Self- and coupled-phase modulations of ultrashort laser pulses

By R.R. Alfano and P.P. Ho

The use of light to control and to generate[1–6] light is most important in the fields of lasers and optical communication. One dramatic and still useful nonlinear optical phenomenon observed by Alfano and Shapiro[1,2] over 19 years ago arising from self-phase-modulation (SPM) is the supercontinuum. Self-,[1–5, 7–10] Induced-,[11,12] and Cross[13–17]-phase modulation processes are responsible for the generation of the spectral broadened supercontinuum ultrashort laser pulses. The supercontinuum spans over 10,000 cm^{-1} from the UV to IR with picosecond to femtosecond duration.[3,5] A femtosecond supercontinuum spectrum generated by passing a 120-fs 625-nm laser pulse through a 1-mm long carbon-tetrachloride liquid and dispersed with a grating (Fig. 1) is displayed on the front cover. In this report, we highlight the result and development of the current status of supercontinuum generation.

Many applications for ultrafast supercontinuum

Over the years, the ultrafast supercontinuum has been used in a variety of applications: time-resolved absorption spectroscopy,[18–21] excitation spectroscopy,[22–34] pulse compression,[3,25–28] squeezed states,[29] fiber diagnostics,[4] and spatial filtering for pulse shaping and information encoding,[30] to name a few. The frequency broadened pulses can play an important role in ranging, imaging, remote sensing, and communications. SPM is the key process behind the pulse compression[25] that leads to the generation of 6-fs laser pulses.[28]

Coupled modulation occurs when two laser pulses of different wavelengths propagate simultaneously in a nonlinear medium.

SPM occurs when a laser pulse propagates through a medium and disturbs the refractive index that changes the phase and frequency of the incident laser pulse. This causes a frequency sweep within the pulse envelope.

Coupled modulation occurs when two laser pulses of different wavelengths propagate simultaneously in a nonlinear medium. An intense pulse alters the phase and frequency of a weak pulse. We have further classified coupled modulations into two types: Induced-Phase-Modulation (IPM) and Cross-Phase-Modulation (XPM) are inter-related. In IPM, both laser pulses are controllable and generated outside the interaction medium. The delays and pulse walk-off between these two pulses can lead to the induced

R.R. ALFANO is Distinguished Professor of Science and Engineering and Director of the Institute for Ultrafast Spectroscopy and Lasers and Photonic Application Laboratory at the City College of CUNY, New York. P.P. HO is Associate Professor of Electrical Engineering at the same institution.

frequency shift. In XPM, a new probe frequency is generated inside a medium through Raman process or harmonic generation. This newly generated pulse in turn interacts with the incident laser becoming frequency and amplitude modulated. Recently, both IAM and XPM have been called XPM only.

Table 1 lists various phase modulation processes responsible for the spectral broadening. The bracket indicates the intensity of laser pulses at a given frequency. In the SPM process, the spectral broadening process involves only one primary wavelength. In the IPM process, strong and weak laser pulses at different wavelengths are generated outside the interaction material. These pulses enter the sample to induce additional spectral broadening on the weak pulse. Depending on time arrival, Stokes or anti-Stokes frequency shift can be controlled. In the Raman-XPM process, a principle wavelength generates a Stokes Raman line. The spectral width is broadened through the coupling of the laser wavelength and internally generated Raman wavelengths. In the SHG-XPM process, the second harmonic generated inside the sample becomes spectral broadened from the primary pulse due to the coupling from the second and third optical nonlinear susceptibilities.

Earlier observations

Major observations of the modulated spectral broadened laser pulses in different states of matter are listed in Table 2. Supercontinua spanning over 10,000 cm^{-1} were observed in liquids, glasses and crystals.[1] SPM spectra evolved through and beyond the electronic absorption were demonstrated by Alfano, et al.[31] using a picosecond laser pulse generated near electronic resonances in PrF_3 crystals in 1974. Temperature-dependent supercontinua in an anti-ferromagnetic crystal $KNiF_3$ was studied in 1976.[32] Ippen et al.[33] observed the supercontinuum in CS_2 filled fibers in 1974. Stolen and Lin[4] verified the SPM pro-

