low-energy regime, which corresponds to the region left of the arrow for each curve, the dependence of the Stokes-radiation energy on the input energy agrees with the previously found $\exp(G_{\text{trans}})/G_{\text{trans}}$ ³ dependence, where the transient gain G_{trans} is proportional

to the square root of the input energy.² The self-phase-modulation energy in this region is not yet strong enough to have a significant effect on the transient stimulated Raman scattering. However, self-focusing manifests itself in two important ways. First, Fig. 6 shows that the threshold energy for stimulated Raman scattering decreases with increasing argon pressure, even though the argon does not directly contribute to Stokes production. This decrease in the threshold energy results from enhanced self-focusing caused by the increased pressure.¹³ Stronger self-focusing leads to a higher energy density in the focal region for a given input energy. Second, as is evidenced by Fig. 4, the self-phase-modulation energy grows with input energy faster than the linear dependence expected in the absence of self-focusing [see relation (B11)].

Medium-Energy Regime

The medium-energy regime extends from the peak in the self-phase-modulation production rate to the beginning of the plateau in this rate (see Fig. 5). In this regime strong self-phase modulation leads to suppression of the Stokes radiation. As can be seen by comparing Figs. 3 and 5, the onset of suppression coincides with the point at which the self-phase-modulation production rate peaks.

For pure hydrogen, Stokes-energy production in the low-energy regime becomes more efficient as the pressure is increased. At the same time, however, the onset of suppression occurs at a lower input energy because of stronger self-phase modulation (see Fig. 3). As a result of these two effects, the amount of Stokes energy generated at the onset of suppression is nearly independent of the hydrogen pressure. When argon is added, on the other hand, the enhancement of self-phase modulation is not countered by a corresponding increase in Stokes production. Thus the Stokes-energy level at the onset of suppression drops with increasing argon pressure (see Fig. 6).

The suppression of the Stokes radiation by self-phase modulation can be understood by analysis of the effect of self-phase modulation on the pump pulse. Self-phase modulation spreads the input pulse energy over various frequencies, reducing the energy at the original frequency. Since only the input pulse energy at the original frequency provides gain for the Stokes radiation that starts building up at the beginning of the Raman cell, we can treat the spectral broadening of the input pulse caused by self-phase modulation as a reduction in the pump energy along the interaction path. Assuming that the temporal profile of the pump pulse does not vary along the interaction region, we may separate the spatial and the temporal dependences of the field amplitude associated with the pump pulse and write $\mathcal{E}_p(z, t) = g_p(z)f_p(t')$, where f_p is the temporal profile and $t' = t - z/v_g$, with v_g the group velocity of the pulse, g_p the spatial envelope, and $g_p(0) = 1$. Following an approach³ similar to that of Carman *et al.*,² an approximate expression for the gain coefficient for transient stimulated Raman scattering can be derived [see Eq. (A25)]:

$$G_{\text{trans}}(z) \approx 4 \left[\eta_1 \eta_2 \int_0^z |g_p(z')|^2 dz' \int_{-\infty}^{\infty} |f_p(t'')|^2 dt'' \right]^{1/2}, \quad (1)$$

where the product $\eta_1 \eta_2$ is proportional to the square of the molecular density (see Appendix A). Without self-phase modulation the spatial dependence $|g_p(z)|$ is unity, and Eq. (1) reduces to the result derived by Carman *et al.* On the other hand, if the pump-pulse amplitude becomes smaller along the interaction path owing to self-phase modulation, then $g_p(z)$ decreases as z increases, and the total stimulated Raman gain decreases accordingly.

