

NONLINEAR OPTICS BASICS

Units and Nomenclature

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Key words: nonlinear optics; lasers; nomenclature; units; dimensions; terminology; nonlinear absorption; nonlinear refraction; nonlinear polarization; nonlinear susceptibility

Nomenclature Associated with the Excitation Light

Particular care must be used when characterizing the excitation beam in nonlinear optical experiments compared to linear measurements. By definition, nonlinear optical phenomena depend on the electric field to high order. The higher the order, the more sensitive the observed behavior depends on the input. Extracting a representative nonlinear coefficient from an experiment, for example, becomes progressively more difficult as the order of the optical nonlinearity increases. In other words: the errors associated with optical beam characterization get magnified by the order of the nonlinearity under study.

There is an extensive nomenclature for characterizing the light (almost always laser light) interacting with the nonlinear optical medium. Different descriptions may be used depending on whether the excitation light is pulsed, continuous, or a continuous train of pulses. When a laser beam is constant or continuous, it is often described by its 'cw power'. 'Cw' stands for 'continuous wave', an acronym taken from the nomenclature of electronics. The cw power of a laser is determined by placing a power meter in the path of a beam. Repeatedly pulsed lasers can be characterized in the same way, provided the response time of the power meter is slower than the pulse separation period. This will almost certainly be the case with a continuously pumped mode-locked laser such as a dye laser, fiber laser, or Ti:sapphire laser, which produce pulses at repetition frequencies of tens of megahertz. Cw power may also be suitable for

describing pulsed flashlamp lasers, gas discharge lasers, or any laser that is excited in a periodic fashion.

When the optical output can be distinguished as individual pulses, additional metrics are used. A pulse, by definition, exists during a window of time, i.e., there is a time when light is present and a time when light is absent. How one characterizes the time light is present – the pulse duration – is critically important and not always obvious. There are a myriad of ways the pulse temporal envelope can manifest itself; common examples include square pulses, triangular pulses, Gaussian pulses, and hyperbolic secant pulses. These names refer to the mathematical waveforms that map the pulse envelope as a function of time.

A pulse waveform that exhibits symmetry about the peak in its temporal envelope is commonly characterized by its 'full-width, half-maximum', abbreviated FWHM. One obtains the full-width, half-maximum by locating the two points on the pulse profile that are at half the peak value. The temporal separation of these two points is the FWHM. One uses this measure because in a strict mathematical sense, waveforms such as the Gaussian or hyperbolic-secant exist even at times $t = \pm\infty$. A less common term is the 'half-width, half-maximum' or HWHM. As the name implies, the HWHM is exactly half the FWHM value. Not all light-pulses are temporally symmetric, however, so greater detail may be needed when giving a mathematical description of asymmetric pulses.

Light pulses are also characterized by their energy and peak power. The temporal envelope recorded by a so-called square-law detector (all laboratory detectors are square-law detectors) shows the power of the pulse as a function of time, provided the detector response time is sufficiently fast. Integrating this waveform gives the pulse energy. It makes no sense to talk about the energy of a cw beam of light, unless that beam is composed of repetitive, distinguishable pulses. Each pulse in the train carries a distinct amount of optical energy.

'Ultrafast' or 'ultrashort' laser pulses generally refer to light pulses that are of such small duration they cannot be measured directly with detectors and oscilloscopes because of bandwidth limitations. To infer the pulse duration, so-called intensity autocorrelation measurements are often made. The temporal power profile is then deduced from the autocorrelation signal with the appropriate mathematical

conversion factor. Equally important is the spectrum of a short pulse. The optical bandwidth determines the lower limit of temporal compression; such ideally compressed pulses are said to be ‘transform limited’. If different components of the spectrum can be distinguished at specific positions on the pulse temporal profile, then the pulse is ‘chirped’. Information on the methods of ultrafast laser science can be found in the chapter by ???.

Nonlinear optical effects take place when light is concentrated on a target – there is a cross-section or ‘footprint’ of the beam on the medium. The interaction of the light and material takes place in a region called the beam area, beam cross-section, focal area, or focal volume. One must then define the appropriate interaction area or volume.

Specification of the beam area allows one to define two important quantities used frequently in nonlinear optics. The power/area is known as irradiance or power density. This quantity is commonly called the intensity, although the strict radiometric definition of intensity is power/(solid angle). The second important quantity is energy/area, which is called the energy density or fluence.