TABLE 1. *Types of Spectral Broadening from Phase Modulation Processes*

	INPUT	COUPLING	OUTPUT	PROCESS
Self-Phase Modulation (SPM)	$[\omega_1]$	$\xrightarrow[\chi^3]{}$	$[\omega_1 \pm \Delta\omega_1]$	: SPM
Induced Phase Modulation (IPM)	$[\omega_1]$	$\xrightarrow[\chi^3]{}$	$[\omega_1 \pm \Delta\omega_1]$	: SPM
	$[\omega_2]$ weak	$\xrightarrow[\chi^3]{}$	$[\omega_2 \pm \Delta\omega_2]$	: SPM
			where $\Delta\omega_1 >> \Delta\omega_2$	
	$[\omega_1] + [\omega_2]$	$\xrightarrow[\chi^3]{}$	$[\omega_1 \pm \Delta\omega_1]$	: SPM
		$+$	$[\omega_2 \pm \Delta\omega]$	: IPM
			where $\Delta\omega >> \Delta\omega_2$	
Cross-Phase Modulation (XPM)				
Raman XPM	$[\omega_1]$	$\xrightarrow[\chi^3]{}$	$[\omega_1 \pm \Delta\omega_1]$	: SPM
		$\xrightarrow[\chi^R]{}$	$[\omega_1 - \Omega \pm \Delta\omega_R]$	: XPM
			where $\Delta\omega_R \sim 2\,\Delta\omega_1$	
Second Harmonic XPM	$[\omega_1]$	$\xrightarrow[\chi^3]{}$	$[\omega_1 \pm \Delta\omega_1]$	: SPM
		$\xrightarrow[\chi^2]{}$	$[2\omega_1]$	: SHG
		$\xrightarrow[\chi^2, \chi^3]{}$	$[2\omega_1 \pm \Delta\omega_0]$	: XPM
			where $\Delta\omega_0 \sim 1{,}000\ cm^{-1}$.	

TABLE 2. *Brief History of Experimental Supercontinuum Generation*

PROCESS	YEAR	MATERIAL	WAVELENGTH LASER/ PULSEWIDTH	SPECTRAL	FREQUENCY SHIFT
SPM[1]	1969-1973	Liquids & Solids	530nm/8ps or 1060nm/8ps	Visible Near IR	10000 cm^{-1}
SPM[4,33]	1974-1976	Fibers	530nm/ns	Visible	500 cm^{-1}
SPM[3]	1983	Glycerol	620nm/100 fs	UV, visible near IR	10000 cm^{-1}
SPM[5]	1985	Semiconductor Dielectrics	10μ/6ps	IR	1000 cm^{-1}
SPM[34]	1986	Gases	600nm/2ps 300nm/0.5ps	Visible UV	5000 cm^{-1}
IPM[11]	1986	Glass	1060nm/530nm 8ps	Visible	1000 cm^{-1}
Harmonic XPM[15,35]	1986	ZnSe	1060nm 8ps	Visible	1000 cm^{-1}
Raman XPM[13,14]	1987	Fibers	503nm 30ps	Visible	1000 cm^{-1}

cess in optical fiber glasses in 1978. In 1983, Shank and his coworkers[3] demonstrated the spectral distribution of SPM in glycerol jet stream using femtosecond laser pulses.

Corkum et al.[5] observed the SPM process spanning from 3-μm to 14-μm using ps CO_2 laser pulses in semiconductors and dielectrics in 1985. Supercontinua in gases using subpicosecond laser pulses in visible and near UV was observed in 1986 by two groups.[34] In coupled modulation, IPM was confirmed in bulk glasses[11] in 1986 and in fiber glasses in 1987.[12] XPM was reported in 1987 by two separate processes.[13–15] One type of XPM is the Raman-XPM in fiber glasses[13,14] and the other type is the Harmonic-XPM in ZnSe.[15] These observations[35] have indicated that phase modulated spectral broadening is a key process for new laser wavelengths, optical information encoding, and possible noise generation in communications.

Explaining the basic mechanism

A simplified model can be used to explain the basic mechanism behind phase modulated spectral broadening within the temporal envelope of an ultrashort laser pulse. When a nearly monochromatic optical pulse propagates in the z-direction in a medium, the electric field amplitude E

. . . phase modulation spectral broadening is a key process for new laser wavelengths, optical information encoding, and possible noise generation in communications.