To quantify the effect of self-phase modulation on the total stimulated Raman gain, let us define the effective interaction length l_D as the length scale over which the average of the square of the spatial envelope function $|g_p(z)|^2$ equals 1/2. If the effective interaction length l_D is much larger than the actual interaction length L set by the confocal parameter of the focusing optics, $l_D \gg L$, then the decrease in the pump-pulse energy that is due to self-phase modulation is negligible over the interaction region. In this case, corresponding to the low-input-energy regime, the effect of self-phase modulation on the pump pulse can be ignored, and the transient gain coefficient in Eq. (1) is approximately proportional to \sqrt{L} . On the other hand, if $l_D \ll L$, then the integration over the interaction path in Eq. (1) becomes

$$\int_0^L |g_p(z)|^2 dz = \frac{l_D}{2} + \int_{l_D}^L |g_p(z)|^2 dz = \frac{l_D}{2} + O\left(\frac{l_D}{L}\right). \quad (2)$$

Thus, when $l_D \ll L$, to first order the transient gain coefficient is proportional to $\sqrt{l_D}$. Under this condition we can write

$$G_{\text{total}}(L) \propto (N_{\text{H}_2}^2 E_i l_D)^{1/2}, \quad (3)$$

where N_{H_2} is the molecular density of hydrogen and E_i is the incident pulse energy.¹⁴

To see the effect of self-phase modulation on the stimulated Raman gain, we must first examine the effect of self-phase modulation on the effective interaction length l_D in Eq. (3). In the absence of self-focusing the effect of self-phase modulation on the pump-pulse amplitude is (see Appendix B)

$$E_p(z, t') = E_p(0, t') [1 + (CN_{\text{H}_2} E_i z)^2]^{-1/4}, \quad (4)$$

where C is a constant. Using this expression to find l_D gives

$$\begin{aligned} \frac{l_D}{2} &= \int_0^{l_D} |g_p(z)|^2 dz \\ &= \int_0^{l_D} [1 + (CN_{\text{H}_2} E_i z)^2]^{-1/2} dz \\ &= \frac{1}{CN_{\text{H}_2} E_i} \ln\{CN_{\text{H}_2} E_i l_D + [1 + (CN_{\text{H}_2} E_i l_D)^2]^{1/2}\}. \end{aligned} \quad (5)$$

Solving self-consistently for l_D shows that

$$l_D \propto \frac{1}{N_{\text{H}_2} E_i}. \quad (6)$$

Substituting Eq. (6) into Eq. (3), we find that when $l_D \ll L$ the transient stimulated Raman gain becomes independent of input energy,

$$G_{\text{total}}(L) \propto \sqrt{N_{\text{H}_2}}. \quad (7)$$

This result explains the nearly flat regions observed in the curves in Fig. 3 immediately after the onset of suppression. The above result also suggests that the transition from the low-energy regime to the medium-energy regime, marked by the onset of suppression, corresponds roughly to the point at which l_D becomes smaller than L .

High-Energy Regime

As the input energy is further increased, the self-phase-modulation production rate drops from its maximum to a constant value that is independent of pressure and input energy. In parallel with this flattening of the self-phase-modulation production rate in Fig. 5, we note a partial recovery of the Stokes production in Fig. 3. Once the self-phase-modulation production rate is constant, the Stokes energy takes on an approximately linear dependence on input energy.

The linear dependence of both the self-phase-modulation energy and the Stokes radiation on input energy in this regime can be explained by the breakup of the laser beam into fine filaments caused by strong self-focusing, an effect that is confirmed by visible observation of the output spatial profile of the pump beam at high input energy. Campillo *et al.*¹⁵ carried out a linear stability analysis and showed that when the input pulse energy density is larger than a critical value the laser beam will break up into filaments with the energy density in each filament inversely proportional to the nonlinear refractive index, and therefore to the molecular density, but independent of the total input energy density.¹⁶ In other words, increasing the input energy will increase the number of filaments but will not increase the energy density in each filament.

Since the input energy density per filament is fixed after beam breakup has occurred, we can use relation (B11) to calculate the self-phase-modulation energy density produced in each filament:

$$F_{\text{SPM}} = \alpha F_i, \quad (8)$$

with

$$\alpha \approx 1 - \frac{1}{[1 + 2(\eta_3 |E_0|^2 L)^2]^{1/2}} \quad (9)$$

being independent of total input energy because the input energy density per filament is fixed. Note that $\eta_3 \equiv 2\pi n_2/\lambda_0$ is proportional to the molecular density, whereas $|E_0|^2 \propto F_i$ is inversely proportional to the molecular density in each filament. Thus α is independent of both total input energy and pressure. Integrating both sides of Eq. (8) over area gives

$$E_{\text{SPM}} = E_0 + \alpha E_i, \quad (10)$$

where E_{SPM} is the total self-phase-modulation energy, E_i is the total input energy, and E_0 is a constant determined by the self-phase-modulation energy produced before the input beam breaks up. Thus in the high-energy regime the self-phase-modulation production rate is given by

$$\frac{dE_{\text{SPM}}}{dE_i} = \alpha \quad (11)$$

and is independent of both input energy and pressure, as is seen in Fig. 5. The numerical value of α for pure

hydrogen, indicated by the high-input-energy results in Fig. 5, is ~ 0.7 .

