

**Intensity Effects on the Stimulated Four Photon Spectra
Generated by Picosecond Pulses in Optical Fibers**

**Patrice L. Baldeck
Robert P. Alfano**

Reprinted from
JOURNAL OF LIGHTWAVE TECHNOLOGY
Vol. LT-5, No. 12, December 1987

Intensity Effects on the Stimulated Four Photon Spectra Generated by Picosecond Pulses in Optical Fibers

PATRICE L. BALDECK AND ROBERT R. ALFANO

Abstract—The intensity dependence of stimulated four photon mixing (SFPM) spectra generated in 15 m of a 4-mode optical fiber by 25-ps pulses has been investigated. Despite the shortness of pulses, the SFPM conversion was highly efficient due to the intrinsic phase matching condition. In addition to usual features of SFPM spectra generated by nanosecond pump pulses, picosecond SFPM spectra were broadened by self phase modulation (SPM) and cross phase modulation (XPM). At the highest pump powers, intensity saturated frequency continua, arising from the combined effects of SFPM, stimulated Raman scattering (SRS), SPM, and XPM were generated all over the visible spectrum.

I. INTRODUCTION

STIMULATED four photon mixing (SFPM) is an ideal process for designing optical-fiber parametric amplifiers and frequency converters. SFPM is produced when two high-intensity pump photons are coupled by the third-order susceptibility $\chi^{(3)}$ to generate a Stokes photon and an anti-Stokes photon. The frequency shifts of the SFPM waves are determined by the phase matching conditions which depend on the optical geometry. In 1970, SFPM was produced in glass by Alfano and Shapiro using picosecond pulses [1]. Later on, SFPM has been successfully demonstrated by a number of investigators in few-mode [2], birefringent [3], and single-mode [4] optical fibers. Most of the earlier experiments using optical fibers have been performed with nanosecond pulses. Recently, large-frequency shifts have been demonstrated with 25-ps pulses [5]; however, the spectral dependence on the input intensity was not investigated.

In the present report, the intensity dependence of SFPM spectra generated by 25-ps pulses in 15 m of a 4-mode optical fiber was experimentally investigated. For such short pulses, it is well known that self-phase modulation (SPM) broadens the spectra [6]. It has been predicted that cross phase modulations (XPM) should enhance the broadening [7].¹ We have studied the broadening of SFPM lines and the formation of frequency supercontinua. Several new properties of SFPM were noticed in this report.

Manuscript received November 11, 1986; revised May 17, 1987. This work was partially supported by the Hamatsu Photonics KK., and by Corning Glass through the University Gift Program.

The authors are with the Institute for Ultrafast Spectroscopy and Lasers and Photonics Application Laboratory, Departments of Physics and Electrical Engineering, The City College of The City University of New York, New York, NY 10031.

IEEE Log Number 8716917.

¹Note: [7, eq. (21)] has been corrected in this paper.

1) Numerous sets of SFPM frequencies could be simultaneously generated. Up to 10 sets of SFPM frequencies with frequency shifts ranging from 60 to 3800 cm^{-1} were generated. 2) The phase matching condition could be satisfied with pump and SFPM photons propagating in totally different modes. 3) For adequate frequency shifts, anti-Stokes lines could be depleted by stimulated Raman scattering and the energy transferred to their Stokes companions. 4) Frequency continua were produced with self limited intensities.

II. EXPERIMENTAL METHOD

The experimental setup is described as follows. A Quantel frequency-doubled mode-locked Nd:YAG laser produced 30-ps pulses. A $\times 20$ microscope lens was used to couple the laser beam into the optical fiber. The spectra of the output pulses were measured using an 1 m-1200/mm grating spectrometer. Spectra were recorded on photographic film and with an optical multichannel analyzer OMA 2. Average powers coupled in the fiber were measured with a power meter at the optical fiber output.

The optical fiber had a core diameter of 8 μm and a normalized frequency $V = 4.44$ at 532 nm. At this wavelength, the 4 first LP modes (LP_{01} , LP_{11} , LP_{21} , and LP_{02}) were allowed to propagate. The theoretical SFPM frequency shifts were calculated for all mode combinations using a generalization of equations derived in [2].

