Theory of nonlinear pulse propagation in silicon-nanocrystal waveguides

Ivan D. Rukhlenko
Advanced Computing and Simulation Laboratory (AχL), Department of Electrical and Computer Systems Engineering, Monash University, Clayton, VIC 3800, Australia
ivan.rukhlenko@monash.edu

Abstract: We develop a comprehensive theory of the nonlinear propagation of optical pulses through silica waveguides doped with highly nonlinear silicon nanocrystals. Our theory describes the dynamics of arbitrarily polarized pump and Stokes fields by a system of four generalized nonlinear Schrödinger equations for the slowly varying field amplitudes, coupled to the rate equation for the number density of free carriers. In deriving these equations, we use an analytic expression for the third-order effective susceptibility of the waveguide with randomly oriented nanocrystals, which takes into account both the weakening of the nonlinear optical response of silicon nanocrystals due to their embedment in fused silica and the change in the tensor properties of the response due to the modification of light interaction with electrons and phonons inside the silicon-nanocrystal waveguide. In order to facilitate the use of our theory by experimentalists, and for reasons of methodology, we provide a great deal of detail on the mathematical treatment throughout the paper, even though the derivation of the coupled-amplitude equations is quite straightforward. The developed theory can be applied for the solving of a wide variety of specific problems that require modeling of nonlinear optical phenomena in silicon-nanocrystal waveguides.

OCIS codes: (999.9999) Silicon nanocrystals; (190.0190) Nonlinear optics; (160.4330) Nonlinear optical materials; (190.4400) Nonlinear optics, materials; (260.2065) Effective medium theory.

References and links

1. R. Soref and J. Lorenzo, "All-silicon active and passive guided-wave components for $\lambda = 1.3$ and $1.6\ \mu$m," IEEE J. Quantum Electron. **22**, 873–879 (1986).
2. J. Leuthold, C. Koos, and W. Freude, "Nonlinear silicon photonics," Nat. Photonics **4**, 535–544 (2010).
3. R. M. Osgood, Jr., N. C. Panoiu, J. I. Dadap, X. Liu, X. Chen, I.-W. Hsieh, E. Dulkeith, W. M. Green, and Y. A. Vlasov, "Engineering nonlinearities in nanoscale optical systems: Physics and applications in dispersion-engineered silicon nanophotonic wires," Adv. Opt. Photonics **1**, 162–235 (2009).
4. Q. Lin, O. J. Painter, and G. P. Agrawal, "Nonlinear optical phenomena in silicon waveguides: Modeling and applications," Opt. Express **15**, 16604–16644 (2007).
5. R. A. Soref, "The past, present, and future of silicon photonics," IEEE J. Sel. Top. Quantum Electron. **12**, 1678–1687 (2006).
6. G. T. Reed and A. P. Knights, *Silicon Photonics: An Introduction* (John Wiley, Hoboken, 2004).
7. L. Pavesi and D. Lockwood, eds., *Silicon Photonics*, vol. 94 of *Topics in Applied Physics* (Springer-Verlag, Berlin, 2004).
8. M. Paniccia, "Integrating silicon photonics," Nat. Photonics **4**, 498–499 (2010).
9. J. I. Dadap, N. C. Panoiu, X. Chen, I.-W. Hsieh, X. Liu, C.-Y. Chou, E. Dulkeith, S. J. McNab, F. Xia, W. M. J. Green, L. Sekaric, Y. A. Vlasov, and J. R. M. Osgood, "Nonlinear-optical phase modification in dispersion-engineered Si photonic wires," Opt. Express **16**, 1280–1299 (2008).

#181788 - $15.00 USD Received 14 Dec 2012; revised 15 Jan 2013; accepted 16 Jan 2013; published 29 Jan 2013

10. C. Koos, L. Jacome, C. Poulton, J. Leuthold, and W. Freude, “Nonlinear silicon-on-insulator waveguides for all-optical signal processing,” Opt. Express. **15**, 5976–5990 (2007).
11. X. Chen, N. C. Panoiu, I. Hsieh, J. I. Dadap, and R. M. Osgood, “Third-order dispersion and ultrafast-pulse propagation in silicon wire waveguides,” IEEE Photonics Technol. Lett. **18**, 2617–2619 (2006).
12. A. Martinez, J. Blasco, P. Sanchis, J. V. Galan, J. Garcia-Ruperez, E. Jordana, P. Gautier, Y. Lebour, S. Hernandez, R. Spano, R. Guider, N. Daldosso, B. Garrido, J. M. Fedeli, L. Pavesi, and J. Marti, “Ultrafast all-optical switching in a silicon-nanocrystal-based silicon slot waveguide at telecom wavelengths,” Nano Lett. **10**, 1506–1511 (2010).
13. C. Manolatou and M. Lipson, “All-optical silicon modulators based on carrier injection by two-photon absorption,” IEEE J. Lightwave Technol. **24**, 1433–1439 (2006).
14. R. Jones, A. Liu, H. Rong, M. Paniccia, O. Cohen, and D. Hak, “Lossless optical modulation in a silicon waveguide using stimulated Raman scattering,” Opt. Express **13**, 1716–1723 (2005).
15. D. J. Moss, L. Fu, I. Littler, and B. J. Eggleton, “Ultrafast all-optical modulation via two-photon absorption in silicon-on-insulator waveguides,” Electron. Lett. **41**, 320–321 (2005).
16. I. D. Rukhlenko, M. Premaratne, I. L. Garanovich, A. A. Sukhorukov, and G. P. Agrawal, “Analytical study of pulse amplification in silicon Raman amplifiers,” Opt. Express **18**, 18324–18338 (2010).
17. M. Krause, H. Renner, and E. Brinkmeyer, “Silicon Raman amplifiers with ring-resonator-enhanced pump power,” IEEE J. Sel. Top. Quantum Electron. **16**, 216–225 (2010).
18. I. D. Rukhlenko, C. Dissanayake, M. Premaratne, and G. P. Agrawal, “Maximization of net optical gain in silicon-waveguide Raman amplifiers,” Opt. Express **17**, 5807–5814 (2009).
19. I. D. Rukhlenko, M. Premaratne, C. Dissanayake, and G. P. Agrawal, “Continuous-wave Raman amplification in silicon waveguides: Beyond the undepleted pump approximation,” Opt. Lett. **34**, 536–538 (2009).
20. M. Krause, H. Renner, S. Fathpour, B. Jalali, and E. Brinkmeyer, “Gain enhancement in cladding-pumped silicon Raman amplifiers,” IEEE J. Quantum Electron. **44**, 692–704 (2008).
21. M. A. Foster, A. C. Turner, J. E. Sharping, B. S. Schmidt, M. Lipson, and A. L. Gaeta, “Broad-band optical parametric gain on a silicon photonic chip,” Nature **441**, 960–963 (2006).
22. J. I. Dadap, R. L. Espinola, R. M. Osgood, S. J. McNab, and Y. A. Vlasov, “Spontaneous Raman scattering in ultrasmall silicon waveguides,” Opt. Lett. **29**, 2755–2757 (2004).
23. D. Liang and J. E. Bowers, “Recent progress in lasers on silicon,” Nat. Photonics **4**, 511–517 (2010).
24. H. Rong, S. Xu, O. Cohen, O. Raday, M. Lee, V. Sih, and M. Paniccia, “A cascaded silicon Raman laser,” Nat. Photonics **2**, 170–174 (2008).
25. M. Krause, H. Renner, and E. Brinkmeyer, “Analysis of Raman lasing characteristics in silicon-on-insulator waveguides,” Opt. Express **12**, 5703–5710 (2004).
26. O. Boyraz and B. Jalali, “Demonstration of a silicon Raman laser,” Opt. Express **12**, 5269–5273 (2004).
27. M. W. Geis, S. J. Spector, M. E. Grein, J. U. Yoon, D. M. Lennon, and T. M. Lyszczarz, “Silicon waveguide infrared photodiodes with >35 GHz bandwidth and phototransistors with 50 AW-1 response,” Opt. Express **17**, 5193–5204 (2009).
28. W. Astar, J. B. Driscoll, X. Liu, J. I. Dadap, W. M. J. Green, Y. A. Vlasov, G. M. Carter, and R. M. Osgood, “Conversion of 10 Gb/s NRZ-OOK to RZ-OOK utilizing XPM in a Si nanowire,” Opt. Express **17**, 12987–12999 (2009).
29. I. D. Rukhlenko, M. Premaratne, and G. P. Agrawal, “Analytical study of optical bistability in silicon-waveguide resonators,” Opt. Express **17**, 22124–22137 (2009).
30. H. K. Tsang and Y. Liu, “Nonlinear optical properties of silicon waveguides,” Semicond. Sci. Technol. **23**, 064007 (2008).
31. M. A. Foster, A. C. Turner, M. Lipson, and A. L. Gaeta, “Nonlinear optics in photonic nanowires,” Opt. Express **16**, 1300–1320 (2008).
32. I.-W. Hsieh, X. Chen, X. Liu, J. I. Dadap, N. C. Panoiu, C.-Y. Chou, F. Xia, W. M. Green, Y. A. Vlasov, and R. M. Osgood, “Supercontinuum generation in silicon photonic wires,” Opt. Express **15**, 15242–15249 (2007).
33. R. Espinola, J. Dadap, R. Osgood, Jr., S. McNab, and Y. Vlasov, “C-band wavelength conversion in silicon photonic wire waveguides,” Opt. Express **13**, 4341–4349 (2005).
34. M. W. Geis, S. J. Spector, R. C. Williamson, and T. M. Lyszczarz, “Submicrosecond submilliwatt silicon-on-insulator thermooptic switch,” IEEE Photonics Technol. Lett. **16**, 2514–2516 (2004).
35. M. Dinu, F. Quochi, and H. Garcia, “Third-order nonlinearities in silicon at telecom wavelengths,” Appl. Phys. Lett. **82**, 2954–2956 (2003).
36. I. D. Rukhlenko, W. Zhu, M. Premaratne, and G. P. Agrawal, “Effective third-order susceptibility of silicon-nanocrystal-doped silica,” Opt. Express **20**, 26275–26284 (2012).
37. L. Pavesi and R. Turan, eds., *Silicon Nanocrystals: Fundamentals, Synthesis and Applications* (WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim, 2010).
38. L. Khriachtchev, ed., *Silicon Nanophotonics: Basic Principles, Present Status and Perspectives* (Pan Stanford, Singapore, 2009).
39. V. A. Belyakov, V. A. Burdov, R. Lockwood, and A. Meldrum, “Silicon nanocrystals: Fundamental theory and implications for stimulated emission,” Adv. Opt. Technol. **2008**, 279502 (2008).