The convention in nonlinear optics is to define the optical electric field as:

$$\mathbf{E}(\mathbf{z}, t) = \frac{1}{2} E_0 \exp[i(\mathbf{k} \cdot \mathbf{z} - \omega t)] + \text{c.c.} \quad [1]$$

where E_0 is the peak field, \mathbf{k} is the propagation vector, ω is the angular frequency of the light, and c.c. stands for complex conjugate. This represents a forward- and backward-propagating (in the z -direction) infinite, transverse plane wave. Other definitions are also used. In SI/mks units and using the convention of eqn [1], the irradiance (I in units of W/m^2) inside a material of refractive index n is related to the electric field vector of the light (\mathbf{E} in units of V/m) as follows:

$$I = \frac{1}{2} c n \epsilon_0 |\mathbf{E}|^2 \quad [2]$$

Here c is the speed of light (2.998×10^8 m/s) and ϵ_0 is the permittivity of free space (8.854×10^{-12} F/m). Nonlinear optics has an unfortunate tradition of mixing mks and cgs units, resulting in a lot of confusion. Irradiance is usually expressed in units of W/cm^2 . One can calculate the peak electric field (in units of V/cm) of a laser beam given its irradiance (in units of W/cm^2) by using this simple formula:

$$E_0 = 38.82 \sqrt{\frac{I}{n}} \left(\frac{\text{V}}{\text{cm}} \right) \quad [3]$$

The root-mean-square value of the electric field is obtained by replacing the factor 38.82 by 27.45. In Gaussian/cgs units, irradiance is expressed in units of $\text{erg}/(\text{cm}^2 \text{ sec})$ and is related to the field as:

$$I = \frac{cn}{8\pi} |\mathbf{E}|^2 \quad [4]$$

where the field is in units of statvolt/cm and $c = 2.998 \times 10^{10}$ cm/sec. Conversion between mks and cgs for the electric field is $3 \times 10^4 \text{ V/m} = 1 \text{ statvolt/cm}$; irradiance is converted using $1 \text{ erg}/(\text{cm}^2 \text{ sec}) = 1 \times 10^{-3} \text{ W}/\text{m}^2$. The following point must be emphasized: depending on how the electric field is defined, there can be factors of 2 or even 4 discrepancies in the irradiance values quoted by different authors. The common definitions are used here, although they are certainly not universal.

A very common realization of the spatial irradiance profile of a laser beam is the radially symmetric Gaussian function:

$$I(r) = I_0 \exp\left(-\frac{2r^2}{w^2}\right) \quad [5]$$

where I_0 is the peak irradiance at the center of the beam ($r = 0$) and w is the ‘spot size’. The spot size changes continuously as the beam propagates and the minimum spot size is known as the waist. One often hears the spot size referred to as the radius, but the radius of a Gaussian beam traditionally denotes the curvature of the phase front. By definition, the phase front radius is infinite at the waist. The factor of 2 appearing in the argument of the exponential in eqn [5] stems from the fact that the spot size w is conventionally defined for the electric field of the Gaussian beam. When the field is squared to obtain the irradiance, the factor of 2 appears. Using this function for the irradiance spatial profile leads to the common parameter ‘ $1/e^2$ diameter’. The irradiance falls to $1/e^2$ (0.1353) of its peak value when $r = w$.

The Gaussian spatial profile or ‘Gaussian beam’ results when the laser has been designed to operate in the lowest-order transverse mode, usually denoted TEM_{00} . The TEM_{00} Gaussian beam is particularly convenient because the same relative power profile is maintained in the near- and far-field, whether or not the beam is collimated or focused.

The cross-sectional area of a TEM_{00} Gaussian beam normally incident on a surface is found as follows. The power in the beam is obtained by integrating the irradiance profile (I) over

the surface:

$$P = \int_{\pm} \int_{\infty} I dA \quad [6]$$

Referring to eqn [5], the integral becomes:

$$P = \int_0^{2\pi} d\theta \int_0^{\infty} r dr I_0 \exp\left(-\frac{2r^2}{w^2}\right) = I_0 \frac{\pi w^2}{2} \quad [7]$$

Hence the effective area of a TEM₀₀ Gaussian beam normally incident on a surface is:

$$A = \frac{\pi w^2}{2}. \quad [8]$$

There are many situations where the Gaussian formulation of eqn [5] is not suitable. Beams produced by unstable laser resonators, for example, are not Gaussian. A multitransverse mode beam profile obtained from a stable resonator does not usually lend itself to a simple mathematical characterization.

