Optical transistor action by nonlinear coupling of stimulated emission and coherent scattering

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ABSTRACT

In the pursuit of improved platforms for computing, communications and internet connectivity, all-optical systems offer excellent prospects for a speed and fidelity of data transmission that will greatly surpass conventional electronics, alongside the anticipated benefits of reduced energy loss. With a diverse range of sources and fiber optical connections already in production, much current effort is being devoted towards forging optical components for signal switching, such as an all-optical transistor. Achievement of the desired characteristics for any practicable device can be expected to depend crucially on the engagement of a strongly nonlinear optical response. The innovative scheme proposed in the present work is based upon a third-order nonlinearity – its effect enhanced by stimulated emission – operating within a system designed to exploit the highly nonlinear response observed at the threshold for laser emission. Here, stimulated emission is strongly driven by coupling to the coherent scattering of a signal input beam whose optical frequency is purposely off-set from resonance. An electrodynamical analysis of the all-optical coupling process shows that the signal beam can significantly modify the kinetics of emission, and so lead to a dramatically enhanced output of resonant radiation. The underlying nonlinear optical mechanism is analyzed, model calculations are performed for realizable three-level laser systems, and the results exhibited graphically. The advantages of implementing this all-optical transistor scheme, compared to several previously envisaged proposals, are then outlined.

Keywords: optical switching, optical transistor, all-optical processing, laser action, nonlinear optics, stimulated emission, quantum electrodynamics, coherent scattering

1. INTRODUCTION

The advantages of all-optical systems for information processing, computing, communications and internet connection are familiar and well-rehearsed. When compared to current electronic implementations, numerous anticipated benefits can be identified including reduced energy losses, and a greatly increased speed and fidelity of data transmission. As the optical communications industry gears up for the second decade of a millennium already transformed by informatics, much effort is being devoted towards devising new optical components such as an all-optical transistor. Like its electronic antecedent, the underlying principle of such a device is to effect the switching or amplification of a source, under the control of a data signal input. Amongst many novel schemes for the realization of such a device, one molecular device based on saturated absorption has very recently been proposed by Hwang et al.[1] Other, all-optical switching systems that have recently been proposed are based on electromagnetic induced transparency,[2-4] the optical Kerr effect,[5,6] nonlinear transmission through coupling with surface plasmons,[7-9] and beam filament rotation by application of a signal beam.[10,11] A particular commonality of principle can be found in each of these; the engagement of a strongly nonlinear optical response. However, most such schemes are tailored for application to a specific combination of material and optical system.

The scheme that is proposed in the present work is based upon a recently identified third-order optical nonlinearity, its effect enhanced by stimulated emission. This mechanism is brought into play in a system that is designed to exploit the highly nonlinear response of a system at the threshold for laser emission. In contrast to the work of Hwang et al., the role of the probe beam, whose optical frequency is purposely off-set from resonance, is to passively engage by coherent forward scattering with resonant stimulated emission. Detailed analysis shows that this beam, acting as the input signal, can modify the kinetics of emission and so lead to an enhanced output.[12,13] Significantly, optically controlled

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fluorescence is a process that is not limited to operation with any one material; with a judicious choice of signal optical frequency, operation of the mechanism is viable in any suitably nonlinear medium. Moreover the symmetry conditions for the existence of the appropriate nonlinearity are not unduly restrictive. Once the principle is proven, a variety of new optical data handling components can be anticipated to emerge. The following results of calculations, for three-level laser systems, highlight the significant potential for device development.

2. LASER-MODIFIED EMISSION

First, we consider the general principle of laser-modified, optically controlled emission. In general, a single matter-photon interaction is responsible for spontaneous emission, as depicted in Fig. 1(*a*), and the standard theory for the process is accordingly developed through first-order time-dependent perturbation theory. Even-order perturbative terms vanish, and higher order odd-rank correction terms are insignificant when no other light is present – they simply denote self-energy corrections. However this is no longer the case when the electronically excited fluorophore is subjected to a throughput of off-resonant, pulsed laser light. Then, significant non-zero corrections can arise even at relatively modest intensity levels – it being assumed that the wavelength of the latter is chosen to preclude stimulated emission or any excitation of the material to higher electronic levels. With a probe laser detuned to a region where the system is transparent, although in consequence there can be no net absorption or stimulated emission, elastic forward-scattering events will occur, photons being annihilated and created into the same radiation mode. The throughout light, which thus emerges unchanged, nonetheless passively engages by nonlinear coupling with the spontaneous emission, and the net effect is to modify the transition moment for electronic decay. This mechanism, represented in Fig. 1(*b*), entails three matter-photon interactions, i.e. third-order perturbation theory.