#181788 - $15.00 USD Received 14 Dec 2012; revised 15 Jan 2013; accepted 16 Jan 2013; published 29 Jan 2013

40. F. Iori, E. Degoli, R. Magri, I. Marri, G. Cantele, D. Ninno, F. Trani, O. Pulci, and S. Ossicini, "Engineering silicon nanocrystals: Theoretical study of the effect of codoping with boron and phosphorus," Phys. Rev. B **76**, 085302 (2007).
41. I. D. Rukhlenko and M. Premaratne, "Optimization of nonlinear performance of silicon-nanocrystal cylindrical nanowires," IEEE Photonics J. **4**, 952–959 (2012).
42. F. D. Leonardis and V. M. N. Passaro, "Dispersion engineered silicon nanocrystal slot waveguides for soliton ultrafast optical processing," Adv. OptoElectron. **2011**, 751498 (2011).
43. L. Sirleto, M. A. Ferrara, T. Nikitin, S. Novikov, and L. Khriachtchev, "Giant Raman gain in silicon nanocrystals," Nat. Commun. **3**, 1220 (2012).
44. K. Imakita, M. Ito, R. Naruiwa, M. Fujii, and S. Hayashi, "Enhancement of ultrafast nonlinear optical response of silicon nanocrystals by boron-doping," Opt. Lett. **37**, 1877–1879 (2012).
45. R. J. Kashtiban, U. Bangert, I. F. Crowe, M. Halsall, A. J. Harvey, and M. Gass, "Study of erbium-doped silicon nanocrystals in silica," J. Phys.: Conference Series **241**, 012097 (2010).
46. R. J. Walters, G. I. Bourianoff, and H. A. Atwater, "Field-effect electroluminescence in silicon nanocrystals," Nat. Mater. **4**, 143–146 (2005).
47. T. Nikitin, R. Velagapudi, J. Sainio, J. Lahtinen, M. Räsänen, S. Novikov, and L. Khriachtchev, "Optical and structural properties of SiO_x films grown by molecular beam deposition: Effect of the Si concentration and annealing temperature," J. Appl. Phys. **112**, 094316–094316 (2012).
48. J. Wei, J. Price, T. Wang, C. Hessel, and M. C. Downer, "Size-dependent optical properties of Si nanocrystals embedded in amorphous SiO_2 measured by spectroscopic ellipsometry," J. Vac. Sci. Technol. B **29**, 04D112 (2011).
49. T. Nikitin, K. Aitola, S. Novikov, M. Räsänen, R. Velagapudi, J. Sainio, J. Lahtinen, K. Mizohata, T. Ahlgren, and L. Khriachtchev, "Optical and structural properties of silicon-rich silicon oxide films: Comparison of ion implantation and molecular beam deposition methods," Physica Status Solidi (a) **208**, 2176–2181 (2011).
50. L. Ding, T. P. Chen, Y. Liu, C. Y. Ng, and S. Fung, "Optical properties of silicon nanocrystals embedded in a SiO_2 matrix," Phys. Rev. B **72**, 125419 (2005).
51. I. D. Rukhlenko, M. Premaratne, and G. P. Agrawal, "Effective mode area and its optimization in silicon-nanocrystal waveguides," Opt. Lett. **37**, 2295–2297 (2012).
52. F. Trani, D. Ninno, and G. Iadonisi, "Role of local fields in the optical properties of silicon nanocrystals using the tight binding approach," Phys. Rev. B **75**, 033312 (2007).
53. C. M. Dissanayake, I. D. Rukhlenko, M. Premaratne, and G. P. Agrawal, "Raman-mediated nonlinear interactions in silicon waveguides: Copropagating and counterpropagating pulses," IEEE Photonics Technol. Lett. **21**, 1372–1374 (2009).
54. I. D. Rukhlenko, M. Premaratne, and G. P. Agrawal, "Nonlinear propagation in silicon-based plasmonic waveguides from the standpoint of applications," Opt. Express **19**, 206–217 (2011).
55. I. D. Rukhlenko, M. Premaratne, and G. P. Agrawal, "Analytical study of optical bistability in silicon ring resonators," Opt. Lett. **35**, 55–57 (2010).
56. B. A. Daniel and G. P. Agrawal, "Vectorial nonlinear propagation in silicon nanowire waveguides: Polarization effects," J. Opt. Soc. Am. B **27**, 956–965 (2010).
57. I. D. Rukhlenko, C. Dissanayake, M. Premaratne, and G. P. Agrawal, "Optimization of Raman amplification in silicon waveguides with finite facet reflectivities," IEEE J. Sel. Top. Quantum Electron. **16**, 226–233 (2010).
58. I. D. Rukhlenko, I. Udagedara, M. Premaratne, and G. P. Agrawal, "Effect of free carriers on pump-to-signal noise transfer in silicon Raman amplifiers," Opt. Lett. **35**, 2343–2345 (2010).
59. M. D. Turner, T. M. Monro, and S. Afshar V., "A full vectorial model for pulse propagation in emerging waveguides with subwavelength structures part II: Stimulated Raman scattering," Opt. Express **17**, 11565–11581 (2009).
60. L. Yin, J. Zhang, P. M. Fauchet, and G. P. Agrawal, "Optical switching using nonlinear polarization rotation inside silicon waveguides," Opt. Lett. **34**, 476–478 (2009).
61. I. D. Rukhlenko, M. Premaratne, C. Dissanayake, and G. P. Agrawal, "Nonlinear pulse evolution in silicon waveguides: An approximate analytic approach," IEEE J. Lightwave Technol. **27**, 3241–3248 (2009).
62. D. Dimitropoulos, B. Houshmand, R. Claps, and B. Jalali, "Coupled-mode theory of Raman effect in silicon-on-insulator waveguides," Opt. Lett. **28**, 1954–1956 (2003).
63. S. Afshar V. and T. M. Monro, "A full vectorial model for pulse propagation in emerging waveguides with subwavelength structures part I: Kerr nonlinearity," Opt. Express **17**, 2298–2318 (2009).
64. R. W. Boyd, *Nonlinear Optics* (Academic Press, San Diego, 2008), 3rd ed.
65. X. Chen, N. C. Panoiu, and R. M. Osgood, Jr., "Theory of Raman-mediated pulsed amplification in silicon-wire waveguides," IEEE J. Quantum Electron. **42**, 160–170 (2006).
66. S. N. Volkov, J. J. Saarinen, and J. E. Sipe, "Effective medium theory for 2D disordered structures: A comparison to numerical simulations," J. Mod. Opt. **59**, 954–961 (2012).
67. X. C. Zeng, D. J. Bergman, P. M. Hui, and D. Stroud, "Effective-medium theory for weakly nonlinear composites," Phys. Rev. B **38**, 10970–10973 (1988).
68. W. Cai and V. Shalaev, *Optical Metamaterials: Fundamentals and Applications* (Springer, New York, 2010).

#181788 - $15.00 USD Received 14 Dec 2012; revised 15 Jan 2013; accepted 16 Jan 2013; published 29 Jan 2013

69. C. M. Dissanayake, M. Premaratne, I. D. Rukhlenko, and G. P. Agrawal, "FDTD modeling of anisotropic nonlinear optical phenomena in silicon waveguides," Opt. Express **18**, 21427–21448 (2010).
70. F. Trojanek, K. Neudert, K. Zidek, K. Dohnalova, I. Pelant, and P. Maly, "Femtosecond photoluminescence spectroscopy of silicon nanocrystals," Physica Status Solidi (c) **3**, 3873–3876 (2006).
71. R. Spano, N. Daldosso, M. Cazzanelli, L. Ferraioli, L. Tartara, J. Yu, V. Degiorgio, E. Giordana, J. M. Fedeli, and L. Pavesi, "Bound electronic and free carrier nonlinearities in silicon nanocrystals at 1550 nm," Opt. Express **17**, 3941–3950 (2009).
72. M. A. Ferrara, I. Rendina, S. N. Basu, L. D. Negro, and L. Sirleto, "Raman amplifier based on amorphous silicon nanoparticles," Int. J. Photoenergy **2012**, 254946 (2012).
73. M. A. Ferrara, I. Rendina, and L. Sirleto, "Stimulated Raman scattering in quantum dots and nanocomposite silicon based materials," in "Nonlinear Optics," N. Kamanina, ed. (InTech, Rijeka, 2012), pp. 53–70.
74. H. Richter, Z. P. Wang, and L. Ley, "The one phonon Raman spectrum in microcrystalline silicon," Solid State Commun. **39**, 625–629 (1981).
75. I. H. Campbell and P. M. Fauchet, "The effects of microcrystal size and shape on the one phonon Raman spectra of crystalline semiconductors," Solid State Commun. **58**, 739–741 (1986).
76. L. Cao, B. Nabet, and J. E. Spanier, "Enhanced Raman scattering from individual semiconductor nanocones and nanowires," Phys. Rev. Lett. **96**, 157402 (2006).
77. R. Hillenbrand, T. Taubner, and F. Keilmann, "Phonon-enhanced light-matter interaction at the nanometre scale," Nature **418**, 159–162 (2002).
78. G. P. Agrawal, *Nonlinear Fiber Optics* (Academic Press, San Diego, 2007).
79. I. D. Rukhlenko, M. Premaratne, and G. P. Agrawal, "Nonlinear silicon photonics: Analytical tools," IEEE J. Sel. Top. Quantum Electron. **16**, 200–215 (2010).
80. I. D. Rukhlenko, I. L. Garanovich, M. Premaratne, A. A. Sukhorukov, G. P. Agrawal, and Y. S. Kivshar, "Polarization rotation in silicon waveguides: Analytical modeling and applications," IEEE Photonics J. **2**, 423–435 (2010).

