Springer Handbook of
Atomic, Molecular, and Optical Physics

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CD-ROM

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Springer Handbooks of Atomic, Molecular, and Optical Physics

Springer Handbook
of Atomic, Molecular, and Optical Physics

Gordon W. F. Drake (Ed.)

With CD-ROM, 288 Figures and 111 Tables

Springer

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Library of Congress Control Number: 2005931256

ISBN-10: 0-387-20802-X e-ISBN: 0-387-26308-X
ISBN-13: 978-0-387-20802-2 Printed on acid free paper

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Printed in Germany.

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Production and typesetting: LE-TeX GbR, Leipzig
Handbook coordinator: Dr. W. Skolaut, Heidelberg
Typography, layout and illustrations: schreiberVIS, Seeheim
Cover design: eStudio Calamar Steinen, Barcelona
Cover production: *design&production* GmbH, Heidelberg
Printing and binding: Stürtz GmbH, Würzburg

SPIN 10948934 100/3141/YL 5 4 3 2 1 0

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List of Abbreviations

2P/2H	two-particle/two-hole

A

AA	average atom
ACT	activated complex theory
ADDS	angular distribution by Doppler spectroscopy
ADO	average dipole orientation
AES	Auger electron spectroscopy
AI	adiabatic ionization
AL	absorption loss
ALS	advanced light source
AMO	atomic, molecular, and optical
ANDC	arbitrarily normalized decay curve
AO	atomic orbital
AOM	acoustooptic modulator
AS	active space
ASD	atomic spectra database
ASF	atomic state functions
ATI	above threshold ionization
AU	absorbance units

B

BEA	binary encounter approximation
BEC	Bose–Einstein condensate (or condensation)
BF	body-fixed
BI	Bell’s inequality
BL	Bethe log
BO	Born–Oppenheimer
BS	Bethe–Salpeter
BW	Brillouin–Wigner

C

CARS	coherent anti-Stokes Raman scattering
CAS	complete active space
CASPT	complete active space perturbation theory
CAUGA	Clifford algebra unitary group approach
CC	coupled cluster
CCA	coupled cluster approximation
CCC	convergent close coupling
CCD	coupled cluster doubles
CCO	coupled-channels optical
CDW	continuum distorted wave
CEAS	cavity enhanced absorption spectroscopy
CES	cavity enhanced spectroscopy
CES	constant energy surface
CETS	cavity enhanced transmission spectroscopy
CFCP	free–free molecular Franck–Condon
CG	Clebsch–Gordan
CH	Clauser–Horne
CI	configuration interaction
CIS	constant ionic state
CL	constant log
CM	center-of-mass
CMA	cylindrical mirror analyzer
COA	classical oscillator approximation
CODATA	Committee on Data for Science and Technology
COIL	chemical-oxygen-iodine
COLTRIMS	cold-target recoil-ion momentum spectroscopy
CP	central potential
CPA	chirped-pulsed-amplification
CQC	classical-quantal coupling
CRDS	cavity ring-down spectroscopy
CSDA	continuous slowing down approximation
CSF	configurational state functions
CTF	common translation factor
CTMC	classical trajectory Monte Carlo
CW	continuous wave
CW-CRDS	continuous-Wave Cavity Ring-Down Spectroscopy
CX	charge exchange
CXO	Chandra X-ray Observatory

D

DB	detailed balance
DCS	differential cross sections
DDCS	doubly differential cross sections
DF	Dirac–Fock
DFB	distributed feedback
DFS	decoherence free subspace
DFT	discrete Fourier transform
DFWM	degenerate four wave mixing
DLR	dielectronic recombination
DODS	different orbitals for different spins
DR	dielectronic recombination
DSPB	distorted wave strong potential Born approximation

E

EA	excitation-autoionization
EBIT	electron beam ion traps
EBS	eikonal Born series

ECP	effective core potential
ECS	exterior complex scaling
EEDF	electron energy distribution functions
EOM	equation of motion
EPR	Einstein–Podolsky–Rosen
ESM	elastic scattering model
ESR	experimental storage ring
EUV	extreme ultraviolet
EW-CRDS	evanescent-wave CRDS
EXAFS	extended X-ray absorption fine structure