A similar argument accounts for the behavior of the Stokes radiation in this regime. As in the case of self-phase modulation, the Stokes-radiation energy density in each filament can be written as

$$F_{\text{Stokes}} = \beta F_i, \quad (12)$$

where, again, β is independent of total input energy because the input energy density per filament is fixed. Following the same procedure as above leads to the result

$$\frac{dE_{\text{Stokes}}}{dE_i} = \beta, \quad (13)$$

which explains the linear dependence of the Stokes-radiation energy on the input energy observed in the high-energy regime (see Figs. 3 and 6). For pure hydrogen (Fig. 3) β is roughly independent of pressure and equal to ~ 0.2 . However, because the addition of argon decreases the total energy per filament in the high-energy regime without enhancing Stokes production, β decreases with increasing argon partial pressure (Fig. 6).

In conclusion, we have demonstrated the effect of self-phase modulation and self-focusing on transient stimulated Raman scattering in high-pressure hydrogen gas. In the low-energy regime self-phase modulation has little effect on transient stimulated Raman scattering, but self-focusing enhances it. Adding argon to the hydrogen enhances self-focusing, which in turn lowers the Stokes-radiation threshold. Increasing the input energy results in suppression of transient stimulated Raman scattering by self-phase modulation, an effect that is strongly enhanced by the addition of argon. At high input energies the beam breaks up into fine filaments, leading to a partial recovery of the Stokes radiation as well as a linear dependence of both the self-phase modulation and Stokes energies on the input energy. This study shows that the complicated interplay among transient stimulated Raman scattering, self-phase modulation, and self-focusing must be taken into account when a system is designed to use stimulated Raman scattering with ultrashort pulses.

APPENDIX A

With no pump-pulse depletion assumed to be due to the Stokes-radiation production, the coupled equations for describing transient stimulated Raman scattering are given by¹

$$E_p(z, t) = g_p(z) f_p(t), \quad (A1)$$

$$\frac{1}{v_s} \frac{\partial E_s}{\partial t} + \frac{\partial E_s}{\partial z} = i\eta_1 E_p Q^*, \quad (A2)$$

$$\left(\frac{\partial}{\partial t} + \Gamma \right) Q^* = -i\eta_2 E_p^* E_s, \quad (A3)$$

where E_p and E_s are the field amplitudes of the pump and the Stokes waves, respectively, Q is the oscillation amplitude of the normal coordinate in the molecular vibration, v_s is the group velocity of the Stokes wave, and Γ corresponds to the linewidth of the spontaneous Raman

scattering of the medium. The coupling parameters η_1 and η_2 are given by

$$\eta_1 = \frac{N}{2\omega_v} \frac{\partial \alpha}{\partial Q}, \quad (A4)$$

$$\eta_2 = \frac{2\pi N \omega_s^2}{c^2 k_s} \frac{\partial \alpha}{\partial Q}, \quad (A5)$$

where N is the number density of Raman-active molecules in the medium, α is the polarizability associated with Raman-active vibrations, ω_s and k_s are the radial frequency and the amplitude of the wave vector of the Stokes radiation, respectively, and ω_r is the radial frequency of the vibration.