Nomenclature Associated with the Nonlinear Optical Medium

In nonlinear optics texts, the subject is often introduced by writing the macroscopic polarization in Maxwell's equations as a power series expansion in the electric field. This approach, proposed by Bloembergen and coworkers in the early 1960s, has been spectacularly successful for interpreting experiments, though it has also led to confusion in the definition of nonlinear optical coefficients. The confusion, which stems from arbitrary definitions and nomenclature used by different authors, cannot be resolved here. The reader should, however, be alert for these discrepancies. Comparison of results published by different laboratories requires that the fundamental equations for extracting the nonlinear coefficients from their data are known. As the discipline has matured, the nonlinear optics community largely recognized the ambiguities and confusion; precise definitions of terms and coefficients are now commonly provided in the research literature.

It should also be noted that the power-series framework may not always be the best formulation for modeling and characterizing nonlinear optical effects. A carefully designed optical limiter, for example, may exhibit an abrupt decrease of transmission at a specific 'threshold irradiance' or 'threshold fluence', perhaps at a given laser pulse

duration. The physical phenomenon or phenomena driving this behavior may not readily succumb to characterization by an n th-order coefficient in a power series expansion. In this situation it may be appropriate to specify a threshold optical input parameter. Another example is a homogeneously broadened saturable absorber, in which the absorption of an optical medium decreases with increasing input irradiance. The irradiance-dependent absorption coefficient α (refer to eqn [26]) is characterized by a 'saturation irradiance' I_{sat} :

$$\alpha(I) = \frac{\alpha_0}{1 + \frac{I}{I_{\text{sat}}}} \quad [9]$$

where α_0 is the linear absorption coefficient.

Nonlinear Susceptibility

The power-series expansion of the macroscopic polarization is the standard approach for modeling nonlinear optical behavior and categorizing the various phenomena. A common way to write the polarization, nonlinear in the electric field, is (mks units):

$$\mathbf{P} = \varepsilon_0 \left(\chi^{(1)} \cdot \mathbf{E} + \chi^{(2)} : \mathbf{E}\mathbf{E} + \chi^{(3)} : \mathbf{E}\mathbf{E}\mathbf{E} + \dots \right) \quad [10]$$

where the polarization \mathbf{P} is a time- and space-dependent vector and the terms $\chi^{(n)}$ are the various orders of the nonlinear susceptibility (the cgs equation is obtained by dropping the free-space permittivity coefficient ε_0). A second-order effect is associated with $\chi^{(2)}$, a third-order with $\chi^{(3)}$, and so on. In general, there are distinguishable electric field vectors (in eqn [10], the fields have not been individually designated for clarity). The susceptibility terms are generally tensors, which means the medium is sensitive to the orientation of the input fields. The dots in eqn [10] indicate tensor products. The input field vectors can have different frequencies and the presence of complex conjugates in eqn [1] indicates that the frequency components will have both + and - signs, i.e. the j th electric field term will have associated frequency factors $\exp(\pm i\omega_j t)$.

The nomenclature used here can be illustrated by example. In the case of the second-order nonlinear polarization (mks units):

$$\mathbf{P}_j = \varepsilon_0 \chi_{jkl}^{(2)} : \mathbf{E}_k \mathbf{E}_l \quad [11]$$

Note that subscripts have been introduced. The indices correspond to Cartesian space vectors.

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This equation says that the polarization in the j -direction results from the tensor product of the appropriate $\chi^{(2)}$ with the input fields at \mathbf{E}_k and \mathbf{E}_l . The j th component of polarization has an oscillation frequency that is determined by mixing of the input fields. In this example, the indices j , k and l can each take the value of x , y , and z . Let each input field be at one of two possible frequencies, call them ω_1 and ω_2 . There are an enormous number of permutations (81 to be exact) of eqn [11]. One term is:

$$P_x(\omega_1 + \omega_2) = \varepsilon_0 \chi_{xzy}^{(2)}(\omega_1 + \omega_2) E_z(\omega_1) E_y(\omega_2) \times \exp[-i(\omega_1 + \omega_2)t] \quad [12]$$

Another possibility is:

$$P_z(\omega_2 - \omega_1) = \varepsilon_0 \chi_{zyx}^{(2)}(\omega_2 - \omega_1) E_y(\omega_2) E_x(-\omega_1) \times \exp[-i(\omega_2 - \omega_1)t] \quad [13]$$

Equation [12] corresponds to sum frequency generation ($\omega_1 + \omega_2$) and eqn [13] shows difference frequency generation ($\omega_2 - \omega_1$).