F

FBA	first Born approximation
FCPC	full-core plus correlation
FEL	free-electron lasers
FFT	fast Fourier transform
FID	free induction decay
FIR	far-infrared
FM	frequency modulation
FOTOS	first-order theory for oscillator strengths
FS	fine-structure
FT	Fourier transform
FTIR	Fourier transform infrared spectroscopy
FTMS	Fourier transform mass spectrometry
FTS	Fourier transform spectroscopy
FUSE	far ultraviolet spectroscopic explorer
FUV	far ultraviolet
FVP	Fermi virtual photon
FWHM	full width at half maximum

G

GBT	generalized Brillouin's Theorem
GFA	Green's function approach
GGA	generalized gradient approximation
GHZ	Greenberger, Horne, Zeilinger
GI	gauge invariant
GIB	guided ion beam
GOME	global ozone monitoring experiment
GOS	generalized oscillator strength
GPE	Gross–Pitaevskii equation
GRPAE	generalized random phase approximationwith exchange

H

HEDP	high energy-density physics
HF	Hartree–Fock equations
HF	Hellman–Feynman
HG	harmonic generation
HOM	Hong–Ou–Mandel
HREELS	high resolution electron energy loss spectroscopy
HRTOF	H-atom Rydberg time-of-flight
HUM	Hylleraas–Undheim–MacDonald theorem

I

IC	intermediate coupling
ICF	inertial confinement fusion
ICOS	integrated cavity output spectroscopy
ICSLS	international conference on spectral line shapes
IERM	intermediate energy R-matrix
IPCC	intergovernmental panel on climate change
IPES	inverse photoemission spectroscopy
IPIR	independent-processes and isolated-resonance
IPM	independent particle model
IPP	impact parameter picture
IR	irreducible representations
IR	infrared
IRI	international reference ionosphere
IRREP	irreducable representation
ISO	infrared space observatory

J

JB	Jeffrey–Born

K

KS	Kohn–Sham
KTA	potassium titanyl arsenate
KTP	potassium titanyl phosphate

L

LieA	Lie algebras
LA	linear algebraic
L-CETS	locked cavity enhanced transmission spectroscopy
LDA	local density approximation
LEED	low energy electron diffraction
LER	laser electric resonance
LG	Lie groups
LHC	left-hand circular
LHV	local hidden variable
LIF	laser-induced-fluorescence
LIGO	laser interferometer gravitational-wave observatory
LISA	laser interferometer space antenna
LL	Landau–Lifshitz
LM	Levenberg–Marquardt
LMR	laser magnetic resonance
LPT	laser photodetachment threshold
LRL	Laplace–Runge–Lenz
LTE	local thermodynamic equilibrium
LYP	Lee, Yang, and Parr
LZ	Landau–Zener

M

MBE	molecular beam epitaxy
MCP	microchannel plate
MDAL	minimum detectable absorption loss
MBPT	many-body perturbation theory
MCDHF	multiconfigurational Dirac–Hartree–Fock
MCHF	multiconfiguration Hartree–Fock
MCSCF	multiconfigurational self-consistent field
MEMS	microelectromechanical systems
MFP	mean free path
MIGO	matter–wave interferometric gravitational-wave observatory
MIM	metal-insulator-metal
MKSA	meters, kilograms, seconds, and amperes
MM	Massey–Mohr
MMCDF	multichannel multiconfiguration Dirac–Fock
MO	molecular orbital
MOPA	master oscillator power amplifier
MOT	magneto-optical trap
MOX	molecular orbital X-radiation
MP2	second order Møller–Plesset perturbation theory
MP3	third order Møller–Plesset perturbation theory
MPI	multiphoton ionization
MQDT	multichannel quantum defect theory
MR	multireference
MR-SDCI	multireference singles/doubles configuration interaction
MUV	middle ultraviolet

N

NAR	nonadiabatic region
NEP	noise-equivalent power
NEXAFS	near-edge X-ray absorption fine structure
NDIR	non-dispersive infrared
NFS	nonfine-structure
NICE-OHMS	noise-immune, cavity-enhanced optical heterodyne molecular spectroscopy
NIM	normal incidence monochromator
NIST	National Institute of Standards and Technology
NMR	nuclear magnetic resonance systems
NNS	nearest-neighbor energy level spacings
NR	nonrelativistic
NRQED	NR quantum electrodynamics