Transforming the temporal coordinate t into a new coordinate $t' = t - z/v_s$, the coupled equations (A1)–(A3) become

$$E_p(z, t') = g_p(z) f_p(t'), \quad (A6)$$

$$\frac{\partial E_s}{\partial z} = i\eta_1 E_p Q^*, \quad (A7)$$

$$\left(\frac{\partial}{\partial t'} + \Gamma \right) Q^* = -i\eta_2 E_p^* E_s. \quad (A8)$$

Integrating both sides of Eq. (A8), we get

$$Q^*(z, t') = -i\eta_2 \exp(-\Gamma t') \int_{-\infty}^{t'} E_p^*(z, t'') E_s(z, t'') \times \exp(\Gamma t'') dt''. \quad (A9)$$

Substituting Eq. (A9) into Eq. (A7) yields

$$\frac{\partial E_s}{\partial z} = \eta_1 \eta_2 E_p(z, t') \exp(-\Gamma t') \int_{-\infty}^{t'} E_p^*(z, t'') E_s(z, t'') \times \exp(\Gamma t'') dt''. \quad (A10)$$

Taking the temporal derivative of the above equation, we find

$$\frac{\partial^2 E_s}{\partial z \partial t'} + \left(\Gamma - \frac{1}{E_p} \frac{\partial E_p}{\partial t'} \right) \frac{\partial E_s}{\partial z} = \eta_1 \eta_2 |E_p|^2 E_s. \quad (A11)$$

Substituting Eq. (A6) into Eq. (A11) yields

$$\frac{\partial^2 E_s}{\partial z \partial t'} + \left(\Gamma - \frac{1}{f_p} \frac{\partial f_p}{\partial t'} \right) \frac{\partial E_s}{\partial z} = \eta_1 \eta_2 |E_p|^2 E_s, \quad (A12)$$

from which we obtain

$$\frac{\partial}{\partial z} \left\{ \frac{\partial}{\partial t'} \left[E_s \frac{\exp(\Gamma t')}{f_p(t')} \right] \right\} = \eta_1 \eta_2 |E_p|^2 E_s \frac{\exp(\Gamma t')}{f_p(t')}. \quad (A13)$$

We next define

$$U(z, t') \equiv E_s(z, t') \frac{\exp(\Gamma t')}{f_p(t')}, \quad (A14)$$

which together with Eq. (A13) gives

$$\frac{\partial^2}{\partial z \partial t'} U(z, t') = \eta_1 \eta_2 |E_p|^2 U(z, t'). \quad (A15)$$

Transforming the temporal and the spatial coordinates using the relations

$$\tau(t') \equiv \sqrt{\eta_1 \eta_2} \int_{-\infty}^{t'} |f_p(t'')|^2 dt'', \quad (\text{A16})$$

$$\xi(z) \equiv \sqrt{\eta_1 \eta_2} \int_0^z |g_p(z')|^2 dz', \quad (\text{A17})$$

we finally obtain

$$\frac{\partial^2}{\partial \xi \partial \tau} U(\xi, \tau) = U(\xi, \tau). \quad (\text{A18})$$

Proceeding similarly, we derive an analogous relation for the normal coordinate

$$\frac{\partial^2}{\partial \xi \partial \tau} W(\xi, \tau) = W(\xi, \tau), \quad (\text{A19})$$

where

$$W(z, t') \equiv Q^*(z, t') \frac{\exp(\Gamma t')}{g_p^*(z)}. \quad (\text{A20})$$

Both Eqs. (A18) and (A19) are standard hyperbolic equations that can be solved for arbitrary initial conditions by Riemann's method.¹⁴ A special solution of this type of equation is the modified Bessel function $I_0(2\sqrt{\xi\tau})$. The solution must satisfy the following initial conditions: (1) the system starts in the ground vibrational state so that $\partial \mathcal{E}_S / \partial z = Q^*(z) = 0$ for $t' \rightarrow -\infty$, and (2) the Stokes radiation field at the edge of the Raman cell $z = 0$ is the constant background spontaneous noise $\mathcal{E}_S(0, t')$. Through the coordinate transformation relations (A16) and (A17) the initial conditions can be converted into

$$\left. \frac{\partial U}{\partial \xi} \right|_{\tau=0} = W|_{\tau=0} = 0, \quad (\text{A21})$$

$$U|_{\xi=0} = \mathcal{E}_S(0, t') \frac{\exp(\Gamma t')}{F_p(t')},$$

$$\left. \frac{\partial W}{\partial \tau} \right|_{\xi=0} = -i \left(\frac{\eta_2}{\eta_1} \right)^{1/2} \mathcal{E}_S(0, t') \frac{\exp(\Gamma t')}{F_p(t')}. \quad (\text{A22})$$