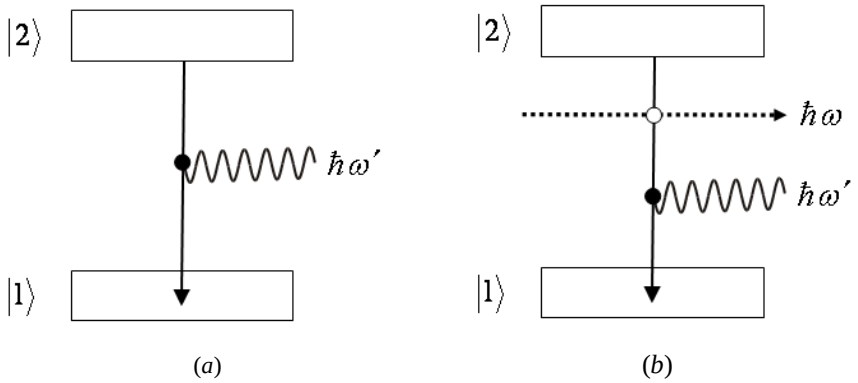

Fig. 1. (*a*) Energy level representation for spontaneous emission. Electronic states (and their vibrational manifolds) are signified by boxes, the wavy line is the radiative emission ($\hbar\omega'$) and the black vertical arrow the decay transition: $|1\rangle$ and $|2\rangle$ are the electronic ground and excited states, respectively, the black dot symbolizing a single matter-photon interaction. (*b*) Same emission but engaging an off-resonant laser beam ($\hbar\omega$) denoted by the horizontal dashed arrow; the open dot symbolizes two matter-photon interaction (i.e. elastic forward-scattering).

The radiant intensity of emission, $I'(\Omega')$ (power per unit solid angle), which follows from the Fermi Golden Rule rate[14] multiplied by the energy of an emission photon, $\hbar ck' \equiv \hbar\omega'$,[15] is now determined from $I'(\Omega') = 2\pi\rho ck' |M^{(1)} + M^{(3)}|^2$, where $M^{(1)}$ and $M^{(3)}$ are the quantum amplitudes for the first- and third-order interaction processes, respectively, and the density of radiation states is $\rho = \left(k^2 V / 8\pi^3 \hbar c\right) d\Omega$, where $d\Omega$ signifies an element of solid angle for the emission.[16] The effects to be considered below now depend on the relative signs of the first- and third-order amplitudes; a common sign leads to emission enhancement, opposite signs its suppression. To proceed, the following is found for a given emission polarization;

$$I'(\Omega') = \left(\frac{ck'^4}{8\pi^2\varepsilon_0}\right)\Big[e'_i e'_j \mu_i^{12}\bar{\mu}_j^{12} + (I/c\varepsilon_0)\, e'_i e_j e_k e'_l \chi_{ijk}^{12}(\omega';-\omega,\omega)\,\bar{\mu}_l^{12}$$
$$+\left(I^2/4c^2\varepsilon_0^2\right) e'_i e_j e_k e'_l e_m e_n \chi_{ijk}^{12}(\omega';-\omega,\omega)\,\bar{\chi}_{lmn}^{12}(\omega';-\omega,\omega)\Big] \quad , \tag{1}$$

where χ_{ijk}^{12} is a transition hyperpolarizability (nonlinear susceptibility) tensor, **e** and $\hbar\omega$ correspond to a probe laser photon, and I is the irradiance of the laser probe. Furthermore, the normal decay transition dipole moment is designated by the shorthand notation $\boldsymbol{\mu}^{12} = \langle 1|\boldsymbol{\mu}|2\rangle$ – in which $|1\rangle$ and $|2\rangle$ denote the states of levels E_1 and E_2, respectively. In equation (1), the Einstein implied summation convention for repeated (Cartesian) indices is deployed. The initial term on the right-hand side of (1) corresponds to spontaneous emission – the usual one-photon transition, intrinsic to the system and independent of the probe laser beam – while the last term signifies coupling of the elastically forward scattered probe beam with the spontaneous emission, overall a three-photon event. The second term, linear in I, represents a quantum interference of these two concurrent processes. In general, it may be assumed that the leading term in (1) is non-zero, and the second term a leading correction.