1. Introduction

In a lapse of two decades, the concept of silicon photonics proposed by Richard Soref [1] in the mid 1980s has culminated in the realization of high-performance passive and active microscale optical devices [2–7]. One of the main advantages offered by such devices is their low fabrication cost and the possibility to integrate them with standard microelectronics components using the mature complementary semiconductor–metal–oxide (CMOS) technology [8]. Another merit is the tight optical confinement, which significantly enhances the efficiency of all nonlinear optical processes, and the small linear absorption in silicon at infrared wavelengths. Being optically dense and strongly nonlinear material, silicon is ideal for the integration of the five key elements enabling optical information processing: waveguides [9–11], modulators [12–15], amplifiers [16–22], lasers [23–26], and detectors [27]. The functionalities of the nonlinear silicon elements are made possible by the ultrafast Kerr effect and stimulated Raman scattering—stemming from the third-order optical nonlinearity—and the fifth-order free-carrier and thermo-optic effects [28–35].

The optical functionalities realized with bulk silicon can be also achieved with low-dimensional silicon in the form of spherical nanocrystals (NCs) embedded in a SiO_2 matrix [36–40]. Indeed, since the refractive index of the Si-NCs/SiO_2 composite may be anywhere between 1.45 and 2.2 depending on the excess of silicon, this composite can confine and guide light better than the pure silica fibers do [41,42]. More importantly, the nonlinear effects associated with the third-order electron and phonon nonlinearities are significantly enhanced in Si NCs as compared to bulk silicon. Recent measurements have shown that the nonlinear Kerr coefficient and Raman gain of the Si-NCs/SiO_2 composite may exceed those of silicon by factors of 100 and 10000 [37,43,44], respectively, even when the volume fraction of the NCs does not exceed 10%. Because of an indirect bandgap, neither bulk nor low-dimensional silicon can naturally emit light in the telecommunication band. However, by simply coupling Si NCs to Er^{3+} ions doping the SiO_2 matrix, one can realize the desired emission from the NCs and thus create from them a promising material for light emitting devices [39,45,46]. All these features allow us to expect that the Si-NCs/SiO_2 composite will eventually replace the

#181788 - $15.00 USD Received 14 Dec 2012; revised 15 Jan 2013; accepted 16 Jan 2013; published 29 Jan 2013

silicon-on-insulator (SOI) technology in creating the key elements of silicon photonics.

In contrast to a large number of experimental studies of the Si-NCs/SiO_2 composite over the past years [12, 37, 38, 43, 47–50], there have been only a few theoretical works centered at the nonlinear propagation through this material [36, 41, 42, 51, 52]. The imbalance between theory and experiment is especially striking in light of the advanced numerical and analytical models of the nonlinear light–matter interaction developed for silicon [3, 4, 53], and the numerous theoretical studies of the nonlinear optical phenomena in SOI waveguides [54–62]. The reason behind this disproportion is the absence of the unified theoretical platform for modeling nonlinear optical propagation through the Si-NCs/SiO_2 composite. The demand for such a platform is even higher than it was for silicon, as it is extremely computationally challenging to solve Maxwell's equations for a macroscopic sample of the composite with hundreds of thousands of tiny NCs treated individually.

In this paper, we present the first theory of nonlinear pulse propagation through a Si-NC-doped silica waveguide. Our theory assumes arbitrary orientation of the NCs in space and describes this situation using an effective third-order susceptibility of the Si-NCs/SiO_2 composite [36]. The paper is organized as follows. In Section 2, we derive a system of coupled nonlinear equations for the slowly varying amplitudes of the pump and Stokes pulses inside a nonlinear waveguide. The system shows that the source of the space and time evolution of the amplitudes is the waveguide's nonlinear material polarization. Section 3 is therefore devoted to the calculation of the electronic, Raman, and free-carrier contributions to the nonlinear polarization induced by the pump and Stokes fields inside the Si-NCs/SiO_2 composite. The free-carrier contribution depends on the generation rate of electron–hole pairs inside the NCs due to the effect of two-photon absorption, whose efficiency is governed by field intensity and thus depends on the effective mode area (EMA) of the waveguide. The ambiguity in defining EMA is emphasized in Section 4, where we provide two alternative definitions of EMA and explain the often ignored difference between them. The results of Sections 2–4 are merged in Section 5 to give the desired coupled amplitude equations governing the nonlinear propagation of the two pulses. These equations are then simplified for the case of continuous waves, before Section 6 summarizes our results and concludes the paper.

2. Nonlinear propagation equations

The general equations governing propagation of optical field through a nonlinear waveguide may be derived by treating the nonlinear material polarization as a small perturbation coupling otherwise independent propagating modes of the same waveguide operating in the linear regime. Let us consider the propagation of narrowband pump ($\mu = p$) and Stokes ($\mu = s$) pulses, whose spectra are centered around frequencies ω_p and ω_s. In this case, the unperturbed propagating modes of the linear waveguide are the solutions to Maxwell's equations for the pump and Stokes fields

$$\nabla \times \widetilde{\mathbf{E}}_\mu^{(0)}(\mathbf{r}, \omega) = i\omega\mu_0 \widetilde{\mathbf{H}}_\mu^{(0)}(\mathbf{r}, \omega) \tag{1a}$$

and

$$\nabla \times \widetilde{\mathbf{H}}_\mu^{(0)}(\mathbf{r}, \omega) = -i\omega\varepsilon_0\varepsilon_L(\mathbf{r}_\perp, \omega)\widetilde{\mathbf{E}}_\mu^{(0)}(\mathbf{r}, \omega), \tag{1b}$$

where $\varepsilon_L(\mathbf{r}_\perp, \omega)$ is the transverse profile of linear permittivity and $\mathbf{r}_\perp$ is the two-dimensional radius vector perpendicular to the waveguide axis z.

The solution to Maxwell's equations for the perturbed fields can be expanded over the complete set of propagating modes $\mathbf{e}_{\mu\nu}(\mathbf{r}_\perp, \omega_\mu)\, e^{i\beta_{\mu\nu}z}$ and $\mathbf{h}_{\mu\nu}(\mathbf{r}_\perp, \omega_\mu)\, e^{i\beta_{\mu\nu}z}$ satisfying Eq. (1) as

#181788 - $15.00 USD Received 14 Dec 2012; revised 15 Jan 2013; accepted 16 Jan 2013; published 29 Jan 2013

$$\widetilde{\mathbf{E}}_\mu(\mathbf{r},\omega) = \sum_\nu \widetilde{a}_{\mu\nu}(z,\omega-\omega_\mu)\frac{\mathbf{e}_{\mu\nu}(\mathbf{r}_\perp,\omega_\mu)}{\sqrt{N_{\mu\nu}}}\,e^{i\beta_{\mu\nu}z} \tag{2a}$$

and

$$\widetilde{\mathbf{H}}_\mu(\mathbf{r},\omega) = \sum_\nu \widetilde{a}_{\mu\nu}(z,\omega-\omega_\mu)\frac{\mathbf{h}_{\mu\nu}(\mathbf{r}_\perp,\omega_\mu)}{\sqrt{N_{\mu\nu}}}\,e^{i\beta_{\mu\nu}z}, \tag{2b}$$

where $\beta_{\mu\nu} \equiv \beta_\nu(\omega_\mu)$ is the real propagation constant of mode ν evaluated at the carrier frequency ω_μ of field μ and we assume the modal profiles $\mathbf{e}_{\mu\nu}$ and $\mathbf{h}_{\mu\nu}$ to be normalized according to the condition

$$\iint (\mathbf{e}^*_{\mu\nu}\times\mathbf{h}_{\mu\nu'}+\text{c.c.})\,\mathrm{d}\mathbf{r}_\perp = \delta_{\nu\nu'}\iint (\mathbf{e}^*_{\mu\nu}\times\mathbf{h}_{\mu\nu}+\text{c.c.})\,\mathrm{d}\mathbf{r}_\perp = 4N_{\mu\nu}, \tag{3}$$

where the integration is taken over the entire area of the xy plane and the constants $N_{\mu\nu}$ are implicitly defined. With this normalization, one finds that the total power carried by field μ is given by

$$P_\mu = \frac{1}{2}\iint \mathrm{Re}\big(\widetilde{\mathbf{E}}_\mu\times\widetilde{\mathbf{H}}^*_\mu\big)\,\mathrm{d}\mathbf{r}_\perp = \sum_\nu |\widetilde{a}_{\mu\nu}|^2. \tag{4}$$

As in Eq. (3), the double integral here is evaluated over the entire transverse plane.