Solving the hyperbolic equations (A18) and (A19) subject to the above boundary conditions, we obtain the solutions

$$\begin{aligned} \mathcal{E}_S(z, t') &= \mathcal{E}_S(0, t') + [\eta_1 \eta_2 \xi(z)]^{1/2} f_p(t') \\ &\times \int_{-\infty}^{t'} \exp[-\Gamma(t' - t'')] f_p^*(t'') \mathcal{E}_S(0, t'') \\ &\times \frac{I_1(2\{\xi(z)[\tau(t') - \tau(t'')]\}^{1/2})}{[\tau(t') - \tau(t'')]^{1/2}} dt'', \end{aligned} \quad (\text{A23})$$

$$\begin{aligned} Q^*(z, t') &= i \eta_1 \int_{-\infty}^{t'} \exp[-\Gamma(t' - t'')] f_p^*(t'') \mathcal{E}_S(0, t'') \\ &\times g_p(z) I_0(2\{\xi(z)[\tau(t') - \tau(t'')]\}^{1/2}) dt''. \end{aligned} \quad (\text{A24})$$

From the asymptotic behavior of the Bessel function, we can obtain an approximate relation for the Stokes field amplitude in the limit of large transient gain. In this limit the Stokes-energy amplification $E_{\text{Stokes}}(z)/E_{\text{Stokes}}(0)$ is approximately proportional to $\exp[G_{\text{trans}}(z)]/[G_{\text{trans}}(z)]^3$,² where the transient stimulated Raman gain coefficient G_{trans} is given by

$$G_{\text{trans}}(z) \approx 4 \left[\eta_1 \eta_2 \int_0^z |g_p(z')|^2 dz' \int_{-\infty}^{\infty} |f_p(t'')|^2 dt'' \right]^{1/2}. \quad (\text{A25})$$

APPENDIX B

In this appendix we derive an expression for the self-phase-modulation energy in hydrogen, assuming that there is no self-focusing and that effects from stimulated Raman scattering can be neglected. The electric field of the incident laser pulse can be written as

$$\mathcal{E}_p(t) = \mathcal{E}_0 f(t) \exp(-i\omega_0 t + ibt^2), \quad (\text{B1a})$$

with

$$f(t) = \exp(-t^2/\tau^2), \quad (\text{B1b})$$

where ω_0 is the center radial frequency of the light wave, b is the chirp parameter, and we have assumed that the pulse shape is Gaussian. Taking the Fourier transform of Eqs. (B1) yields

$$\begin{aligned} S(\omega) &= \int_{-\infty}^{\infty} \mathcal{E}_p(t) \exp(i\omega t) dt \\ &= \frac{\mathcal{E}_0 \sqrt{\pi} \tau}{(1 - ib\tau^2)^{1/2}} \exp \left[-\frac{(\omega - \omega_0)^2 \tau^2}{4(1 - ib\tau^2)} \right] \end{aligned} \quad (\text{B2})$$

for the frequency dependence of the electric field of the laser pulse. The power spectrum is thus

$$|S(\omega)|^2 = \frac{|\mathcal{E}_0|^2 \pi \tau^2}{(1 + b^2 \tau^4)^{1/2}} \exp \left[-\frac{(\omega - \omega_0)^2 \tau^2}{2(1 + b^2 \tau^4)} \right]. \quad (\text{B3})$$

After the laser pulse is sent through the nonlinear-optical medium, the electrical field of the pump pulse becomes

$$\mathcal{E}_p'(t) = \mathcal{E}_0 f(t) \exp[i\eta_3 |\mathcal{E}_0|^2 f(t)^2 L] \exp(-i\omega_0 t + ibt^2), \quad (\text{B4})$$

where L is the interaction length and $\eta_3 \equiv 2\pi n_2/\lambda_0$. The frequency dependence of the electric field associated with self-phase modulation is then

$$\begin{aligned} S'(\omega) &= \int_{-\infty}^{\infty} \mathcal{E}_0 f(t) \exp[i\eta_3 |\mathcal{E}_0|^2 f(t)^2 L] \exp(-i\omega_0 t + ibt^2) \\ &\times \exp(i\omega t) dt. \end{aligned} \quad (\text{B5})$$