The key parameter within equation (1) is the nonlinear transition (inelastic) susceptibility, χ_{ijk}^{12}, mediating radiative decay of the molecular excited state – in consequence of which, the first frequency parameter ω', registering the molecular decay, differs from the sum of those which follow in the argument of χ. The explicit form of this tensor is determined from well-attested and reported methods,[15,17-19] and the result is given by:

$$\chi_{ijk}^{12}(\omega';-\omega,\omega) = \sum_r \sum_{s\neq 2}\left(\frac{\mu_i^{1s}\mu_j^{sr}\mu_k^{r2}}{E_{s2}(E_{r2}-\hbar\omega)} + \frac{\mu_i^{1s}\mu_k^{sr}\mu_j^{r2}}{E_{s2}(E_{r2}+\hbar\omega)}\right)$$
$$+\sum_r \sum_s\left(\frac{\mu_j^{1s}\mu_i^{sr}\mu_k^{r2}}{(E_{s2}-\hbar\omega+\hbar\omega')(E_{r2}-\hbar\omega)} + \frac{\mu_k^{1s}\mu_i^{sr}\mu_j^{r2}}{(E_{s2}+\hbar\omega+\hbar\omega')(E_{r2}+\hbar\omega)}\right)$$
$$+\sum_{r\neq 1}\sum_s\left(\frac{\mu_j^{1s}\mu_k^{sr}\mu_i^{r2}}{(E_{s2}-\hbar\omega+\hbar\omega')(E_{r2}+\hbar\omega')} + \frac{\mu_k^{1s}\mu_j^{sr}\mu_i^{r2}}{(E_{s2}+\hbar\omega+\hbar\omega')(E_{r2}+\hbar\omega')}\right) \quad , \tag{2}$$

where ω is the signal beam frequency, and the transition moments are defined in the same manner as $\boldsymbol{\mu}^{12}$; $|r\rangle$ and $|s\rangle$ are intermediate states, and $E_{xy} = E_x - E_y$ is an energy difference between two states. In passing, we observe that the nonlinear mechanism may alternatively be interpreted in terms of a 'dressing' of the molecular states by the throughput beam, manifest in a modification to the $E_2 \rightarrow E_1$ transition moment; although a different derivation method ensues, the same expression as equation (2) will emerge. The third-rank polar tensor (2) has non-vanishing elements if the product of the initial and final state symmetries spans one or more components of suitable symmetry. In fact, any transition that is electric-dipole allowed will also support a non-vanishing χ^{12}.

3. OPTICAL TRANSISTOR ACTION

We now focus consideration on a typical three-level laser system optically pumped within a microcavity. The kinetics of emission are primarily determined by a pump rate R_p driving population from the ground state E_0 into a metastable upper level E_2, lasing action from E_2 into E_1, and ultrafast relaxation from E_1 (Fig. 2). Following Siegman,[20] the rate equations corresponding to the temporal behavior of the cavity photon number, n, and the E_2 population, N_2, are as follows:

$$\frac{dn}{dt} = K(n+1)N_2 - \gamma_c n \quad , \tag{3}$$

$$\frac{dN_2}{dt} = R_p - nKN_2 - \gamma_2 N_2 \quad . \tag{4}$$

Here, the population of E_1 is assumed to be vanishingly small; K denotes the coupling coefficient for the laser transition, whilst γ_c and γ_2 signify the cavity and population decay rate, respectively. Under steady state conditions, equations (3) and (4) may be solved to give the result;

$$n = \frac{R_p - g\gamma_c p + \left[\left(R_p - g\gamma_c p\right)^2 + 4\gamma_c R_p\right]^{\frac{1}{2}}}{2\gamma_c} \quad . \tag{5}$$