is given by:

$$E = Re\{E_o(t)\ \exp(i\omega t) - kz)\} \qquad (1)$$

where E_o varies in a period $2\pi/w$. The instantaneous phase of the light wave is given by:

$$\phi(t) = \omega\ t - n(t)\ \omega\ z/c \qquad (2)$$

In an isotropic medium, the index of refraction change due to the third order optical nonlinearity is:

$$\delta n\ (t) = \tfrac{1}{2}\ n_2\ |E_o(t)|^2 \qquad (3)$$

The electric field in ω-space can be evaluated using the method of stationary phase. The frequency chirp Δω can be written as:

$$\Delta\omega(t) = -\omega\,(z/c)\,\partial/\partial t\,(\delta n) \qquad (4)$$

The spectral broadening as a function of the pulse temporal envelope is shown in Fig. 2. All new wavelengths are generated within the original pulse duration. The Stokes pulses are located in the leading edge of the incident pulse, while the anti-Stokes pulses are located in the trailing edge. Assuming δn as a bell shaped curve, t_1 and t_2 its inflection points, and t′ and t″ the solutions of the critical points equation, the maximum frequency broadening of Stokes and anti-Stokes occurs at t_1 and t_2.

Using the Fourier transformation, the spectrum distribution of the pulse is:

$$S(\omega) = c/4\pi\,|E(\omega)|^2 \qquad (5)$$

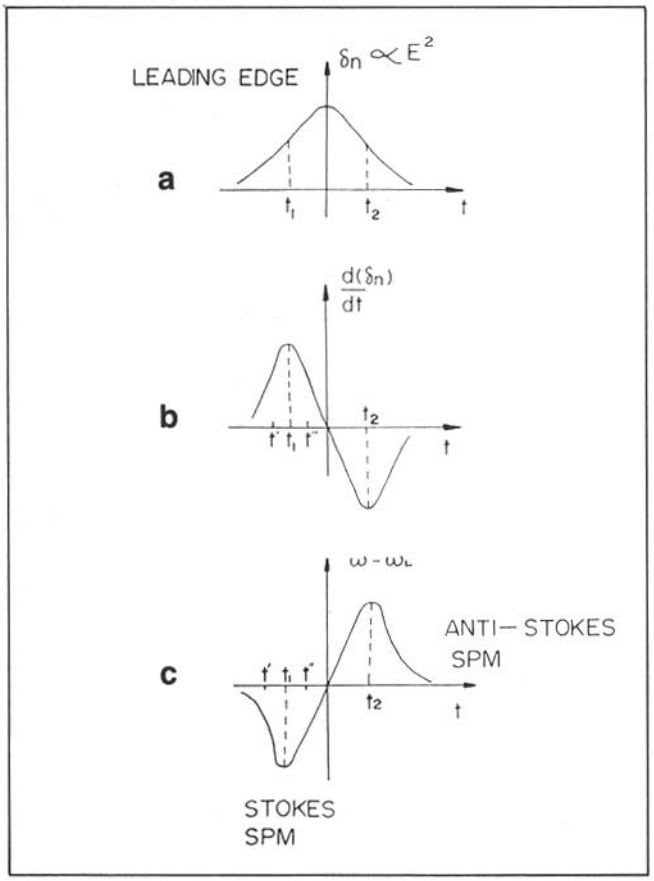

FIGURE 2. Frequency generation with a Gaussian pulse envelope by the SPM process. (a) Refractive index change as a function of time. (b) Time derivative of the refractive index. (c) Frequency broadening as a function of time.

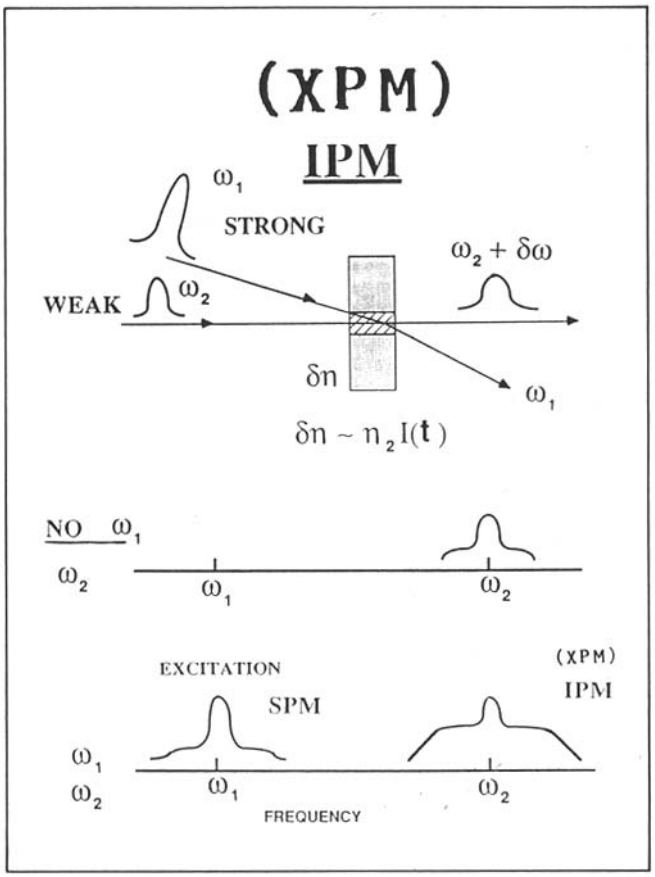

FIGURE 3. Couple-phase-modulation in a nonlinear medium. For IPM, weak ω2-pulse is externally sent into the nonlinear medium. For XPM, ω2-pulse is internally generated in the nonlinear medium through Raman or harmonic generation process.