If $\widetilde{\mathbf{P}}^{\mathrm{NL}}_\mu(\mathbf{r},\omega)$ is the sum of material polarizations stemming from all types of nonlinearities in the waveguide, then the perturbed fields $\widetilde{\mathbf{E}}_\mu$ and $\widetilde{\mathbf{H}}_\mu$ must obey the equations

$$\nabla\times\widetilde{\mathbf{E}}_\mu(\mathbf{r},\omega) = i\omega\mu_0\widetilde{\mathbf{H}}_\mu(\mathbf{r},\omega) \tag{5a}$$

and

$$\nabla\times\widetilde{\mathbf{H}}_\mu(\mathbf{r},\omega) = -i\omega\varepsilon_0\varepsilon_L(\mathbf{r}_\perp,\omega)\widetilde{\mathbf{E}}_\mu(\mathbf{r},\omega) - i\omega\widetilde{\mathbf{P}}^{\mathrm{NL}}_\mu(\mathbf{r},\omega). \tag{5b}$$

The perturbed and unperturbed fields are also interrelated through the reciprocity theorem [3, 59, 63]

$$\frac{\partial}{\partial z}\iint\left(\widetilde{\mathbf{E}}^{(0)}_\mu\times\widetilde{\mathbf{H}}^*_\mu+\widetilde{\mathbf{E}}^*_\mu\times\widetilde{\mathbf{H}}^{(0)}_\mu\right)\mathrm{d}\mathbf{r}_\perp = \iint\nabla\left(\widetilde{\mathbf{E}}^{(0)}_\mu\times\widetilde{\mathbf{H}}^*_\mu+\widetilde{\mathbf{E}}^*_\mu\times\widetilde{\mathbf{H}}^{(0)}_\mu\right)\mathrm{d}\mathbf{r}_\perp. \tag{6}$$

Using the vector identity $\nabla\cdot(\mathbf{f}\times\mathbf{g}) = \mathbf{g}\cdot(\nabla\times\mathbf{f})-\mathbf{f}\cdot(\nabla\times\mathbf{g})$, together with Eqs. (1) and (5), it is possible to express the gradient in Eq. (6) by means of the scalar product

$$\nabla\left(\widetilde{\mathbf{E}}^{(0)}_\mu\times\widetilde{\mathbf{H}}^*_\mu+\widetilde{\mathbf{E}}^*_\mu\times\widetilde{\mathbf{H}}^{(0)}_\mu\right) = -i\omega\left(\widetilde{\mathbf{E}}^{(0)}_\mu\cdot\widetilde{\mathbf{P}}^{NL*}_\mu\right). \tag{7}$$

By introducing this expression into Eq. (6), substituting in the resulting relation both the unperturbed solutions

$$\widetilde{\mathbf{E}}^{(0)}_\mu = \frac{\mathbf{e}_{\mu\nu}(\mathbf{r}_\perp,\omega_\mu)}{\sqrt{N_{\mu\nu}}}\,e^{i\beta_\nu(\omega)z}\quad\text{and}\quad\widetilde{\mathbf{H}}^{(0)}_\mu = \frac{\mathbf{h}_{\mu\nu}(\mathbf{r}_\perp,\omega_\mu)}{\sqrt{N_{\mu\nu}}}\,e^{i\beta_\nu(\omega)z} \tag{8}$$

and the perturbed ones given in Eq. (2), we find using Eq. (3) that

$$\frac{\partial}{\partial z}\left\{\widetilde{a}_{\mu\nu}(z,\omega-\omega_\mu)\,e^{i[\beta_\nu(\omega_\mu)-\beta_\nu(\omega)]z}\right\} = \frac{i\omega}{4\sqrt{N_{\mu\nu}}}\iint\mathbf{e}^*_{\mu\nu}(\mathbf{r}_\perp,\omega_\mu)\cdot\widetilde{\mathbf{P}}^{\mathrm{NL}}_\mu(\mathbf{r},\omega)\,e^{-i\beta_\nu(\omega)z}\,\mathrm{d}\mathbf{r}_\perp. \tag{9}$$

#181788 - $15.00 USD Received 14 Dec 2012; revised 15 Jan 2013; accepted 16 Jan 2013; published 29 Jan 2013

The evaluation of the derivative in this equation gives

$$\left(\frac{\partial}{\partial z}+i\sum_{n=1}^{\infty}\frac{1}{n!}\frac{\partial\beta_{\nu}}{\partial\omega}\bigg|_{\omega_{\mu}}(\omega-\omega_{\mu})^{n}\right)\widetilde{a}_{\mu\nu}(z,\omega-\omega_{\mu})$$
$$=\frac{i\omega}{4\sqrt{N_{\mu\nu}}}\iint\mathbf{e}^{*}_{\mu\nu}(\mathbf{r}_{\perp},\omega_{\mu})\cdot\widetilde{\mathbf{P}}^{\mathrm{NL}}_{\mu}(\mathbf{r},\omega)\,e^{-i\beta_{\nu}(\omega_{\mu})z}\,\mathrm{d}\mathbf{r}_{\perp}.\quad(10)$$

By multiplying both sides of Eq. (10) by $e^{-i(\omega-\omega_{\mu})t}$ and integrating with respect to ω, we arrive at the following time-domain equation:

$$\left(\frac{\partial}{\partial z}+\sum_{n=1}^{\infty}\frac{i^{n+1}}{n!}\frac{\partial\beta_{\nu}}{\partial\omega}\bigg|_{\omega_{\mu}}\frac{\partial^{n}}{\partial t^{n}}\right)a_{\mu\nu}(z,t)$$
$$=-\frac{e^{-i\beta_{\mu\nu}z}}{4\sqrt{N_{\mu\nu}}}\iint\mathbf{e}^{*}_{\mu\nu}(\mathbf{r}_{\perp},\omega_{\mu})\,\frac{\partial\mathbf{P}^{\mathrm{NL}}_{\mu}(\mathbf{r},t)}{\partial t}\,e^{i\omega_{\mu}t}\,\mathrm{d}\mathbf{r}_{\perp},\quad(11)$$

in which we have set

$$a_{\mu\nu}(z,t)=\frac{1}{2\pi}\int_{-\infty}^{+\infty}\widetilde{a}_{\mu\nu}(z,\omega)\,e^{-i\omega t}\mathrm{d}\omega\quad\text{and}\quad\mathbf{P}^{\mathrm{NL}}_{\mu}(\mathbf{r},t)=\frac{1}{2\pi}\int_{-\infty}^{+\infty}\widetilde{\mathbf{P}}^{\mathrm{NL}}_{\mu}(\mathbf{r},\omega)\,e^{-i\omega t}\mathrm{d}\omega.\quad(12)$$

If we now represent the polarization vector as a product of the slowly varying amplitude and the rapidly oscillating exponential, $\mathbf{P}^{\mathrm{NL}}_{\mu}(\mathbf{r},t)=\mathbf{P}^{\mathrm{NL}}_{\omega_{\mu}}(\mathbf{r},t)\,e^{-i\omega_{\mu}t}$, then Eq. (11) acquires the form

$$\frac{\partial a_{\mu\nu}}{\partial z}+\sum_{n=1}^{\infty}\frac{i^{n+1}\beta^{(n)}_{\mu\nu}}{n!}\frac{\partial^{n}a_{\mu\nu}}{\partial t^{n}}=\frac{i\omega_{\mu}}{4\sqrt{N_{\mu\nu}}}\left(1+\frac{i}{\omega_{\mu}}\frac{\partial}{\partial t}\right)e^{-i\beta_{\mu\nu}z}\iint\left(\mathbf{e}^{*}_{\mu\nu}\cdot\mathbf{P}^{\mathrm{NL}}_{\omega_{\mu}}\right)\mathrm{d}\mathbf{r}_{\perp},\quad(13)$$

where $\beta^{(n)}_{\mu\nu}=\partial\beta_{\nu}/\partial\omega|_{\omega=\omega_{\mu}}$ is the nth-order dispersion parameter at the frequency ω_{μ}. The term with the time derivative on the right-hand side of this equation accounts for the effect of self steepening, which is significant for ultrashort optical pulses [64].

Equation (13) shows that the variation of the amplitudes $a_{\mu\nu}$ of pump and Stokes fields is due to the nonlinear material polarization of Si-NCs/SiO_2 waveguide. This equation is similar to those derived in Refs. [59, 63, 65].

In this paper, we shall calculate the nonlinear polarization of the Si-NCs/SiO_2 waveguide under the frame of the effective medium theory [47, 66, 67]. According to this theory, the effective permittivity of the Si-NCs/SiO_2 composite is related to the permittivity ε_1 of silicon and the permittivity ε_2 of silica as [68] $\varepsilon_{\mathrm{eff}}=(1/4)\left[u+(u^2+8\varepsilon_1\varepsilon_2)^{1/2}\right]$, where $u=(3f-1)\varepsilon_1+(2-3f)\varepsilon_2$ and f is the volume fraction of Si NCs in the composite. The dispersion of $\varepsilon_{\mathrm{eff}}$ needs to be taken into account when one calculates the parameters $\beta^{(n)}_{\mu\nu}$, but may generally be ignored in the cases where $\varepsilon_{\mathrm{eff}}$ explicitly appears in the coupled amplitude equations. Specifically, for narrowband pump and Stokes pulses in the 1.55-μm spectral region, the effective permittivity of the Si-NCs/SiO_2 composite may be calculated by taking $\varepsilon_1\approx 12.1$ and $\varepsilon_2\approx 2.1$. An allowance for the size dependency of the NCs' permittivity can be made using ε_1 extracted from the spectroscopic ellipsometry data presented in Ref. [48].