where $g = \gamma_2/\gamma_{\text{rad}}$ and $K = \gamma_{\text{rad}}/p$ (in which γ_{rad} is the radiative decay rate and p is the number of resonant cavity modes). The relaxation from E_2 into E_1 is not entirely radiative, i.e. $\gamma_2 \neq \gamma_{\text{rad}}$, since non-radiative relaxation processes (the excitation of lattice phonons etc.) arise. For present calculational purposes, given that the level E_2 decay rate γ_2 subsumes (but is dominated by) the rate of radiative decay, a value of 5/4 is to be assumed for g in the absence of the off-resonant input signal considered below. Proceeding from equation (5), employing typical values p = 10^{10} and $\gamma_c = 10^8$ s^{-1}, the familiar vertical climb in cavity photons at laser threshold emerges, as graphically illustrated by Fig. 3 (solid line). All-optical control of such a pumped active medium may be achieved by nonlinear optical engagement of the laser emission with stimulated elastic forward scattering of off-resonant (signal) laser pulses, effecting a modification to the dipole transition moment for the $E_2 \rightarrow E_1$ laser transition.

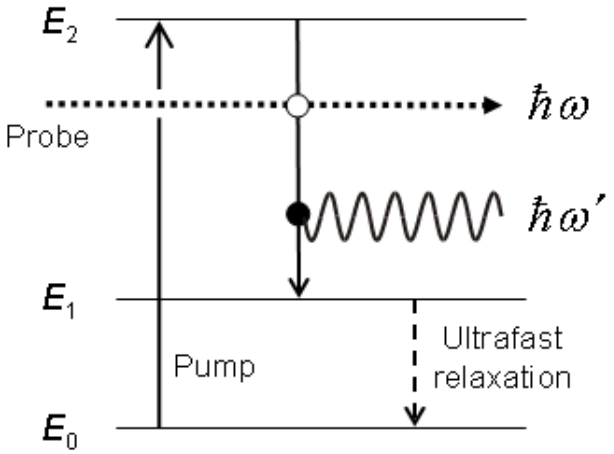

Fig. 2. Energy level diagram for the three-level laser system: black solid arrows denote electronic transitions, the wavy line emission ($\hbar\omega'$), and the dashed arrow, the off-resonant probe beam ($\hbar\omega$). Black and open dots symbolize single and dual matter-photon interactions, respectively. Discrete energy levels are depicted for simplicity.

Returning to equation (2), r and s now equate to either 0, 1 or 2 in the three-level system of Fig. 2 – except when precluded in certain summations, as indicated in (2). We may also suppose $\omega' < \omega$, and that these frequencies determine an offset, $\Delta E = E_{20} - \hbar\omega - \hbar\omega'$, that is a small fraction of the energy for a typical electronic transition. Under these conditions the fourth term of equation (2) is dominant, delivering the following:

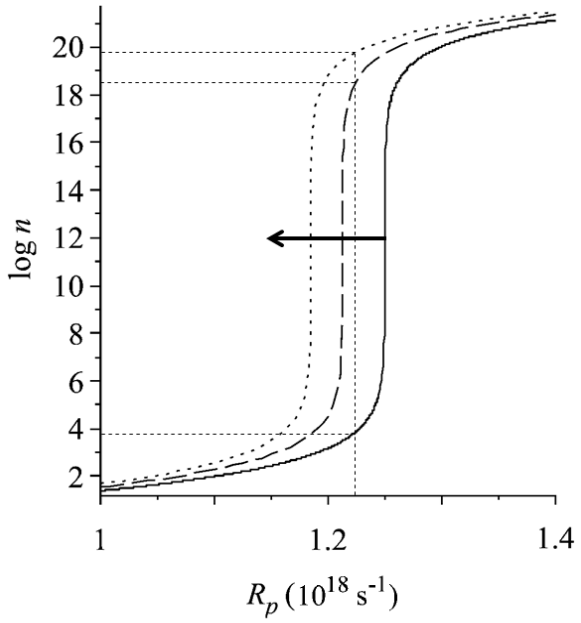

Fig. 3. Plot of log *n*, where *n* is the number of cavity photons, against the pumping rate, R_p, for absent (solid line) and present signal beam; example irradiances of the latter are 2×10^{11} W cm^{-2} (dashed curve) and 4×10^{11} W cm^{-2} (dotted). Horizontal arrow illustrates movement of the lasing threshold to the left for increasing laser intensities. The vertical dotted line represents a constant R_p at which an introduction of the signal beam produces above-threshold operation (denoted by the upper pair of horizontal dotted lines).