The maximum frequency extent can be expressed as

$$|\Delta\omega| \sim \omega\, n_2 z E_0^2/[2\pi\,\ell n2)^{0.5}\, c\,\tau \qquad (6)$$

where τ is the pulse duration of the incident laser.

Spectral broadening due to the coupled-phase modulation (IPM and XPM) is sketched in Fig. 3. An intense pulse E_1 at frequency ω_1 is used to dynamically alter the phase and frequency of a weak pulse in different spectral regimes. The induced phase shift, $\delta\phi_2$, of E_2 can be expressed as:

$$\delta\phi_2 = \delta\phi(\text{SPM}) + \delta\phi(\text{IPM or XPM})$$
$$= (\omega_2/c)n_2E_2^2z + 2\,(\omega_2/c)n_2E_1^2z \qquad (7)$$

In a theoretical analysis of coupled phase modulation,[16,17] first order coupled wave equations have been obtained for IPM, Raman-XPM, and harmonic-XPM. In all

three cases, the spectral bandwidth of the weak probe laser is increased as the probe pulses travels in the medium. In Raman-XPM, by assuming the phase matching and neglecting group velocity dispersion and higher-order Raman processes, the calculated ratio[13] of the Raman spectral width to the primary spectral width is $\Delta\omega_{Raman}/\Delta\omega_{Laser} > 2$.. The spectral broadening of the Raman line is larger than the incident laser line broadening. This has been experimentally confirmed.[13]

In the SHG-XPM,[15,35] when an intense laser pulse propagates through noncentrosymmetric media where both χ^2 and χ^3 are operative, coupled spectral broadening around the non-phase matching second harmonic frequency occurs. The calculated temporal distribution of harmonic-XPM has shown a destructive interference pattern when the harmonic is phase mismatched. Spectral broadened SHG-XPM pulses appear to be mainly generated at the entrance and exit surfaces of the crystal. This has been observed[15] by sending an 1054-nm picosecond laser pulse through ZnSe. The output second harmonic spectral around the 527-nm has been broadened to about 2,000-cm^{-1} and all these wavelengths have been observed to be emitted at nearly the same time as the 1054-nm pulse, even though there is a 0.8 difference between their group indices.

In the first and key IPM experiment,[11] the supercontinuum generated by a weak 527-nm picosecond pulse in a BK-7 glass from 410 nm to 660 nm wavelength was increased 11 times when an intense 1054-nm pulse was added. Supercontinua generated by the 1054-nm pulse alone in this spectral region were less than 1% of the total signal. In a most recent IPM experiment,[12] 1.06-μm and 0.53-μm 30-ps laser pulses from a mode-locked YAG laser were passed into an optical fiber. Due to the walk-off between 1064-nm and 530 pulses, the peak wavelength of a weak 530-nm pulse was found to be shifted as a function of an added intense laser pulse in a 1-m long fiber. The magnitude of the induced peak wavelength shift of the

weak pulse was found to be a function of the delay time between these two pulses. IPM can be adapted to encode THz optical information in a multiplex-demultiplex shift register by transferring the time delay variation of pulses into a frequency shift.

Knowledge of the spectral broadening mechanism[36] of the phase modulation processes as well as the capability to control the spectrum and pulse propagation will be useful in future optical communications, signal processing, squeezed state, quantum demolition detection, and optical computation systems.

Acknowledgment

This research is supported in part by Hamamatsu Photonics.