Not explicitly written on the right-hand side of the above equations is the exponential containing the mixing of the propagation vectors. To maximize the nonlinear polarization, which is the source of nonlinear behavior in Maxwell's equations, the vector sum $\sum \mathbf{k}_j$ for *all* the interacting fields should be close to zero. Arranging the propagation vectors to accomplish this is known as 'phase matching'.

It is important to point out that there are eight more terms describing the nonlinear polarization at $P_x(\omega_1 + \omega_2)$ in addition to eqn [12]; likewise for $P_z(\omega_2 - \omega_1)$. Also note that the indices k and l may be identical, hence a nonlinear polarization driven by $\chi_{xyy}^{(2)}$ for example, is allowed. Higher-order nonlinear polarizations get progressively more complicated. The general third-order nonlinear polarization is

$$\mathbf{P}_j = \varepsilon_0 \chi_{jklm}^{(3)} \mathbf{E}_k \mathbf{E}_l \mathbf{E}_m \quad [14]$$

Sometimes the second-order nonlinear susceptibility is written with the coefficient d instead of $\chi^{(2)}$. The relation between the d -coefficient and $\chi^{(2)}$ depends arbitrarily on how it is defined; a common definition is

$$d_{jkl} = \frac{1}{2} \chi_{jkl}^{(2)} \quad [15]$$

but the reader is cautioned that the factor $\frac{1}{2}$ is sometimes missing. In the preceding discussion it was pointed out how there can, in principle, be a large number of tensor components involved in the

expansion of the nonlinear polarization. Simplification occurs when it is realized that there is no discernible physical difference between the frequency terms ω_j and $-\omega_j$ and that ordering of the fields in eqn [14] – which suggests a time order in the arrival of the fields – is irrelevant. These symmetry arguments show that $d_{jkl} = d_{jlk}$, which reduces the number of independent d -coefficients from 81 to 18. A simpler subscript notation is then introduced that uses the integers 1–6 to represent pairs of Cartesian components k and l . This reduces the number of subscripts from three to two. An example of this notation is $d_{xzz} = d_{14}$.

In certain situations, one can take advantage of crystal symmetry to further reduce the complicated summations to a single scalar coefficient for the nonlinear susceptibility, which is referred to as 'd-effective'. For these specialized cases, the nonlinear polarization is

$$P = \varepsilon_0 d_{\text{eff}} E_1 E_2 \quad [16]$$

In SI/mks units, the polarization is in units of coul/m². The second-order susceptibility (i.e. $\chi^{(2)}$, d_{jkl}) must therefore have units of m/volt. The units of $\chi^{(3)}$ are m²/volt², $\chi^{(4)}$ is m³/volt³, and so on. In Gaussian/cgs units, $\chi^{(2)}$ has dimensions cm/statvolt, $\chi^{(3)}$ will be cm²/statvolt², etc. although sometimes in the Gaussian system *all* the coefficients $\chi^{(n)}$ are discussed with shorthand 'electrostatic units' or 'esu'.

Complex Quantities

In linear optics, the susceptibility has real and imaginary parts

$$\chi^{(1)} = \chi_{\text{real}}^{(1)} + i\chi_{\text{imaginary}}^{(1)} \quad [17]$$

The complex linear index is, in turn, written as the sum of real and imaginary components, derived from the linear susceptibility as follows (mks units):

$$\sqrt{1 + \chi^{(1)}} = n_0 + i\kappa \quad [18]$$

where n_0 is the linear refractive index and κ is the imaginary term leading to absorption of light. In general, the nonlinear susceptibility $\chi^{(n)}$ is also a complex number:

$$\chi^{(n)} = \chi_{\text{real}}^{(n)} + i\chi_{\text{imaginary}}^{(n)} \quad [19]$$