$$e'_i e_j e_k \chi^{12}_{ijk}\left(\omega';-\omega,\omega\right) \approx \frac{\mu^3}{\Delta E\left(\Delta E+\hbar\omega'\right)} \quad . \tag{6}$$

To secure a more quantitative assessment of the generic scheme, it is expedient to assume that the relevant transition dipole moment components have broadly similar direction and magnitude – the latter now simply represented as μ. In calculations on specific systems, this approximation can of course be surrendered for greater accuracy. It should be observed that both factors in the denominator of (6) have negative values, so that the resulting susceptibility is always positive and in consequence leads to enhanced emission; under other conditions the susceptibility components may assume a negative value, representative of reduced emission. On insertion of equation (6) into (1), typical values of $I'\left(\Omega'\right)$ may be calculated for various signal beam intensities. As indicated in Section 2, it is the second term of (1) (linear in *I*) that represents the leading correction. With this in mind, the degree of enhancement (or in other cases any suppression) of the emission can be measured by taking the ratio of the second term against the first in equation (1); the corresponding parameter η may be approximated as:

$$\eta = \frac{I\mu^2}{c\varepsilon_0 \Delta E\left(\Delta E+\hbar\omega'\right)} \quad . \tag{7}$$

Returning to equation (1), it is clear that the variable *g* will be affected by introduction of the input signal beam, since the radiative decay rate, γ_{rad}, and population decay rate, γ_2, both thereby suffer change (but to differing degrees); the non-radiative decay rate, γ_{nr}, can be assumed to be constant. By simple manipulation, an expression for *g* is given by;

$$g\left(I\right) = 1 + \frac{1-Y\left(I\right)}{Y\left(I\right)+\eta Y\left(I\right)} \quad , \tag{8}$$

where $Y(I) = \gamma_{\mathrm{rad}}(I)/\gamma_2(I)$ and $\gamma_{\mathrm{nr}}/\gamma_2(I) = 1 - Y(I)$. With the previous condition that g = 5/4 for I = 0, and adopting indicative values $\mu = 16 \times 10^{-30}$ C m, $\Delta E = 10^{-20}$ J and $\hbar\omega' = 10^{-19}$ J, insertion of (8) into (5) generates the results exhibited in Fig. 3. It is clearly evident that transistor action with respect to the signal beam occurs. For a constant pumping rate at a level indicated by the dotted vertical line, the system operates below threshold when no signal laser present; however on the introduction of an off-resonant beam with an irradiance approaching 2×10^{11} W cm^{-2}, the device output climbs by fourteen orders of magnitude, rising to sixteen if the signal input is doubled.

4. DISCUSSION

It is interesting to compare the scheme set forth above with the three-level proposal recently offered by Hwang et al.[1] These authors devised a system based on the variable transmission of a tunable probe, which is a continuous-wave narrow bandwidth beam whose optical frequency is scanned across a 0-1 transition (Fig. 3). When the probe beam is coincident in time with ultrashort pulses from a dye laser acting as pump, their frequency locked onto the 0-2 transition, the probe absorption loses intensity as the pump intensity is increased (optical bleaching) – eventually being observed to marginally increase the on-resonance throughput intensity. The authors have interpreted this enhancement of the probe as amplification; logically the effect derives from stimulated emission of the transition between the two lower levels, level 1 being populated by rapid decay from level 2. A decay channel from level 1 into a vibrationally excited sub-level of the ground state affords an optical output that can be registered against zero background.

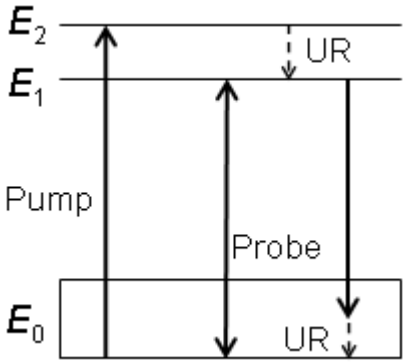

Fig. 3. Energy level scheme for the three-level laser system of Hwang et al.: double-headed arrow denotes electronic transition due to a continuous-wave (and resonant) probe beam, upward arrow represents a pumping dye laser transition, downward arrow is a Stokes-shifted decay channel, and dashed arrows denote non-radiative ultrafast relaxation (UR).