III. RESULTS AND DISCUSSION

A. Observations

Spectra are displayed in Figs. 1, 2, and 3. At low intensity $I < 10^8 \text{ W/cm}^2$, the output spectrum contains only the pump wavelength $\lambda = 532 \text{ nm}$ (Fig. 1(a)). At approximately $5 \times 10^8 \text{ W/cm}^2$, 3 sets of symmetrical SFPM lines (at $\Omega = 50, 160, \text{ and } 210 \text{ cm}^{-1}$) and the first SRS Stokes line (at 440 cm^{-1}) appear (Fig. 1(b) and 1(c)). As the intensity increases the SFPM and SRS lines broaden, and a Stokes frequency continuum is generated (Fig. 1(d) and 1(e)). Above an intensity threshold of $20 \times 10^8 \text{ W/cm}^2$, new sets of SFPM lines appear on the Stokes and anti-Stokes sides with frequency shifts ranging from 2700 to 3865 cm^{-1} . Finally, the large shift lines merge (Fig. 1(f)) and contribute to the formation of a 4000 cm^{-1} frequency continuum (Fig. 1(g)). Fig 2 shows how the large-

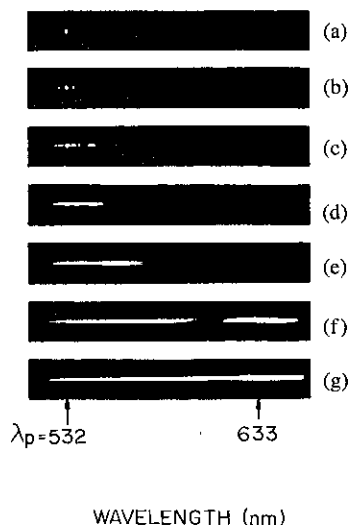


Fig. 1. Evolution of a stimulated four photon spectrum with the increase of the pulse intensity. (a) $I < 10^8$ W/cm². (b) and (c) $I = 5 \times 10^8$ W/cm². (d) $I = 10 \times 10^8$ W/cm². (e) $I = 15 \times 10^8$ W/cm². (f) $I = 30 \times 10^8$ W/cm². (g) $I = 35 \times 10^8$ W/cm².

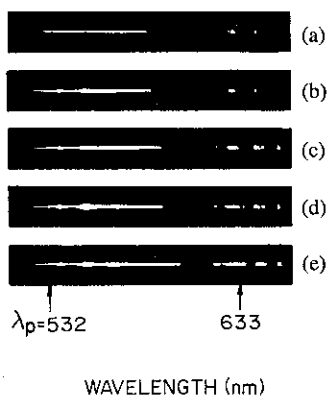


Fig. 2. (a)-(e) Sequence of the large-shift SFPM-lines broadening. The peak intensity is increased from $I = 20 \times 10^8$ W/cm² in (a) to $I = 30 \times 10^8$ W/cm² in (e) in steps of 2.5×10^8 W/cm².

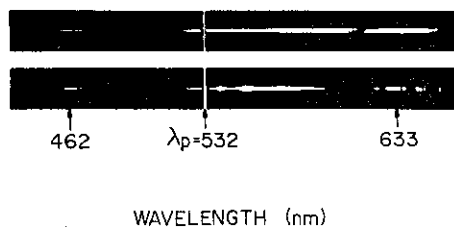


Fig. 3. Examples of large-shift Stokes lines with their corresponding anti-Stokes lines. Photographs of the Stokes and anti-Stokes regions were spliced together.

Stokes-shift SFPM lines are generated and broaden when the pump intensity increases from 20×10^8 W/cm² to 30×10^8 W/cm². Fig. 3 gives two examples of complete spectra including the large-shift anti-Stokes and Stokes lines.