As in linear optics, the complex notation is a bookkeeping method that conveniently accounts for what is known as 'resonant enhancement'. Nonlinear optics research has revealed that the nonlinear susceptibility $\chi^{(n)}$ can be a strong function of frequency. Specifically, this quantity will be strongly

enhanced when sums and/or differences of the photon energies in the interacting light beams coincide with quantum mechanical energy resonances in the material. When a resonance condition is achieved, $\chi^{(n)}$ will be dominated by its imaginary component. Far from the resonances, $\chi^{(n)}$ behaves more like a real quantity. These complex terms directly enter the wave equations describing how light propagates in a nonlinear medium and are particularly important for $\chi^{(3)}$; the complex quantity $\chi^{(3)}$ determines whether light will be refracted or absorbed and by how much. The real part of $\chi^{(3)}$ drives nonlinear refraction while the imaginary portion characterizes nonlinear absorption (e.g., two-photon absorption) and the inverse effect – gain. This rich subject – in particular the critical importance of the wavelength dependence of the nonlinear phenomena – is considered in the chapter by Sheik-Bahae in this encyclopedia.

written in mks units as:

$$\Delta n = n_2 I = \frac{\text{Re}(\chi^{(3)})}{4\epsilon_0 n^2 c} I \quad [23]$$

where ‘Re’ denotes the real part of $\chi^{(3)}$. In cgs units we have:

$$\Delta n = \frac{1}{2} \tilde{n}_2 |E|^2 = \frac{4\pi^2 \text{Re}(\tilde{\chi}^{(3)})}{n^2 c} I \quad [24]$$

where I is in units of ergs/(cm² sec) defined by eqn [4]. The reader is again advised that different authors will show discrepancies of 2, 4, or even 8 when writing these equations. The susceptibilities $\chi^{(3)}$ are related as follows:

$$\tilde{\chi}^{(3)}(\text{cgs}) = \frac{9 \times 10^8}{4\pi} \chi^{(3)}(\text{mks}) \quad [25]$$

If the coefficients n_0 and n_2 exist, does the nomenclature imply there are terms n_1 , n_3 , and others? These coefficients are certainly allowed, but not often seen in discussions of nonlinear refraction. When one refers to nonlinear refraction, the common understanding is that the index depends linearly on irradiance, which is conveniently modeled by eqn [20]. But there are other physically relevant situations to consider. The coefficient n_1 , for example, describes the linear electro-optic effect in which the change in index is linearly proportional to the electric field (although it is called the linear electro-optic, it is actually derived from the second-order nonlinear susceptibility). The electric field can be the oscillating field of a laser beam, for example, or a dc field applied to an electro-optic crystal (e.g., Pockel’s cell). When other terms are added to eqns [20] or [21], the units of the coefficients must be chosen to keep the equation dimensionless.

In the preceding discussion, there is an implication that the nonlinear index is an instantaneous function of the irradiance or field. Although the material system can never respond instantaneously, this is a good approximation in many situations. Sometimes it is not. Consider a pulsed laser beam that heats the material. When the local temperature increases, the linear refractive index can change. A short, Q-switched laser can have a pulse duration far less than a microsecond, while the temperature change it induces can last many orders of magnitude longer than this. This means optical modification of the refractive index may persist long after the exciting pulse has vanished, i.e., when the irradiance is at zero.

Another example is when a laser beam promotes electrons from their ground state to higher energy

S0025 Nonlinear Refraction

P0105 The most common manifestation of nonlinear refraction arises from the third-order nonlinear susceptibility, the so-called optical Kerr effect. In this case, the refractive index is linearly proportional to the irradiance (I) of a monochromatic light beam. The irradiance-dependent refractive index is

$$n(I) = n_0 + n_2 I \quad [20]$$

where n_0 is the linear, irradiance-independent refractive index and n_2 is the nonlinear refractive index coefficient. Because n is a dimensionless quantity, n_2 must be in units of area/power.