REFERENCES

1. R.R. Alfano and S.L. Shapiro, Phys. Rev. Lett., **24**, 584, 592, 1219 (1970).
2. R.R. Alfano and S.L. Shapiro, Scientific Am., **28**, 42 (1973); R.R. Alfano, L. Hope, and S.L. Shapiro, Phys. Rev. **A6** 433 (1972).
3. R.L. Fork, C.V. Shank, C. Hirlimann, and R. Yen, Opt. Lett., **8**, 1 (1983).
4. R.H. Stolen and C. Lin, Phys. Rev. **A17**, 1448 (1978).
5. P. Corkum, P. Ho, R. Alfano, and J. Manassah, Opt. Lett., **10**, 624 (1985).
6. R. Byer and R. Herbst, in Y. Shen, ed. *Nonlinear infrared generation*, Springer Verlag, Berlin (1977), p. 81.
7. N. Bloembergen, Opt. Comm., **8**, 285 (1973).
8. F. Shimizu, Phys. Rev. Lett., **19**, 1097 (1967).
9. T.K. Gustafson, J.P. Taran, H.A. Haus, J.R. Lifsitz, and P.L. Kelley, Phys. Rev. **177**, 306 (1969).
10. V. Kozobkin, A. Malyntin, A, Prokhrov, JETP Lett., **12**, 150 (1970).
11. R.R. Alfano, Q. Li, T. Jimbo, J. Manassah, and P.P. Ho, Opt. Lett., **11**, 626 (1986).
12. R.R. Alfano, P. Baldeck, and P. Agrawal, Appl. Phys. Lett., **52**, 1939 (1988).
13. R.R. Alfano, P. Baldeck, F. Raccah, and P.P. Ho, Appl. Opt., **6**, 3491 (1987).
14. M. Islam, L. Mollenauer, R. Stolen, J. Simpson, and H. Shang, Opt. Lett., **12**, 625 (1987).
15. R.R. Alfano, Q.Z. Wang, T. Jimbo, P.P. Ho, R. Bhargava, and B. Fitzpatrick, Phys. Rev. A **35**, 459 (1987).
16. J. Manassah and O. Cockings, Opt. Lett., **12**, 1005 (1987).
17. J. Gersten, R. Alfano, and M. Belic, Phy. Rev. **A35**, 1222 (1980).
18. R.R. Alfano, ed. *Biological Events Probed by Ultrafast Laser Spectroscopy*, Academic Press (1982); R.R. Alfano and S. Shapiro, Chem, Phys. Lett., **8**, 631 (1971).
19. D. Hulin, A. Mysyrowicz, A. Antonetti, A. Migus, W. Masselink, H. Morkoc, H. Gibbs, and N. Peyghambarian, Phys. Rev. **B33**, 4389 (1984).
20. M. Berg, A. Harris, J. Brown, and C. Harris, Opt. Lett., **9**, 50 (1984).
21. A. Matveetz Yu, S. Chekalin, and A. Sharkov, JOSA **B2**, 634 (1985).
22. T. Jedju and L. Rothberg, Appl. Opt., **26**, 2877 (1987).
23. L. Goldberg, Picosecond Phenomena III, ed. by K. Eisenthal, R. Hochstrasser, W. Kaiser, and A. Lauberau, p. 94, Springer Verlag, New York (1982).
24. P. Delfyett, R. Dorinsville, and R.R. Alfano, Opt. Lett., **12**, 1002 (1987).
25. E. Treacy, IEEE J. Quant. Elect. **QE-5**, 454 (1969).
26. D. Grischkowsky and A. Balant, Appl. Phys. Lett., **41**, 1 (1982).
27. A. Johnson and M. Simpson, IEEE J. Quant. Electron., **QE-22**, 133 (1986).
28. R. L. Fork, C.H. Brito-Cruz, P.C. Baker, and C.V. Shank, Opt. Lett., **12**, 483 (1987).
29. M. Kitagawa and Y. Yamamato, Phys. Rev. **A34**, 3974 (1986).
30. A. Weiner, J. Heritage, and E. Kirschner, JOSA **B5**, 1563 (1988).
31. R.R. Alfano, J. Gersten, G. Zawadzkas, and N. Tzoar, Phys. Rev. **A10**, 698 (1974).
32. R.R. Alfano, P. Ho, P. Fleury, and H. Guggenheim, Opt. Comm. **19**, 261 (1976).
33. E. Ippen, C. Shank, and T. Gustafson, Appl. Phys. Lett., **33**, 1765 (1974).
34. P.B. Corkum, C. Rolland, and T. Raq, Phys. Rev. Lett., **57**, 2268 (1986). J. Glownia, G. Arjavalingam, P. Sorokin, and J. Rothenberg, Opt. Lett., **11**, 79 (1986).
35. R.R. Alfano and P. Ho, IEEE, J-QE **24**, 351 (1988).
36. R.R. Alfano, ed. *Supercontinuum Laser Source*, Springer Verlag, New York, (1989).