Assuming a Gaussian form for $f(t)$ as in Eq. (B1b), the dominant contribution to the integral in Eq. (B5) occurs when $t^2/\tau^2 < 1$. So we may approximate $f(t)$ in this integral by $1 - t^2/\tau^2$, yielding

$$\begin{aligned} S'(\omega) &\approx \frac{\mathcal{E}_0 \sqrt{\pi} \tau}{(1 + i2\eta_3 |\mathcal{E}_0|^2 L - ib\tau^2)^{1/2}} \\ &\times \exp \left[-\frac{(\omega - \omega_0)^2 \tau^2}{4(1 + i2\eta_3 |\mathcal{E}_0|^2 L - ib\tau^2)} \right]. \end{aligned} \quad (\text{B6})$$

Using the spectrum in relation (B6), we can now derive an expression for F_{SPM} , the total energy density of the pulse minus the energy density in the original spectral region of the incident laser pulse. To calculate the final energy density remaining in the original spectral region, we integrate the final power spectrum $|S'(\omega)|^2$ by using the weighting function $\exp[-(\omega - \omega_0)^2 \tau^2 / 2(1 + b^2 \tau^4)]$

from the original power spectrum in Eq. (B3). Let us define the energy density inside the original spectral width of the incident pulse to be F_δ . Using Eq. (B5), we get

$$\begin{aligned} F_\delta &= B \int_{-\infty}^{\infty} |S'(\omega)|^2 \exp\left[-\frac{(\omega - \omega_0)^2 \tau^2}{2(1 + b^2 \tau^4)}\right] d\omega \\ &= \frac{B \pi \tau^2 |\mathcal{E}_0|^2}{[1 + (2\eta_3 |\mathcal{E}_0|^2 L - b\tau^2)^2]^{1/2}} \int_{-\infty}^{\infty} \exp\left[-\frac{(\omega - \omega_0)^2 \tau^2}{2(1 + b^2 \tau^4)}\right] \\ &\quad \times \exp\left\{\frac{(\omega - \omega_0)^2 \tau^2}{2[1 + (2\eta_3 |\mathcal{E}_0|^2 L - b\tau^2)^2]}\right\} d\omega \\ &= \pi \sqrt{2\pi} B \tau |\mathcal{E}_0|^2 \left[\frac{1 + b^2 \tau^4}{2 + b^2 \tau^4 + (2\eta_3 |\mathcal{E}_0|^2 L - b\tau^2)^2} \right]^{1/2}, \end{aligned} \quad (\text{B7})$$

where B is a normalization constant. In our case, the spectral broadening due to the chirp is negligible compared with the effect of self-phase modulation.⁹ So ignoring the chirp parameter b , F_δ becomes

$$F_\delta \approx \frac{\pi \sqrt{\pi} B \tau |\mathcal{E}_0|^2}{[1 + 2(\eta_3 |\mathcal{E}_0|^2 L)^2]^{1/2}}. \quad (\text{B8})$$

We define F_{SPM} to be the energy density associated with frequency components other than those composing the center spectral range of the incident pulse,

$$F_{\text{SPM}} \equiv F_i - F_\delta, \quad (\text{B9})$$

where F_i is the energy density of the incident laser pulse. In the absence of self-phase modulation, $F_i = F_\delta$, which allows us to determine the normalization constant:

$$F_i = \frac{cn_0 \tau}{2\sqrt{2\pi}} |\mathcal{E}_0|^2 = \pi \sqrt{\pi} B \tau |\mathcal{E}_0|^2, \quad (\text{B10})$$

so $B = cn_0/(2\sqrt{2}\pi^2)$. Therefore, without self-focusing, the self-phase-modulation energy density is given by

$$F_{\text{SPM}} \approx F_i - \frac{F_i}{[1 + 2(\eta_3 |\mathcal{E}_0|^2 L)^2]^{1/2}}. \quad (\text{B11})$$