3. Nonlinear polarization of Si-NCs/SiO_2 waveguide

The nonlinear material polarization of the Si-NCs/SiO_2 waveguide may originate from the nonlinearities of both silicon crystallites and silica matrix. Since the nonlinear effects in Si NCs are

#181788 - $15.00 USD Received 14 Dec 2012; revised 15 Jan 2013; accepted 16 Jan 2013; published 29 Jan 2013

typically much stronger than those in SiO_2, one may safely neglect the third-order susceptibility of silica, provided the volume fraction of the NCs is larger than 0.1% [36]. With this simplification, the total material polarization is a sum of three terms [3, 4, 53, 69]

$$\mathbf{P}^{\mathrm{NL}}_{\omega_\mu}(\mathbf{r},t) = \mathbf{P}^{\mathrm{K}}_{\omega_\mu}(\mathbf{r},t) + \mathbf{P}^{\mathrm{R}}_{\omega_\mu}(\mathbf{r},t) + \mathbf{P}^{\mathrm{FC}}_{\omega_\mu}(\mathbf{r},t), \tag{14}$$

which represent the electronic (Kerr), Raman, and free-carrier contributions.

The electronic polarization stems from the nonlinear interaction of an optical field with electronic clouds of silicon atoms and leads to an almost instantaneous (with a response time less than 100 fs) change in the refractive index of Si NCs (Kerr effect), as well as to the two-photon absorption (TPA) [12]. The Raman polarization allows for the interaction of light with optical phonons and is responsible for the stimulated Raman scattering (SRS) and lasing [17–19, 26]. Free electrons and holes generated inside the NCs *via* TPA also change the refractive index of the NCs, which is usually referred to as the effect of free-carrier dispersion (FCD), and result in the free-carrier absorption (FCA) of the pump and Stokes fields [19, 29]. The effects of FCD and FCA are included in the last polarization term in Eq. (14).

Consider the three types of polarization contributions separately, while assuming that the mean NC size is large enough for the quantum confinement to not increase the bandgap of silicon significantly.

3.1. Electronic contribution

The Kerr polarization induced in the Si-NCs/SiO_2 waveguide with randomly oriented nanocrystals is given by the tensor product

$$\mathbf{P}^{\mathrm{K}}_{\omega_\mu}(\mathbf{r},t) = \varepsilon_0 \chi^{(3)}_{\mathrm{K}}(\omega_\mu;\omega_\mu,-\omega_\mu,\omega_\mu)\,\mathbf{E}_{\omega_\mu}(\mathbf{r},t)\mathbf{E}^*_{\omega_\mu}(\mathbf{r},t)\mathbf{E}_{\omega_\mu}(\mathbf{r},t), \tag{15}$$

where $\mathbf{E}_{\omega_\mu}(\mathbf{r},t)$ is the slowly varying amplitude of the electric field, which may be calculated by Fourier-transforming Eq. (2), and the third-order susceptibility tensor is of the form [36]

$$\chi^{(3)}_{\mathrm{K}}(\omega_\mu;\omega_\mu,-\omega_\mu,\omega_\mu) = \chi_\mu \left(\frac{8+7\rho}{45}\left(\delta_{kl}\delta_{mn} + \delta_{km}\delta_{ln} + \delta_{kn}\delta_{lm}\right) + \frac{1-\rho}{9}\,\delta_{kl}\delta_{lm}\delta_{mn} \right), \tag{16}$$

where $\{k,l,m\} = \{x,y,z\}$, δ_{ij} is the Kronecker delta, and ρ is the nonlinear anisotropy factor ($\rho \approx 1.27$ near the 1.55 μm wavelength). Owing to a uniform distribution of Si-NC orientations in space, the Kerr tensor of the Si-NCs/SiO_2 composite is no longer described with respect to the crystallographic basis, but rather is given in the reference frame associated with the nonlinear waveguide.

The strength of the electronic nonlinearity is determined by the complex parameter [4, 56, 69]

$$\chi_\mu = c\varepsilon_0\varepsilon_{\mathrm{eff}}\left[n_2 + i\beta_{\mathrm{TPA}}/(2k_\mu)\right]\xi, \tag{17}$$

where n_2 and β_{TPA} are the nonlinear Kerr parameter and TPA coefficient of a Si NC, $k_\mu = \omega_\mu/c$, and

$$\xi = \frac{1}{f}\left(\frac{\partial\varepsilon_{\mathrm{eff}}}{\partial\varepsilon_1}\right)^2 = \frac{\left[(3f-1)\,\varepsilon_{\mathrm{eff}} + \varepsilon_2\right]^2}{f(u^2 + 8\varepsilon_1\varepsilon_2)} \tag{18}$$

is the third-order susceptibility attenuation factor [36].

Both the Kerr parameter and the TPA coefficient at 1.55 μm are enhanced in Si NCs when compared to bulk silicon. The Kerr parameter may be enhanced in the Si-NCs/SiO_2 composite by as much as two orders of magnitude and lie in the range $(2\pm1)\times10^{-12}$ cm^2/W [37, 38, 44], while the reported spread of the TPA coefficient is larger and ranges 5 to 170 cm/GW [37, 42, 44, 70]. The values of $\beta_{\rm TPA}$ and n_2 depend on the doping of Si NCs [44] and may vary with powers and repetition rates of optical pulses, due to the band-filling-induced saturation of the nonlinear absorption [71]. In borrowing these parameters from the experimental papers, it is important to distinguish between their values related to a single Si NC and the entire composite. For example, Spano *et al.* [71] found the intensity-dependent TPA coefficient of the Si-NCs/SiO$_2$ composite with $f = 8\%$ to be given by the expression $\beta^{(c)}_{\rm TPA} = \beta^{(0)}_{\rm TPA}/[1+(I/I_s)^2]$, where $\beta^{(0)}_{\rm TPA} = (7.0\pm 0.6)$ cm/GW and $I_s = 4070$ GW/cm^2. The TPA coefficient of a single NC in this case may be estimated as $\beta_{\rm TPA} = \beta^{(c)}_{\rm TPA}/\xi$.

The electronic contribution to the nonlinear material polarization can be written entirely in vector form as

$$\mathbf{P}^{\rm K}_{\omega_\mu} = \varepsilon_0 \chi_\mu \left[\frac{8+7\rho}{45} \left(2|\mathbf{E}_{\omega_\mu}|^2 \mathbf{E}_{\omega_\mu} + \mathbf{E}^2_{\omega_\mu} \mathbf{E}^*_{\omega_\mu} \right) + \frac{1-\rho}{9} \sum_\eta E^2_\eta E^*_\eta \hat{\eta} \right], \tag{19}$$

where E_η is the ηth component of the vector $\mathbf{E}_{\omega_\mu}$ and $\hat{\eta}$ is the unit vector in the direction of the ηth Cartesian axis.

In the rest of the paper, we shall restrict ourselves to the special case of only two unperturbed transverse modes ($\mathbf{e}_{\mu x}$ and $\mathbf{e}_{\mu y}$) in the expansion of Eq. (2), assuming them to be polarized along the x and y axes. In this case, the orthogonality relations $(\mathbf{e}_{\mu\nu} \cdot \mathbf{e}_{\mu\nu'}) = \mathbf{e}^2_{\mu\nu} \delta_{\nu\nu'}$ and $(\mathbf{e}_{\mu\nu} \cdot \mathbf{e}^*_{\mu\nu'}) = |\mathbf{e}_{\mu\nu}|^2 \delta_{\nu\nu'}$, the expansion

$$\mathbf{E}_{\omega_\mu} = \sum_{\nu=x,y} a_{\mu\nu} \frac{\mathbf{e}_{\mu\nu}}{\sqrt{N_{\mu\nu}}} e^{i\beta_{\mu\nu} z}, \tag{20}$$

and Eq. (19) enable one to obtain

$$\begin{aligned} &\frac{e^{-i\beta_{\mu\nu} z}}{\sqrt{N_{\mu\nu}}} \iint \left(\mathbf{e}^*_{\mu\nu} \cdot \mathbf{P}^{\rm K}_{\omega_\mu} \right) {\rm d}\mathbf{r}_\perp \\ &\quad = \varepsilon_0 \chi_\mu \left[\frac{8+7\rho}{45} \left(2a_{\mu\nu} \sum_{\nu'} \Gamma^{(\mu)}_{\nu\nu'} |a_{\mu\nu'}|^2 + a^*_{\mu\nu} \sum_{\nu'} a^2_{\mu\nu'} \Lambda^{(\mu)}_{\nu\nu'} e^{2i(\beta_{\mu\nu'} - \beta_{\mu\nu})z} \right) \right. \\ &\qquad \left. + \frac{1-\rho}{9} \Gamma^{(\mu)}_{\nu\nu} a_{\mu\nu} |a_{\mu\nu}|^2 \right], \end{aligned} \tag{21}$$

where

$$\Gamma^{(\mu)}_{\nu\nu'} = \frac{1}{N_{\mu\nu} N_{\mu\nu'}} \iint |\mathbf{e}_{\mu\nu}|^2 |\mathbf{e}_{\mu\nu'}|^2 \, {\rm d}\mathbf{r}_\perp \tag{22a}$$

and

$$\Lambda^{(\mu)}_{\nu\nu'} = \frac{1}{N_{\mu\nu} N_{\mu\nu'}} \iint \mathbf{e}^{*2}_{\mu\nu} \mathbf{e}^2_{\mu\nu'} \, {\rm d}\mathbf{r}_\perp. \tag{22b}$$

It is easy to prove using Maxwell's equations that $(\mathbf{h}_{\mu\nu})_\perp = (\beta_{\mu\nu}/\mu_0 \omega_\mu) \hat{z} \times \mathbf{e}_{\mu\nu}$ for the transverse electric modes, so that the normalization condition yields

$$N_{\mu\nu} = \frac{\beta_{\mu\nu}}{2\mu_0 \omega_\mu} \iint |\mathbf{e}_{\mu\nu}|^2 \, {\rm d}\mathbf{r}_\perp. \tag{23}$$