P0110 The optical Kerr effect is usually written in mks units with the expression shown in eqn [20]. In cgs units, the common (but by no means universal) convention is to write:

$$n(E) = n_0 + \frac{1}{2} \tilde{n}_2 |E|^2 \quad [21]$$

where the field is in units of statvolts/cm. The nonlinear coefficient \tilde{n}_2 (cgs) must therefore be in units of cm²/statvolt², which is sometimes abbreviated to ‘esu’. The conversion for n_2 between these two equations is:

$$\tilde{n}_2(\text{cgs}) = \frac{n_0 c}{40\pi} n_2(\text{mks}) \quad [22]$$

where $n_2(\text{mks})$ is in units of m²/W and c is in m/sec. Also useful is the relation between n_2 and $\chi^{(3)}$,

states of the material system. These excited electrons may modify the refractive index of the material. In general, the excited electrons remain in high-energy states for some period of time before relaxing to the ground level – if this time is longer than the excitation, the change of the refractive index persists beyond the time the laser beam is present. In these situations, the model of nonlinear refraction suggested by the above equations is inaccurate. One cannot obtain the nonlinear refraction from a simple algebraic analysis. A system of time-dependent equations – describing the dynamics of excitation and relaxation – must be solved using numerical procedures. Such phenomena are sometimes loosely categorized as ‘dynamic nonlinear optical effects’ or ‘effective third-order nonlinearities’. Refer to the chapter(s) by ??? in this encyclopedia.

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S0030 Nonlinear Absorption

P0125 Consider a single-frequency light beam passing through an optically absorbing region of length L . For simplicity, neglect reflections caused by surfaces that may define the region of interest, i.e., ignore reflections at surfaces that may be located on the optical axis z at points $z = 0$, $z = L$, or any other point in the path. Linear absorption means that the optical power extracted from the light beam as it traverses the absorbing medium is a direct function of the power at a given point. This is described mathematically by an elementary, linear differential equation known as Beer’s law:

$$\frac{d}{dz}I(z) = -\alpha I(z) \quad [26]$$

The constant of proportionality is α , which is the linear absorption coefficient. Equation [23] is solved by direct integration:

$$\int_{I(0)}^{I(L)} \frac{dI}{I} = -\alpha \int_0^L dz \quad [27]$$

This has the solution:

$$\frac{I(L)}{I(0)} = \exp(-\alpha L) \quad [28]$$

which means the irradiance decreases exponentially as a function of propagation distance in a linearly absorbing medium.

P0130 In the nonlinear regime, we don’t expect a classical Beer’s law model to hold. By definition, the absorption will be a nonlinear function of irradiance at a given point. One makes the following power-series expansion to describe nonlinear absorption of

monochromatic light:

$$\frac{d}{dz}I = -\alpha I - \beta I^2 - \gamma I^3 - \dots \quad [29]$$

The coefficient β corresponds to a two-photon absorption process and γ is the coefficient of three-photon absorption. What about four-photon and even higher-order processes? These are rarely encountered, but certainly possible. To make these distinctions, eqn [29] is sometimes written with coefficients K_i or α_i instead of the sequential Greek alphabet:

$$\frac{d}{dz}I = -K_1 I - K_2 I^2 - K_3 I^3 - K_4 I^4 - \dots \quad [30]$$

The units of the various absorption coefficients must maintain the dimensional consistency of eqns [29] and [30], which is irradiance/length or power/(length)³. These are shown in Table 1.

If there is linear absorption, the nonlinear processes at that wavelength are often (but not always) negligibly weak. This means we have the following inequalities:

$$K_1 I \gg K_i I^i, \quad \text{where } i \geq 2 \quad [31]$$

Nonlinear absorption is generally observed in the wavelength regime where the medium is transparent to low-irradiance light, i.e., where linear absorption is negligible. It should be emphasized that there are situations where linear absorption is large and in fact a crucial component of the aggregate nonlinear effect. We will return to this point later in the discussion. For the moment, we neglect linear absorption. If the energy of the incident photons and energy levels of the system permit it, the lowest-order nonlinear process is two-photon absorption, described by the equation

$$\frac{d}{dz}I = -K_2 I^2 \quad [32]$$

which can be solved by elementary integration:

$$\frac{I(L)}{I(0)} = \frac{1}{1 + I(0)K_2 L} \quad [33]$$

Unfortunately, the situation is rarely this convenient. The above integration has ignored the fact

Table 1 Dimensions of optical absorption coefficients

Process	Coefficient	Units
Linear absorption	α, K_1	(length) ⁻¹
Two-photon absorption	β, K_2	length/power
Three-photon absorption	γ, K_3	length ³ /power ²
Four-photon absorption	K_4	length ⁵ /power ³

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that nonlinear absorption may enable significant linear absorption. This is possible because nonlinear absorption promotes electrons from low- to high-energy states in the medium. The change in excited electron density associated with absorption of light (both linearly and nonlinearly) is called ‘photocarrier generation’. These photocarriers (e.g., photo-electrons) may modify the linear absorption as well as the refractive properties of the material. If two-photon absorption causes the linear absorption to increase, for example, the assumption that allowed us to ignore K_1 when writing eqn [32] ceases to be valid. While eqn [32] was appropriate at the very start of the light–matter interaction, the generation of photocarriers after the passage of time may change that condition. The behavior of the system is therefore time-dependent, i.e., it is dynamic. Simple analytic solutions are almost always not possible when dealing with nonlinear absorption.