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REFERENCES AND NOTES

1. J. C. White in *Tunable Lasers*, L. F. Mollenauer and J. C. White, eds. (Springer-Verlag, Berlin, 1987), pp. 115–207.
2. M. D. Duncan, R. Mahon, L. L. Tankersley, and J. Reintjes, "Transient stimulated Raman amplification in hydrogen," *J. Opt. Soc. Am. B* **5**, 37 (1988).
3. R. L. Carman, F. Shimizu, C. S. Wang, and N. Bloembergen, "Theory of Stokes pulse shapes in transient stimulated Raman scattering," *Phys. Rev. A* **2**, 60 (1970).
4. S. A. Akhmanov, K. N. Drabovich, A. P. Sukhorukov, and A. S. Chirkin, "Stimulated Raman scattering in a field of ultrashort light pulses," *Sov. Phys. JETP* **32**, 266 (1971).
5. S. A. Akhmanov, K. N. Drabovich, A. P. Sukhorukov, and A. K. Shchednova, "Combined effects of molecular relaxation and medium dispersion in stimulated Raman scattering of ultrashort light pulses," *Sov. Phys. JETP* **35**, 279 (1972).
6. P. G. May and W. Sibbett, "Transient stimulated Raman scattering of femtosecond laser pulses," *Appl. Phys. Lett.* **43**, 624 (1983).
7. J. H. Glowina, J. Misewich, and P. P. Sorokin, "Subpicosecond time-resolved infrared spectral photography," *Opt. Lett.* **12**, 19 (1987).
8. Our amplifier is similar to the one described in M. M. Murnane and R. W. Falcone, "High-power femtosecond dye-laser system," *J. Opt. Soc. Am. B* **5**, 1573 (1988).
9. H. J. Lehmeier, W. Leupacher, and A. Penzkofer, "Nonresonant third order hyperpolarizability of rare gases and N₂ determined by third harmonic generation," *Opt. Commun.* **56**, 67 (1985).
10. Data on spontaneous Raman scattering at various pressures in hydrogen are given in F. De Martini, F. Simoni, and E. Santamato, "High resolution nonlinear spectroscopy. Dicke narrowing and dispersion of the third-order nonlinear susceptibility of H₂ near the Q₀₁(1) vibrational resonance," *Opt. Commun.* **9**, 176 (1973). Assuming negligible inhomogeneous linewidth broadening in gases, the dephasing time is given by $T_2 = 1/\pi\Delta\nu$, where $\Delta\nu$ is the homogeneous linewidth.
11. Assuming that the uncompressed laser pulses have a Gaussian-shaped temporal profile described by $\mathcal{E}(t) = \mathcal{E}_0 \exp(-t^2/\tau^2) \exp(-i\omega t + ibt^2)$, the spectral width that is due to self-phase modulation is approximately given by $-2b\tau_p + 8\pi n_2 L |\mathcal{E}|^2 / \lambda \tau_p \exp(1/2)$, where b is the chirp parameter ($b = 10 \text{ ps}^{-2}$), τ_p is the pulse width, n_2 is the nonlinear refractive index, L is the interaction length, \mathcal{E} is the field strength of the laser beam, and λ is the laser wavelength. Substituting values for these parameters corresponding to our experimental conditions indicates that the first term is at least 5 orders of magnitude smaller than the second term, which is independent of chirp. For the theory of linear pulse propagation, see A. E. Siegman, *Lasers* (University Science, Mill Valley, Calif., 1986), Chap. 9.
12. S. W. Allendorf and A. Szöke, "High-intensity multiphoton ionization of H₂," *Phys. Rev. A* **44**, 518 (1991).
13. P. Lallemand and N. Bloembergen, "Self-focusing of laser beams and stimulated Raman gain in liquids," *Phys. Rev. Lett.* **15**, 1010 (1965).
14. J.-K. Wang, "Femtosecond nonlinear optics in gases and solids," Ph.D. Dissertation (Harvard University, Cambridge, Mass., 1992), Chap. 5.
15. A. J. Campillo, S. L. Shapiro, and B. R. Suydam, "Periodic breakup of optical beams due to self-focusing," *Appl. Phys. Lett.* **23**, 628 (1973).
16. B. R. Suydam, "Self-focusing of very powerful laser beams II," *IEEE J. Quantum Electron.* **QE-10**, 837 (1974).