In contrast, the all-optical transistor system we propose offers several apparent advantages. First and foremost, off-resonant activation of laser emission is effected by a throughput beam which operates entirely passively; the latter beam experiences no loss or gain of intensity, and the optical output that is fully delivered into a zero background channel. The reported mechanism offers ultrafast response with high repetition rate, high efficiency, and a straightforward experimental setup. Moreover, it is based on a principle that is not limited to operation with any one specific material; with judicious choice of signal optical frequency, the mechanism is viable in any suitably nonlinear medium. Our analysis paves a new pathway for all-optical switching and amplification. The realization of a system suitable for implementing this mechanism is an enticing goal, whose achievement will require the identification of systems for which the key tensor parameters can be optimized; all the necessary theory is now delivered.

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REFERENCES

[1] Hwang, J., Pototschnig, M., Lettow, R., Zumofen, G., Renn, A., Götzinger, S. and Sandoghdar, V., "A single-molecule optical transistor," Nature 460, 76-80 (2009).

[2] Ham, B. S. and Hemmer, P. R., "Coherence switching in a four-level system: quantum switching," Phys. Rev. Lett. 84, 4080 (2000).

[3] Bermel, P., Rodriguez, A., Johnson, S. G., Joannopoulos, J. D. and Soljacic, M., "Single-photon all-optical switching using waveguide-cavity quantum electrodynamics," Phys. Rev. A 74, 043818 (2006).

[4] Raymond Ooi, C. H., "Controlling irreversibility and directionality of light via atomic motion: optical transistor and quantum velocimeter," New J. Phys. 10, 123024 (2008).

[5] Yanik, M. F., Fan, S. H., Soljacic, M. and Joannopoulos, J. D., "All-optical transistor action with bistable switching in a photonic crystal cross-waveguide geometry," Opt. Lett. 28, 2506-2508 (2003).

[6] Singh, M. R. and Lipson, R. H., "Optical switching in nonlinear photonic crystals lightly doped with nanostructures," J. Phys. B 41, 015401 (2008).

[7] Janke, C., Rivas, J. G., Bolivar, P. H. and Kurz, H., "All-optical switching of the transmission of electromagnetic radiation through subwavelength apertures," Opt. Lett. 30, 2357-2359 (2005).

[8] Wurtz, G. A., Pollard, R. and Zayats, A. V., "Optical bistability in nonlinear surface-plasmon polaritonic crystals," Phys. Rev. Lett. 97, 057402 (2006).

[9] Chang, D. E., Sorensen, A. S., Demler, E. A. and Lukin, M. D., "A single-photon transistor using nanoscale surface plasmons," Nature Phys. 3, 807-812 (2007).

[10] Dawes, A. M. C., Illing, L., Clark, S. M. and Gauthier, D. J., "All-optical switching in rubidium vapour," Science 308, 672-674 (2005).

[11] Schumacher, S., Kwong, N. H., Binder, R. and Smirl, A. L., "Low intensity directional switching of light in semiconductor microcavities," Phys. Status Solidi RRL 3, 10-12 (2009).

[12] Bradshaw, D. S. and Andrews, D. L., "Mechanism for optical enhancement and suppression of fluorescence," J. Phys. Chem. A 113, 6537-6539 (2009).

[13] Bradshaw, D. S. and Andrews, D. L., "All-optical control of molecular fluorescence," Phys. Rev. A 81, 013424 (2010).

[14] Mandel, L. and Wolf, E., [Optical Coherence and Quantum Optics], University Press, Cambridge, 871 (1995).

[15] Andrews, D. L. and Allcock, P., [Optical Harmonics in Molecular Systems], Wiley-VCH, Weinheim (2002).

[16] Craig, D. P. and Thirunamachandran, T., [Molecular Quantum Electrodynamics], Dover, New York (1998).

[17] Kuzmany, H., [Solid-state Spectroscopy: An Introduction], Springer-Verlag, Berlin, 184 (2002).

[18] Wagnière, G. H., [Linear and Nonlinear Optical Properties of Molecules], VCH-Verlag, Weinheim (1993).

[19] Andrews, D. L. and Bradshaw, D. S., "A photonic basis for deriving nonlinear optical response," Eur. J. Phys. 30, 239-251 (2009).

[20] Siegman, A. E., [Lasers], Oxford University Press, Oxford, 510-516 (1986).