The SFPM shifts observed experimentally correspond well with the calculated shifts as shown in Table I. Notice that the shifts of 1950 and 2270 cm⁻¹ correspond to the respective mode distributions LP₀₁(pump)-LP₀₂(pump)-

TABLE 8
COMPARISON OF THE CALCULATED AND EXPERIMENTAL SFPM SHIFTS

| CALCULATED | | | PHOTOGRAPH | | OMA 2 | |
|------------------------------|------------------|------------------|------------------|------------------|------------------|------------------|
| Ω (cm ⁻¹) | λ_a (nm) | λ_s (nm) | λ_a (nm) | λ_s (nm) | λ_a (nm) | λ_s (nm) |
| 40 | 531 | 533 | 530 | 534 | 530 | 534 |
| 170 | 527 | 537 | 528 | 536 | 527 | 537 |
| 210 | 526 | 538 | 526 | 538 | 526 | 538 |
| 815 | 510 | 556 | | | | |
| 1150 | 501 | 567 | | | | |
| 1220 | 500 | 569 | | | | |
| 1470 | 493 | 577 | | | 499 | |
| 1950 | 482 | 594 | | | 494 | |
| 2120 | 478 | 600 | | | 485 | |
| 2270 | 475 | 605 | | | *478 | |
| 2300 | 474 | 606 | | | 475 | |
| 2440 | 471 | 611 | | | | |
| 2580 | 468 | 617 | | | *463 | *617 |
| 2700 | 465 | 621 | 467 | 622 | 466 | 622 |
| 2990 | 459 | 633 | *459 | *630 | *460 | *631 |
| 3130 | 456 | 638 | 455 | 630 | 456 | 638 |
| 3230 | 454 | 643 | 453 | *643 | *454 | *643 |
| 3330 | 452 | 647 | 452 | 647 | 453 | 648 |
| 3430 | 450 | 651 | | | 451 | |
| 3460 | 449 | 652 | | 652 | | |
| 3550 | 447 | 656 | 448 | 655 | 448 | 656 |
| 3640 | 446 | 660 | | | | |
| 3865 | 441 | 670 | 443 | | 444 | |
| 3950 | 440 | 674 | | | | |
| 4050 | 438 | 678 | | | | |
| 4135 | 436 | 682 | | | | |
| 4215 | 435 | 686 | | | | |
| 4570 | 428 | 703 | | | | |
| 4645 | 427 | 707 | | | | |
| 4716 | 425 | 710 | | | | |

* Multiple Lines.

LP₁₁(Stokes)-LP₂₁(anti-Stokes) and LP₀₁(pump)-LP₂₁(pump)-LP₁₁(Stokes)-LP₀₂(anti-Stokes). In both cases, all waves propagated in totally different modes. More generally, the SFPM shifts observed in this experiment corresponded to the most diverse mode combinations. Thus, there are many more mode combinations which satisfy the SFPM phase-matching than the usual pump-divided and single-mode pump processes [2]. There are more frequency shifts allowed than expected earlier, and the potentiality of SFPM to generate new frequencies are even more important.

The generation of SFPM lines is very sensitive to the beam-optical fiber coupling, which determines the mode excitation. This is why some SFPM lines predicted by the theory were observed only during the experiments recorded on the photographic film, or only during those recorded on the OMA graphs, or not observed at all. The Stokes SFPM lines with medium shifts ($1000 \text{ cm}^{-1} < \Omega < 2500 \text{ cm}^{-1}$) are not listed in Table I for they were indiscernable in the frequency continuum.

B. Depletion of Anti-Stokes SFPM Lines at High Intensities

SFPM spectra centered around the laser pump and measured with an OMA 2 are displayed in Fig. 4. In Fig. 4(a),

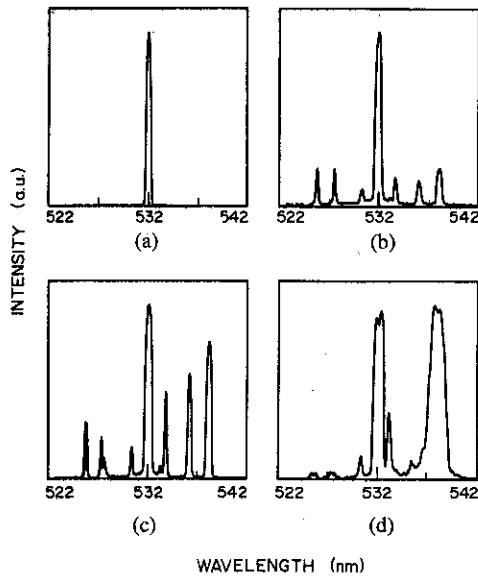


Fig. 4. (a)-(d) Depletion of anti-Stokes SFPM lines for increasing intensities at $I \approx 5 \times 10^8 \text{ W/cm}^2$. The pump intensity is slightly increased between (a) and (d).