P0140 The time rate of change of the photocarrier population (N) is modeled by the following equation:

$$\frac{\partial}{\partial t} N = \frac{K_1 I}{\hbar \omega} + \frac{K_2 I^2}{2\hbar \omega} + \frac{K_3 I^3}{3\hbar \omega} + \frac{K_4 I^4}{4\hbar \omega} + \dots - \text{recombination} - \text{diffusion} \quad [34]$$

where ω is the angular frequency ($\omega = 2\pi f$) of the incident light. Also included are place-holders for loss processes such as recombination and diffusion, which may themselves have complicated mathematical descriptions. The coefficients K_i have exactly the same units, dimensions, and interpretation as in eqn [30] and [Table 1](#); in the spirit of the preceding discussion, these coefficients may be time dependent. Note that the dimensions of eqn [30] describe absorption of light (irradiance/length), while eqn [34] models the photocarrier population density (length⁻³ time⁻¹). The units of these two equations are very different.

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List of Units and Nomenclature

Acronym	Full description
CARS	Coherent anti-Stokes Raman scattering
c.c.	Complex conjugate
CSRS	Coherent Stokes Raman scattering
DFG	Difference frequency generation
DFM	Difference frequency mixing
DFWM, FWM	(Degenerate) Four wave mixing

DRO	Doubly resonant OPO
EIT	Electromagnetically induced transparency
ESA	Excited state absorption
GVD	Group velocity dispersion
GVM	Group velocity mismatch
NLA	Nonlinear absorption
NLR	Nonlinear refraction
OFID	Optical free-induction decay
OPA	Optical parametric amplification/amplifier
OPG	Optical parametric generation/generator
OPL	Optical power limiter
OPO	Optical parametric oscillation/oscillator
2PA, TPA	Two-photon absorption
PCM	Phase conjugate mirror
QPM	Quasi phase matching
RIKES	Raman-induced Kerr effect spectroscopy
RSA	Reverse saturable absorption
SBS	Stimulated Brillouin scattering
SFG	Sum frequency generation
SHG	Second harmonic generation
SIT	Self-induced transparency
SRO	Singly resonant OPO
SRS	Stimulated Raman scattering
SRWS	Stimulated Rayleigh wing scattering
SVEA, SVAA	Slowly varying envelope (amplitude) approximation
THG	Third harmonic generation
TPF	Two-photon fluorescence

See also

Nonlinear Optics: (00700). **Nonlinear Optics Basics:** Ultrafast and Intense-Field Nonlinear Optics (00743). **Laser in Physics:** Nonlinear Laser Spectroscopy (00897).

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Further Reading

- Bloembergen N (1992) *Nonlinear Optics*. Redwood City, CA: Addison-Wesley.
- Boyd RW (1991) *Nonlinear Optics*. Boston: Academic Press.
- Levenson MD and Kano S (1988) *Introduction to Nonlinear Laser Spectroscopy*. Boston: Academic Press.
- Miller A, Miller DAB and Smith SD (1981) Dynamic nonlinear optical processes in semiconductors. *Advances in Physics* 30: 697–800.
- Mills DL (1998) *Nonlinear Optics: Basic Concepts*. New York: Springer.
- Newell C and Moloney JV (1992) *Nonlinear Optics*. Redwood City, CA: Addison-Wesley.

Sauter EG (1996) *Nonlinear Optics*. New York: Wiley.

Shen YR (1984) *The Principles of Nonlinear Optics*.
New York: Wiley.

Sutherland RL (1996) *Handbook of Nonlinear Optics*.
New York: Marcel Dekker.

Yariv A (1991) *Optical Electronics*, 3rd edn. New York:
Holt, Rinehart, and Winston.

Yariv A (1989) *Quantum Electronics*. New York: Wiley.

Zernike F and Midwinter JE (1973) *Applied Nonlinear
Optics*. New York: Wiley.

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