Received 14 Dec 2012; revised 15 Jan 2013; accepted 16 Jan 2013; published 29 Jan 2013
11 February 2013 / Vol. 21, No. 3 / OPTICS EXPRESS 2840

3.2. *Raman contribution*

The Raman polarization of the Si-NCs/SiO_2 waveguide with randomly oriented nanocrystals can be represented as

$$\begin{aligned}\mathbf{P}^{\mathrm{R}}_{\omega_\mu}(\mathbf{r},t) = e^{i\omega_\mu t}\int_{-\infty}^{t}\mathrm{d}t_1\int_{-\infty}^{t}\mathrm{d}t_2\int_{-\infty}^{t}\mathrm{d}t_3\,\varepsilon_0\chi^{(3)}_{\mathrm{R}}(t-t_1,t-t_2,t-t_3)\\ \times\Big(\mathbf{E}_{\omega_{\mu'}}(\mathbf{r},t_1)\mathbf{E}^*_{\omega_{\mu'}}(\mathbf{r},t_2)\mathbf{E}_{\omega_\mu}(\mathbf{r},t_3)\,e^{-i(\omega_{\mu'}t_1-\omega_{\mu'}t_2+\omega_\mu t_3)}\\ +\mathbf{E}_{\omega_\mu}(\mathbf{r},t_1)\mathbf{E}^*_{\omega_{\mu'}}(\mathbf{r},t_2)\mathbf{E}_{\omega_{\mu'}}(\mathbf{r},t_3)\,e^{-i(\omega_\mu t_1-\omega_{\mu'}t_2+\omega_{\mu'}t_3)}\Big),\end{aligned} \quad (24)$$

where $\{\mu,\mu'\}=\{p,s\}$, $\mu\neq\mu'$, and the effective susceptibility tensor is given by the expressions [3,4,36,69]

$$\chi^{(3)}_{\mathrm{R}}(t_1,t_2,t_3) = \frac{1}{2}\left[\delta(t_1-t_2)\delta(t_3)\mathscr{R}_{klmn}+\delta(t_1)\delta(t_2-t_3)\mathscr{R}_{knml}\right]\xi H(t_2), \quad (25a)$$

$$\mathscr{R}_{klmn} = \frac{29}{45}\left(\delta_{km}\delta_{ln}+\delta_{kn}\delta_{lm}\right)-\frac{16}{45}\,\delta_{kl}\delta_{mn}-\frac{2}{9}\,\delta_{kl}\delta_{lm}\delta_{mn}, \quad (25b)$$

and

$$H(t) = 2\chi_R\,\frac{\Gamma_R\Omega_R}{\left(\Omega_R^2-\Gamma_R^2\right)^{1/2}}\,e^{-t/\tau_2}\sin(t/\tau_1), \quad (25c)$$

where χ_R is the peak value of the Raman susceptibility of an individual Si NC, $2\Gamma_R = 2/\tau_2$ is the amplification bandwidth, and $1/\tau_1 = (\Omega_R^2-\Gamma_R^2)^{1/2}\approx\Omega_R$ is the Stokes shift.

Recent experiments on SRS in the Si-NCs/SiO_2 composite have shown [43] that the Raman gain of the composite may be 10 000 times larger than that of bulk silicon ($\sim 10^{-17}$ m^2/V^2) [3]. Since the third-order susceptibility of Si NCs is attenuated by the factor ξ in the composite, this gives us an estimate $\xi\chi_R\sim 10^{-13}$ m^2/V^2. The peak position and bandwidth of gain spectrum in Si NCs also change significantly with respect to their values $2\Gamma_R\approx 105$ GHz and $\Omega_R =$ 15.6 THz in bulk silicon. In particular, for spherical NCs of 2 nm in diameter, the gain peak broadens up to 2 THz and red shifts by about 0.6 THz [72,73]. The downshift and broadening of the Raman spectrum with the change in size of Si NCs can be calculated numerically using the phenomenological theory of Richter *et al.* [74], developed by Campbell and Fauchet [73,75].

The origin of giant Raman gain in Si NCs is currently unclear and warrants further investigation. One possible reason behind this effect may be the resonant field enhancement inside the nanocrystals caused by the interaction of light with optical phonons and the formation of the localized phonon-polariton modes [76,77]. The enhancement occurs within the *reststrahlen* band of phonon-polariton dispersion and is analogous to the plasmon-induced field enhancement in the vicinity of metallic nanoparticles.

By introducing Eq. (25a) into Eq. (24) and evaluating the integrals, we find that

$$\mathbf{P}^{\mathrm{R}}_{\omega_\mu}(\mathbf{r},t) = \varepsilon_0\xi\int_{-\infty}^{t}\mathrm{d}t_1 H(t-t_1)\mathscr{R}_{klmn}\mathbf{E}_{\omega_\mu}(\mathbf{r},t_1)\mathbf{E}^*_{\omega_{\mu'}}(\mathbf{r},t_1)\mathbf{E}_{\omega_{\mu'}}(\mathbf{r},t)\,e^{i(\omega_\mu-\omega_{\mu'})(t-t_1)}. \quad (26)$$

In deriving this expression, we have omitted the nonresonant terms in Eq. (24) and taken into account that the remaining two terms are equal. Now using Eqs. (20) and (25b), we get

$$\frac{e^{-i\beta_{\mu\nu}z}}{\varepsilon_0\xi\sqrt{N_{\mu\nu}}}\iint\left(\mathbf{e}^*_{\mu\nu}\cdot\mathbf{P}^{\mathrm{R}}_{\omega_\mu}\right)\mathrm{d}\mathbf{r}_\perp$$
$$=\frac{29}{45}\sum_{\nu'}\Lambda^{\mu\mu'}_{\nu\nu'}\exp\left(i\beta^{\mu\nu'\mu'\nu'}_{\mu\nu\mu'\nu}z\right)a_{\mu'\nu'}(t)\int_{-\infty}^{t}a^*_{\mu'\nu}(t_1)a_{\mu\nu'}(t_1)H(t-t_1)\,e^{i\omega_{\mu\mu'}(t-t_1)}\mathrm{d}t_1$$
$$+\frac{29}{45}\sum_{\nu'}\Psi^{\mu\mu'}_{\nu\nu'}\exp\left(i\beta^{\mu\nu'\mu'\nu}_{\mu\nu\mu'\nu'}z\right)a_{\mu'\nu}(t)\int_{-\infty}^{t}a^*_{\mu'\nu'}(t_1)a_{\mu\nu'}(t_1)H(t-t_1)\,e^{i\omega_{\mu\mu'}(t-t_1)}\mathrm{d}t_1$$
$$-\frac{16}{45}\sum_{\nu'}\Gamma^{\mu\mu'}_{\nu\nu'}a_{\mu'\nu'}(t)\int_{-\infty}^{t}a^*_{\mu'\nu'}(t_1)a_{\mu\nu}(t_1)H(t-t_1)\,e^{i\omega_{\mu\mu'}(t-t_1)}\mathrm{d}t_1$$
$$-\frac{2}{9}\Lambda^{\mu\mu'}_{\nu\nu}a_{\mu'\nu}(t)\int_{-\infty}^{t}a^*_{\mu'\nu}(t_1)a_{\mu\nu}(t_1)H(t-t_1)\,e^{i\omega_{\mu\mu'}(t-t_1)}\mathrm{d}t_1, \quad (27)$$

where $\mu\neq\mu'$, $\beta^{\mu\nu'\mu'\nu'}_{\mu\nu\mu'\nu}=\beta_{\mu\nu'}+\beta_{\mu'\nu'}-\beta_{\mu\nu}-\beta_{\mu'\nu}$, $\omega_{\mu\mu'}=\omega_\mu-\omega_{\mu'}$,

$$\Lambda^{\mu\mu'}_{\nu\nu'}=\iint\frac{(\mathbf{e}^*_{\mu\nu}\mathbf{e}^*_{\mu'\nu})(\mathbf{e}_{\mu\nu'}\mathbf{e}_{\mu'\nu'})}{\sqrt{N_{\mu\nu}N_{\mu'\nu}N_{\mu\nu'}N_{\mu'\nu'}}}\,\mathrm{d}\mathbf{r}_\perp, \quad (28a)$$

$$\Psi^{\mu\mu'}_{\nu\nu'}=\iint\frac{(\mathbf{e}^*_{\mu\nu}\mathbf{e}_{\mu'\nu})(\mathbf{e}_{\mu\nu'}\mathbf{e}^*_{\mu'\nu'})}{\sqrt{N_{\mu\nu}N_{\mu'\nu}N_{\mu\nu'}N_{\mu'\nu'}}}\,\mathrm{d}\mathbf{r}_\perp, \quad (28b)$$

and

$$\Gamma^{\mu\mu'}_{\nu\nu'}=\frac{1}{N_{\mu\nu}N_{\mu'\nu'}}\iint|\mathbf{e}_{\mu\nu}|^2|\mathbf{e}_{\mu'\nu'}|^2\,\mathrm{d}\mathbf{r}_\perp. \quad (28c)$$

3.3. Free-carrier effects

The last contribution to the nonlinear polarization, stemming from free-carrier effects, may be written as follows:

$$\mathbf{P}^{\mathrm{FC}}_{\omega_\mu}(\mathbf{r},t)=2\zeta\varepsilon_0 n_{\mathrm{eff}}\left[\Delta n_{\mathrm{FC}}+ic/(2\omega_\mu)\Delta\alpha_{\mathrm{FC}}\right]\mathbf{E}_{\omega_\mu}(\mathbf{r},t), \quad (29)$$

where

$$\zeta=\frac{\partial n_{\mathrm{eff}}}{\partial n_1}=\left(\frac{\varepsilon_1}{\varepsilon_{\mathrm{eff}}}\right)^{1/2}\frac{(3f-1)\varepsilon_{\mathrm{eff}}+\varepsilon_2}{\sqrt{u^2+8\varepsilon_1\varepsilon_2}} \quad (30)$$

is the linear susceptibility attenuation factor, $n_{\mathrm{eff}}=\sqrt{\varepsilon_{\mathrm{eff}}}$, and $n_1=\sqrt{\varepsilon_1}$. The amounts of changes to the refractive index and absorption coefficient of Si NCs are related to the number density N of the TPA-generated electron–hole pairs as [4, 56]

$$\Delta n_{\mathrm{FC}}=-\sigma_n(\omega_0/\omega_\mu)^2\left(1+\varsigma N^{0.2}\right)N^{0.8}\quad\text{and}\quad\Delta\alpha_{\mathrm{FC}}=\sigma_\alpha(\omega_0/\omega_\mu)^2N, \quad (31)$$

where $\sigma_n=1.35\times10^{-22}$ m$^{2.4}$, $\varsigma=6.53\times10^{-6}$ m$^{0.6}$, $\sigma_\alpha=14.5\times10^{-21}$ m^2, and $\omega_0=2\pi c/(1.55\ \mu\mathrm{m})$.