the pump intensity is below the SFPM intensity threshold and the spectrum contains only the pump wavelength λ_p . In Fig. 4(b), the spectrum contains the 3 small-shift SFPM line sets which correspond to the mixed mode distribution. In this configuration, the SFPM phase-matching condition is satisfied when the pump power is divided in the Stokes and the anti-Stokes modes [2]. For example, the smallest shift ($\Omega = 40 \text{ cm}^{-1}$) is obtained for the mode distribution LP₂₁(pump and anti-Stokes)-LP₁₁(pump and Stokes).

Fig. 4(c) and 4(d) shows how the SFPM spectrum changes when the intensity is slightly increased. In a first step, all anti-Stokes and Stokes lines are amplified with the increase of the nonlinear interaction, but nonequally. The Stokes lines become more intense than the anti-Stokes lines, and the amplification is more sensible for the largest shifts (Fig. 4(c)). In a second step, the second and third anti-Stokes lines deplete while the third Stokes line becomes much stronger and broader (Fig. 4(d)). The evolution of asymmetric Stokes and anti-Stokes lines are unusual since it has been established that SFPM, contrary to SRS, might generate similar Stokes and anti-Stokes lines [1]. However, it should be noticed that the frequency spacing between these anti-Stokes and Stokes lines is about the 440 cm^{-1} of the Raman shift in silica. Thus, it is very likely that the anti-Stokes photon population is depleted by stimulated Raman scattering while the SFPM is building up.

Several line splittings were observed as shown in Fig. 4(c) for the second anti-Stokes and first Stokes lines, and Fig. 4(d) for the pump and SFPM. The splittings of Fig. 4(d) are characteristic of SPM spectral broadening. However, SPM does seem responsible for the splittings observed in Fig. 4(c). An explanation could be that the different mode combinations lead to about-equal frequency

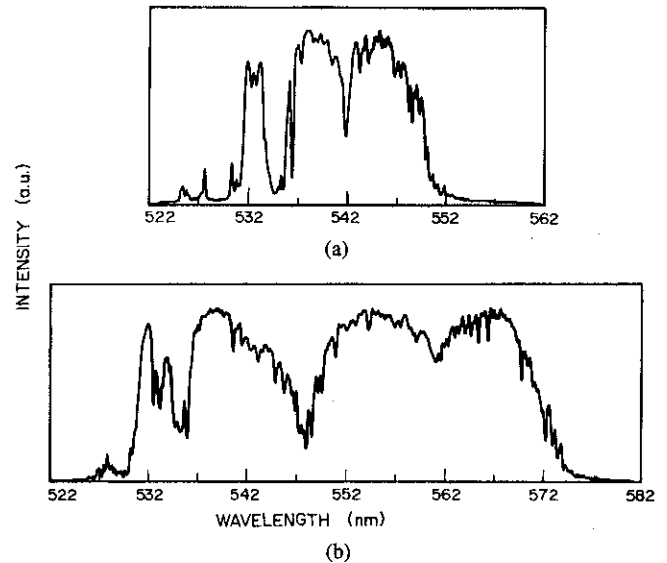


Fig. 5. Supercontinuum generation. (a) The pump, SFPM, and first SRS Stokes lines are broadened at $I = 10 \times 10^8 \text{ W/cm}^2$. (b) The broadened second and third SRS Stokes lines appear and extend the spectrum toward the Stokes wavelengths at $I = 15 \times 10^8 \text{ W/cm}^2$.

shifts. For example, the shift $\Omega = 170 \text{ cm}^{-1}$ is obtained with the mode combination LP₀₁(pump and Stokes)-LP₁₁(pump and anti-Stokes), and with the mode combination LP₀₂(pump and Stokes)-LP₁₁(pump and anti-Stokes). Another explanation could be the degeneracy of LP modes, which are superposition of TE, TM, EH, and HE modes.