O

OAO-2	orbiting astronomical observatory
OB	ordinary Bremsstrahlung
OBE	optical Bloch equations
OBK	Oppenheimer–Brinkman–Kramers
OCP	one-component plasma
OHCE	one-and-a-half centered expansion
OMI	ozone monitoring instrument
OPO	optical parametric oscillator

P

P-CRDS	pulsed-cavity ringdown spectroscopy
PADDS	angular distribution by Doppler spectroscopy
PAH	polycyclic aromatic hydrocarbon
PBS	polarizing beam splitters
PCDW	projectile continuum distorted wave approximation
PDM	phase diffusion model
PEC	potential energy curves
PES	photoelectron spectroscopy
PES	potential energy surface
PH/HP	particle–hole/hole–particle
PI	photoionization
PID	particle identification
PIMC	path-integral Monte Carlo
PMT	photomultiplier tubes
PNC	parity nonconservation
PPT	positive partial transposes
PR	polarization radiation
PSD	postion senitive detectors
PSS	perturbed stationary state
PT	perturbation theory
PWBA	plane wave Born approximation
PZT	piezo-electric transducer

Q

QCD	quantum chromodynamics
QED	quantum electrodynamics
QIP	quantum information processing
QKD	quantum key distribution
QMC	quantum Monte Carlo
QND	quantum nondemolition
QS	quasistatic
QSS	quasi-steady state

R

RATIP	relativistic atomic transition and ionization properties
RDC	ring-down cavity
READI	resonant excitation auto-double ionization
REC	radiative electron capture
REDA	resonant excitation double autoionization
REMPI	resonance-enhanced multiphoton ionization
RES	rotational energy surface
RHC	right-hand circular
RHIC	relativistic heavy ion collider

RIMS	recoil-ion momentum spectroscopy
RMI	relativistic mass increase
RMPS	*R*-matrix with pseudostates
RNA	Raman–Nath approximation
RPA	random-phase approximation
RPA	retarding potential analyzer
RPAE	random phase approximation with exchange
RR	radiative recombination
RRKM	Rampsberger–Rice–Karplus–Marcus
RSE	radial Schrödinger equation
RSPT	Rayleigh–Schrödinger perturbation theory
RT	Ramsauer–Townsend
RTE	resonant transfer and excitation
RWA	rotating wave approximation

S

SA-MCSCF	state averaged multiconfiguration self-consistent field
SACM	statistical adiabatic channel model
SBS	stimulated Brillouin scattering
SCA	semiclassical approximation
SCF	self-consistent field
SCIAMACHY	scanning imaging absorption spectrometer for atmospheric chartography
SD	spin-dependent
SD	single and double
SDS	singly differential cross section
SDTQ	single, double, triple, quadruple
SE	Schrödinger equation
SEP	stimulated emission pump
SEPE	simultaneous electron photon excitation
SEXAFS	surface extended X-ray absorption fine structure
SF	space-fixed
SI	spin-independent
SIAM	Society for Industrial and Applied Mathematics
SM	submillimeter
SM-FIR	submillimeter far-infrared
SMS	specific mass shift
SOHO	solar and heliospheric observatory
SP	stationary phase
SPA	stationary phase approximations
SQL	standard quantum limit
SQUID	superconducting quantum interference detector
SR	synchrotron radiation
SRS	stimulated Raman scattering
SS	strong-short
STIRAP	stimulated Raman adiabatic passage
STO	Slater type orbital
STP	standard temperature and pressure

T

TCDW	target continuum distorted wave
TDCS	triply differential cross section
TDHF	time-dependent Hartree–Fock
TDS	thermal desorption spectroscopy
TDSE	time dependent Schrödinger equation
TEA	transverse-excitation-atmospheric-pressure
TF	toroidal field
TOF	time-of-flight
TOP	time orbiting potential
TPA	two-photon absorption
TSR	test storage ring
TuFIR	tunable far-infrared

U

UGA	unitary group approach
UHF	unrestricted Hartree–Fock
UPS	ultraviolet photoelectron spectroscopy
UV	ultraviolet
UV-VIS	ultraviolet-visible

V

VASP	Vienna ab-initio simulation package
VCSEL	vertical-cavity surface-emitting laser
VECSEL	vertical external cavity surface-emitting laser
VES	vibrational energy surfaces
VUV	vacuum ultraviolet