With the above form of the nonlinear polarization, one finds that

$$\frac{e^{-i\beta_{\mu\nu}z}}{\sqrt{N_{\mu\nu}}}\iint\left(\mathbf{e}^*_{\mu\nu}\cdot\mathbf{P}^{\mathrm{FC}}_{\omega_\mu}\right)\mathrm{d}\mathbf{r}_\perp=4(\zeta/c)(n_{\mathrm{eff}}/n_{\mu\nu})\left[\Delta n_{\mathrm{FC}}+ic/(2\omega_\mu)\Delta\alpha_{\mathrm{FC}}\right]a_{\mu\nu}, \quad (32)$$

#181788 - $15.00 USD Received 14 Dec 2012; revised 15 Jan 2013; accepted 16 Jan 2013; published 29 Jan 2013

where the modal refractive index is defined as $n_{\mu\nu} = \beta_{\mu\nu}/k_\mu$.

The number density of electron–hole pairs entering Eq. (31) grows with light intensity and may be calculated from the rate equation

$$\frac{\partial N}{\partial t} = -\frac{N}{\tau_c} - \sum_\mu \frac{1}{2\hbar\omega_\mu A_{\text{eff}}} \frac{\partial P_\mu}{\partial z}, \tag{33}$$

where $\partial P_\mu/\partial z$ is the rate of mode μ power dissipation due to the TPA and τ_c is the effective free-carrier lifetime. The same average effective mode area (EMA), A_{eff}, for all modes have been assumed in this equation for the sake of simplicity. As will be discussed below, this assumption has to be abandoned if one needs to write the propagation equations in terms of the average field intensities inside the Si-NCs/SiO_2 composite.

By assuming continuous pump and Stokes waves and neglecting the time derivatives in Eq. (13), it may be readily shown using Eq. (21) that

$$\begin{aligned}\frac{\partial P_\mu}{\partial z} &= \sum_\nu \left(a_{\mu\nu} \frac{\partial a^*_{\mu\nu}}{\partial z} + a^*_{\mu\nu} \frac{\partial a_{\mu\nu}}{\partial z} \right) \\ &= -\frac{1}{4}\,\xi c^2 \varepsilon_0^2 \varepsilon_{\text{eff}} \beta_{\text{TPA}} \sum_\nu \left(\frac{8+7\rho}{45} \sum_{\nu'} 2\Gamma^{(\mu)}_{\nu\nu'} |a_{\mu\nu}|^2 |a_{\mu\nu'}|^2 + \frac{13+2\rho}{45}\, \Gamma^{(\mu)}_{\nu\nu} |a_{\mu\nu}|^4 \right).\end{aligned} \tag{34}$$

In evaluating the power dissipation rate, we have taken into account that $\Lambda^{(\mu)}_{\nu\nu} = \Gamma^{(\mu)}_{\nu\nu}$ and omitted the spatially varying terms, which arise due to the waveguide birefringence $(n_{\mu\nu} \neq n_{\mu\nu'})$ and do not contribute to the TPA-induced power dissipation.

4. Effective mode area

It should be recognized that the parameters $\Gamma^{\mu\mu'}_{\nu\nu'}$, $\Lambda^{\mu\mu'}_{\nu\nu'}$, and $\Psi^{\mu\mu'}_{\nu\nu'}$ allow one to introduce three types of EMAs, dependent on the frequencies and polarizations of both the pump and Stokes fields. For simplicity, we define the EMA of mode ν at ω_μ in the common fashion as [4, 10, 78]

$$A^{\mu\nu}_{\text{eff}} = \left(\iint |\mathbf{e}_{\mu\nu}|^2 \, \mathrm{d}\mathbf{r}_\perp \right)^2 \Big/ \iint |\mathbf{e}_{\mu\nu}|^4 \, \mathrm{d}\mathbf{r}_\perp. \tag{35}$$

Using this definition, we adopt the following definition of the average EMA:

$$A_{\text{eff}} = \prod_{\mu,\nu} \left(A^{\mu\nu}_{\text{eff}} \right)^{1/4}. \tag{36}$$

When using Eqs. (33), (35), and (36), one should keep in mind that the quantities $|a_{\mu\nu}|^2/A^{\mu\nu}_{\text{eff}}$ and $|a_{\mu\nu}|^2/A_{\text{eff}}$ are not modal intensities. In order to treat the nonlinear propagation in terms of the average mode intensities inside the nonlinear composite, one needs to use a different EMA [51],

$$\mathscr{A}^{\mu\nu}_{\text{eff}} = \mathscr{A}_{\text{NL}} \iint_\infty |\mathbf{e}_{\mu\nu}|^2 \, \mathrm{d}\mathbf{r}_\perp \Big/ \iint_{\text{NL}} |\mathbf{e}_{\mu\nu}|^2 \, \mathrm{d}\mathbf{r}_\perp, \tag{37}$$

in which $\mathscr{A}_{\text{NL}}$ is the cross section area of the nonlinear core of the waveguide (filled with Si-NCs/SiO_2 composite) and the symbols NL and ∞ denote integrations over the nonlinear core and the entire transverse plane, respectively. It is easy to see that $\mathscr{I}_{\mu\nu} = |a_{\mu\nu}|^2/\mathscr{A}^{\mu\nu}_{\text{eff}}$ gives the average intensity of field μ at ω_ν, i.e., the ratio of the power carried by this field through the waveguide core to the core area $\mathscr{A}_{\text{NL}}$.

#181788 - $15.00 USD Received 14 Dec 2012; revised 15 Jan 2013; accepted 16 Jan 2013; published 29 Jan 2013

5. Coupled amplitude equations

By combining Eqs. (13), (14), (21), (27), and (31)–(34), we arrive at the set of coupled equations for the slowly varying amplitudes $a_{\mu\nu}$ and free-carrier density N. These equations may be simplified by neglecting the effects of self steepening and assuming that no phase matching occurs between the waves of different frequencies and polarizations. In this case, the coupled amplitude equations acquire the form of the generalized nonlinear Schrödinger equation

$$\begin{aligned}\frac{\partial a_{\mu\nu}}{\partial z} &+ \sum_{n=1}^{\infty} \frac{i^{n+1}\beta_{\mu\nu}^{(n)}}{n!}\frac{\partial^n a_{\mu\nu}}{\partial t^n} + \frac{\alpha_{\mu\nu}}{2} a_{\mu\nu} \\ &= -\frac{1}{8}\,\xi c^2\varepsilon_0^2\varepsilon_{\text{eff}}\left(\beta_{\text{TPA}} - 2in_2k_\mu\right)\left(\frac{8+7\rho}{45}\,2\Gamma_{\nu\nu'}^{(\mu)}|a_{\mu\nu'}|^2 + \frac{29+16\rho}{45}\,\Gamma_{\nu\nu}^{(\mu)}|a_{\mu\nu}|^2\right)a_{\mu\nu} \\ &\quad + \frac{8i\varepsilon_0\omega_\mu}{45}\,\xi\Gamma_{\nu\nu}^{\mu\mu'} a_{\mu'\nu}(t)\int_{-\infty}^{t} a_{\mu'\nu}^*(t_1)a_{\mu\nu}(t_1)H(t-t_1)\,e^{i\omega_{\mu\mu'}(t-t_1)}\mathrm{d}t_1 \\ &\quad - \frac{4i\varepsilon_0\omega_\mu}{45}\,\xi\Gamma_{\nu\nu'}^{\mu\mu'} a_{\mu'\nu'}(t)\int_{-\infty}^{t} a_{\mu'\nu'}^*(t_1)a_{\mu\nu}(t_1)H(t-t_1)\,e^{i\omega_{\mu\mu'}(t-t_1)}\mathrm{d}t_1 \\ &\quad - \zeta\,\frac{n_{\text{eff}}}{n_{\mu\nu}}\left(\frac{\omega_0}{\omega_\mu}\right)^2\left(i\sigma_n k_\mu\left(1+\varsigma N^{0.2}\right) + \frac{\sigma_\alpha}{2}N^{0.2}\right)N^{0.8}a_{\mu\nu}, \end{aligned} \tag{38}$$

while the rate equation becomes

$$\frac{\partial N}{\partial t} = -\frac{N}{\tau_c} + \frac{\xi c^2\varepsilon_0^2\varepsilon_{\text{eff}}}{4A_{\text{eff}}}\sum_{\mu,\nu}\frac{\beta_{\text{TPA}}}{2\hbar\omega_\mu}\left(\frac{8+7\rho}{45}\,2\Gamma_{\nu\nu'}^{(\mu)}|a_{\mu\nu'}|^2 + \frac{29+16\rho}{45}\,\Gamma_{\nu\nu}^{(\mu)}|a_{\mu\nu}|^2\right)|a_{\mu\nu}|^2. \tag{39}$$

Here we have collected the terms with $\nu = \nu'$ and explicitly written out the remaining terms with $\nu' \neq \nu$. We have also employed the identity $\Lambda_{\nu\nu}^{\mu\mu'} = \Psi_{\nu\nu}^{\mu\mu'} = \Gamma_{\nu\nu}^{\mu\mu'}$ and phenomenologically added the terms $\alpha_{\mu\nu}a_{\mu\nu}/2$, which account for waveguide losses through the linear absorption coefficients $a_{\mu\nu}$.