C. Supercontinuum Generation

Fig. 5 shows the development of a Stokes continuum from the combined effects of SFPM, SRS, SPM, and XPM. As the intensity is increased, the pump, SFPM, and first SRS lines broaden and merge (Fig. 5(a)). For stronger pump intensities, the SFPM-SRS continuum is duplicated by stimulated Raman scattering, and the continuum expands towards the lowest optical frequencies (Fig. 5(b)). As shown, the maximum intensities of new frequencies are self limited.

The broadening of the SFPM and SRS lines arises from self and cross phase modulation effects. It is established that spectral broadenings generated by SPM are inversely proportional to the pulse duration and linearly proportional to the pump intensity [6]. In this experiment, SPM effects are important because of the pump pulse shortness (30 ps) and intensity (100 MW/cm^2). Moreover, it has been predicted that XPM enhances the spectral broadening in the ratio $\Delta\omega_{\text{Raman}}/\Delta\omega_{\text{pump}} \approx 2$ [7]. The experimental ratios for the SFPM and SRS broadenings are, respectively, 2.7 and 3.6 (Fig. 5(a)). Furthermore, the modulation which is seen in the continuum spectrum fits well with the spectrum modulation predicted by phase modulation theories [6].

Fig. 6 shows the spectral broadening of the anti-Stokes SFPM line of $\lambda = 460 \text{ nm}$ ($\Omega = 2990 \text{ cm}^{-1}$). This line is a large-shift SFPM anti-Stokes line generated simulta-

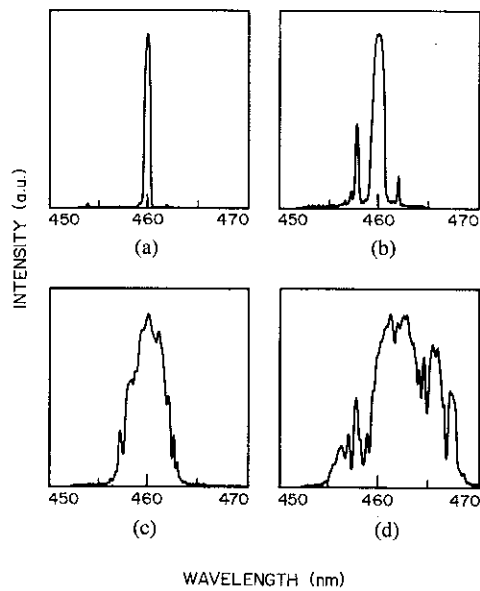


Fig. 6. (a)-(d) Spectral broadening of the anti-Stokes SFPM line generated at 460 nm. The pulse peak intensity increases from $20 \times 10^8 \text{ W/cm}^2$ (a) to $30 \times 10^8 \text{ W/cm}^2$ (d) in steps of $2.5 \times 10^8 \text{ W/cm}^2$.

neously with the $\lambda = 633\text{-nm}$ SFPM Stokes line by the laser pump of $\lambda = 532 \text{ nm}$ (see Fig. 3). The corresponding frequency shift and mode distribution are $\Omega = 2990 \text{ cm}^{-1}$ and LP_{01} (pump)- LP_{11} (Stokes and anti-Stokes), respectively. From Fig. 6(a) to 6(d), the peak intensity of the $\lambda = 460 \text{ nm}$ line increases from approximately 20 to $30 \times 10^8 \text{ W/cm}^2$ in steps of $2.5 \times 10^8 \text{ W/cm}^2$. In Fig. 6(a), the spectrum contains only the 460-nm SFPM line generated by the laser pump ($\lambda = 532 \text{ nm}$). In Fig. 6(b), the line begins to broaden and two symmetrical lines appear with a frequency shift of 100 cm^{-1} . This set of lines is a new set of small-shift SFPM lines generated by the 460-nm SFPM line acting as a new pump wavelength. Fig. 6(c) and 6(d) shows significant broadening, by a combined action of SFPM, SPM, and XPM of the 460 nm into a frequency continuum. Similar effects were observed on the Stokes side as displayed in Fig. 2.