There are several features of the derived equations that clearly distinguish them from the analogous equations for SOI waveguides [3, 56, 59, 63, 65, 79]. First, the factors ξ and ζ entering Eqs. (38) and (39) are responsible for the weakening of the nonlinear effects of Si NCs due to their embedment in fused silica. For small filling factors ($f \lesssim 0.01$), the extent of weakening may be estimated using the approximate expressions $\xi \approx (3\varepsilon_2)^4 f/(\varepsilon_1 + 2\varepsilon_2)^4 \approx 0.022f$ and $\zeta \approx 9f\varepsilon_2\sqrt{\varepsilon_1\varepsilon_2}/(\varepsilon_1 + 2\varepsilon_2)^2 \approx 0.36f$. We wish to iterate that the parameters β_{TPA}, n_2, χ_R, σ_n, and σ_α are the characteristics of individual nanocrystals, whereas the products $\xi\beta_{\text{TPA}}$, ξn_2, $\xi\chi_R$, $\zeta\sigma_n$, and $\zeta\sigma_\alpha$ characterize the Si-NCs/SiO_2 composite as a whole. Second, the strengths of self-phase modulation and TPA are reduced by about $45/(8+7\rho) \approx 2.7$ times, while the effects of cross-phase modulation and cross-TPA are enhanced by a factor of $(26+16\rho)/45 \approx 1.1$, as compared to their values in SOI waveguides fabricated along the $[0\bar{1}1]$ direction [3, 4, 79, 80]. Third, the second integral term on the right-hand side of Eq. (38) appears due to a modification of the interaction between the optical field and phonons inside the Si-NCs/SiO_2 composite and is completely absent in the equations governing the nonlinear propagation through silicon. It should be also emphasized that the coupled amplitude equations written in a coordinate basis aligned with the waveguide edges are uniform, as the information on crystallographic directions in silicon is completely lost due to the random orientation of the crystallites in the composite.

Equations (38) and (39) provide a sound theoretical foundation for modeling nonlinear optical phenomena in Si-NCs/SiO_2 waveguides and constitute the main result of this paper.

#181788 - $15.00 USD Received 14 Dec 2012; revised 15 Jan 2013; accepted 16 Jan 2013; published 29 Jan 2013

5.1. A continuous-wave regime

In the CW regime, when the amplitudes $a_{\mu\nu}$ are independent of time, the above set of equations can be simplified even further, by evaluating the two integral terms responsible for the Raman interaction as

$$\frac{4i\varepsilon_0\omega_\mu}{45}\,\xi\widetilde{H}(\omega_{\mu\mu'})\left(2\Gamma^{\mu\mu'}_{\nu\nu}|a_{\mu'\nu}|^2-\Gamma^{\mu\mu'}_{\nu\nu'}|a_{\mu'\nu'}|^2\right)a_{\mu\nu}, \tag{40}$$

where

$$\widetilde{H}(\omega)=\int_0^\infty H(t)\,e^{i\omega t}\mathrm{d}t=\frac{2\chi_R\Gamma_R\Omega_R}{\Omega_R^2+2i\Gamma_R\omega-\omega^2} \tag{41}$$

is the Raman gain profile and χ_R is related to the the Raman gain coefficient g_R as [4, 69] $\chi_R=2\varepsilon_0\varepsilon_{\text{eff}}c^2g_R/\omega_\mu$.

We can also formally solve Eq. (39) and introduce its solution into Eq. (38) to get a set of four coupled equations solely for the field amplitudes. In doing so, it is more convenient to use an approximate relation $\Delta n_{\text{FC}}\approx-\bar{\sigma}_n(\omega_0/\omega_\mu)^2N$, with $\bar{\sigma}_n=5.3\times10^{-27}$ m^3 [4] for typical free-carrier densities in the range 10^{22} to 10^{23} m^{-3} [the measurements of Spano *et al.* [71] give $\zeta\bar{\sigma}_n=(1.2\pm0.3)\times10^{-28}$ m^3 for $f=8\%$]. Some algebra shows that Eqs. (38) and (39) in this case yield

$$\begin{aligned}\frac{\partial\ln a_{\mu\nu}}{\partial z}+\frac{\alpha_{\mu\nu}}{2}=&-\xi\left(\frac{\beta_{\text{TPA}}}{2}-in_2k_\mu\right)\left(\frac{8+7\rho}{45}2\gamma^{(\mu)}_{\nu\nu'}I_{\mu\nu'}+\frac{29+16\rho}{45}\gamma^{(\mu)}_{\nu\nu}I_{\mu\nu}\right)\\&+\frac{32}{45}\xi\widetilde{g}_R(\omega_{\mu\mu'})\left(2\gamma^{\mu\mu'}_{\nu\nu}I_{\mu'\nu}-\gamma^{\mu\mu'}_{\nu\nu'}I_{\mu'\nu'}\right)-\zeta\xi\tau_c\frac{n_{\text{eff}}}{n_{\mu\nu}}\left(\frac{\omega_0}{\omega_\mu}\right)^2\left(\frac{\sigma_\alpha}{2}+i\bar{\sigma}_nk_\mu\right)\\&\times\sum_{\mu,\nu}\frac{\beta_{\text{TPA}}}{2\hbar\omega_\mu}\left(\frac{8+7\rho}{45}2\gamma^{(\mu)}_{\nu\nu'}I_{\mu\nu'}+\frac{29+16\rho}{45}\gamma^{(\mu)}_{\nu\nu}I_{\mu\nu}\right)I_{\mu\nu},\end{aligned} \tag{42}$$

where $I_{\mu\nu}=|a_{\mu\nu}|^2/A_{\text{eff}}$, $\gamma^{(\mu)}_{\nu\nu'}=\gamma^{\mu\mu}_{\nu\nu'}$,

$$\gamma^{\mu\mu'}_{\nu\nu'}=\frac{n^2_{\text{eff}}}{n_{\mu\nu}n_{\mu'\nu'}}\frac{A_{\text{eff}}\iint|\mathbf{e}_{\mu\nu}|^2|\mathbf{e}_{\mu'\nu'}|^2\,\mathrm{d}\mathbf{r}_\perp}{\iint|\mathbf{e}_{\mu\nu}|^2\,\mathrm{d}\mathbf{r}_\perp\iint|\mathbf{e}_{\mu'\nu'}|^2\,\mathrm{d}\mathbf{r}_\perp}, \tag{43a}$$

and

$$\widetilde{g}_R(\omega)=\frac{2ig_R\Gamma_R\Omega_R}{\Omega_R^2+2i\Gamma_R\omega-\omega^2}. \tag{43b}$$

For exact resonance, $\omega_p-\omega_s=\Omega_R$, the last expression gives $\widetilde{g}_R(\Omega_R)=g_R$.

As we mentioned earlier, the quantities $I_{\mu\nu}$ entering Eq. (42) do not represent modal intensities, as they are defined through the average EMA of the waveguide. One may rewrite the propagation equations in terms of the average field intensities $\mathscr{I}_{\mu\nu}$, by simply replacing A_{eff} with $\mathscr{A}^{\mu'\nu'}_{\text{eff}}$ in the definition of the confinement factors $\gamma^{\mu\mu'}_{\nu\nu'}$.

Although it is now possible to use the developed theory for modeling various nonlinear effects in silicon-nanocrystal waveguides, a thorough analysis of even the simplest propagation scenario (involving one continuous wave) is rather complicated and warrants a comprehensive discussion. We therefore intend to present such an analysis in future publications on nonlinear optics in the Si-NCs/SiO$_2$ waveguides.

#181788 - $15.00 USD Received 14 Dec 2012; revised 15 Jan 2013; accepted 16 Jan 2013; published 29 Jan 2013

6. Conclusions

We have developed a general theory of two-frequency pulse propagation through silicon-nanocrystal-doped silica waveguides in the presence of the Kerr effect, stimulated Raman scattering, two-photon absorption (TPA), and the associated nonlinear optical phenomena. The dynamics of arbitrarily polarized pump and Stokes pulses was described using the system of four coupled nonlinear equations for the slowly varying amplitudes of the pulses, and the rate equation for the number density of TPA-generated free carriers. The equations were derived by assuming a uniform distribution of nanocrystal axes in space and treating the third-order nonlinear response of the waveguide within the framework of the effective medium theory. They allow for the modification of light interaction with electron and phonon subsystems of silicon nanocrystals, and take into account the weakening of the nanocrystals' nonlinear response due to their embedment in silica matrix. The mathematics in the paper was dealt with in much detail, for methodological reasons and to facilitate the application of the developed theory by experimentalists. Our theory may be used to study a wide variety of nonlinear phenomena in silicon-nanocrystal silica waveguides, including the polarization rotation induced in optical pulses by self- and cross-phase modulation, Raman amplification and lasing, as well as TPA-induced optical modulation.

Acknowledgments

This work is sponsored by the Australian Research Council, through its Discovery Early Career Researcher Award DE120100055.