IV. SUMMARY

The intensity effects on SFPM spectra generated by 25-ps pulses propagating optical fibers have been experimentally investigated. In contrast to SFPM lines generated by nanosecond pulses, spectra were broadened by self and cross-phase modulations. Intensity saturated wide frequency continua, covering the whole visible spectrum, were generated for increasing intensities. Applications are for the design of wide-band optical amplifiers, the generation of "white" picosecond pulses, and the generation by pulse compression of femtosecond pulses at new wavelengths. In addition to broadening effects, several new properties of SFPM were also observed: 1) numerous sets of SFPM frequencies could be simultaneously generated, 2) the phase matching condition could be satisfied with pump and SFPM photons propagating in totally different modes, and 3) for adequate frequency shifts, anti-Stokes

SFPM lines depleted by stimulated Raman scattering and the energy were transferred to the companion Stokes. 4) Frequency continua had self limited intensities.

REFERENCES

- [1] R. R. Alfano and S. L. Shapiro, "Emission in the region 4000-7000 Å via four-photon coupling in glass," *Phys. Rev. Lett.*, vol. 24, pp. 584-587, 1970.
- [2] R. H. Stolen, "Phase-matched stimulated four-photon mixing," *IEEE J. Quantum Electron.*, vol. QE-11, pp. 100-103, 1975.
- [3] R. H. Stolen, M. A. Bosch, and C. Lin, "Phase-matching in birefringent fibers," *Opt. Lett.*, vol. 6, pp. 213-215, 1981.
- [4] K. Washio, K. Inoue, and T. Tanigawa, "Efficient generation near-IR stimulated light scattering in optical fibers pumped in low-dispersion region at $1.3 \mu\text{m}$," *Electron. Lett.*, vol. 16, pp. 331-333, 1980.
- [5] C. Lin, M. A. Bosch, "Large Stokes-shift stimulated four-photon mixing in optical fibers," *Appl. Phys. Lett.*, vol. 38, pp. 479-481, 1981.
- [6] R. R. Alfano and S. L. Shapiro, "Observation of Self Phase Modulation and small scale filaments in crystals and glasses," *Phys. Rev. Lett.*, vol. 24, pp. 592-594, 1970.
- [7] J. I. Gersten, R. R. Alfano, and Milivoj Belic, "Combined stimulated Raman scattering and continuum self-phase modulations," *Phys. Rev. A*, vol. 21, pp. 1222-1224, 1980.

*



Patrice L. Baldeck was born on February 16, 1956. He graduated the Maitrise d'Electronique, Electrotechnique et Automatique, in 1984, and received the Diplome d'Etudes Approfondies in optoelectronics and microwaves in 1985 from the Université Pierre et Marie Curie, Paris, France. His D.E.A. thesis research, done at the Thomson-CSF antennas laboratory in Cormeilles en Parisis, involved the automatic measurements of radom dielectric properties in the 8-18 GHz range.

Since September 1985, he has been a Research Assistant at the Institute for Ultrafast Spectroscopy and Photonics Application Laboratory, Electrical Engineering Department, The City College of New York. He has been working toward the Ph.D degree in electrical engineering on femtosecond and picosecond nonlinear fiber optics. His experimental interests involve ultrafast nonlinear optics, femtosecond pulse technology, active optical fibers, optical fiber sensors, and applications of laser technology.

*



Robert R. Alfano was born in New York, NY, on May 7 1941. He received the B.S. and M.S. degrees from Fairleigh Dickinson University, Teaneck, NJ, in 1963 and 1964, respectively, and the Ph.D. degree in physics from New York University, New York, in 1972.

From 1964 to 1972 he was a member of the Technical Staff at GTE Research Laboratories, Bayside, NY, where he conducted studies on linear and nonlinear optical properties of materials and picosecond spectroscopy and discovering the supercontinuum. Since 1972 he has been a Professor with the Department of Physics, the City College of New York, and in 1983 became the Herbert Kayser Professor of Electrical Engineering. In 1987 he became Distinguished Professor of Science and Engineering. He is the Director of both the Institute for Ultrafast Spectroscopy and Lasers and the Photonic Application Laboratory.

Dr. Alfano is a Fellow of the American Physical Society and was previously an Alfred P. Sloan Fellow.