W

WCG	Wigner–Clebsch–Gordan
WDM	warm dense matter
WKB	Wentzel, Kramers, Brillouin
WL	weak-long
WMAP	Wilkinson microwave anisotropy probe
WPMD	wavepacket molecular dynamics

X

XPS	X-ray photoelectron spectroscopy

Y

YAG	Yttrium Aluminum Garnet

Nonlinear Opt

72. Nonlinear Optics

Nonlinear optics is concerned with the propagation of intense beams of light through a material system. The optical properties of the medium can be modified by the intense light beam, leading to new processes that would not occur in a material that responded linearly to an applied optical field. These processes can lead to the modification of the spectral, spatial, or polarization properties of the light beam, or the creation of new frequency components. More complete accounts of nonlinear optics including the origin of optical nonlinearities can be found in references [72.1–4].

Both the Gaussian and MKS system of units are commonly used in nonlinear optics. Thus, we have chosen to express the equations in this chapter in both the Gaussian and MKS systems. Each equation can be interpreted in the MKS system as written or in the Gaussian system by omitting the prefactors (e.g., $1/4\pi\varepsilon_0$) that appear in square brackets at the beginning of the expression on the right-hand-side of the equation.

72.1 Nonlinear Susceptibility

In linear optics it is customary to describe the response of a material in terms of a macroscopic polarization $\tilde{\boldsymbol{P}}$ (i.e., dipole moment per unit volume) which is linearly related to the applied electric field $\tilde{\boldsymbol{E}}$ through the linear susceptibility $\chi^{(1)}$. In order to extend the relationship between $\tilde{\boldsymbol{P}}$ and $\tilde{\boldsymbol{E}}$ into the nonlinear regime, the polarization is expanded in a power series of the electric field strength. We express this relationship mathematically by first decomposing the field and the polarization into their frequency components such that

$$\tilde{\boldsymbol{E}}(\boldsymbol{r},t)=\sum_l \boldsymbol{E}(\boldsymbol{r},\omega_l)\,\mathrm{e}^{-\mathrm{i}\omega_l t}\,, \tag{72.1}$$

$$\tilde{\boldsymbol{P}}(\boldsymbol{r},t)=\sum_l \boldsymbol{P}(\boldsymbol{r},\omega_l)\,\mathrm{e}^{-\mathrm{i}\omega_l t}\,, \tag{72.2}$$

where the summations are performed over both positive and negative frequencies. The reality of $\tilde{\boldsymbol{E}}$ and $\tilde{\boldsymbol{P}}$ is then assured by requiring that $\boldsymbol{E}(\boldsymbol{r},\omega_l)=\boldsymbol{E}^*(\boldsymbol{r},-\omega_l)$ and $\boldsymbol{P}(\boldsymbol{r},\omega_l)=\boldsymbol{P}^*(\boldsymbol{r},-\omega_l)$. In this case the general expression for the Cartesian component i of the polarization at frequency ω_σ is given by

$$\begin{aligned}P_i(\omega_\sigma)=[\varepsilon_0]\Big[&\sum_j \chi_{ij}^{(1)}(\omega_\sigma)E_j(\omega_\sigma)\\&+\sum_{jk}\sum_{(mn)}\chi_{ijk}^{(2)}(\omega_\sigma;\omega_m,\omega_n)\\&\times E_j(\omega_m)E_k(\omega_n)\\&+\sum_{jkl}\sum_{(mno)}\chi_{ijkl}^{(3)}(\omega_\sigma;\omega_m,\omega_n,\omega_o)E_j(\omega_m)\\&\times E_k(\omega_n)E_l(\omega_o)\\&+\cdots\Big]\,,\end{aligned}\tag{72.3}$$

where $ijkl$ refer to field components, and the notation (mn), for example, indicates that the summation over n and m should be performed such that $\omega_\sigma=\omega_m+\omega_n$ is held constant. Inspection of (72.3) shows that the $\chi^{(n)}$ can be required to satisfy intrinsic permutation symmetry, i. e., the Cartesian components and the corresponding frequency components [e.g., (j,ω_j) but not (i,ω_σ)] associated with the applied fields may be permuted without changing the value of the susceptibility. For example, for the second-order susceptibility,

$$\chi_{ijk}^{(2)}(\omega_\sigma;\omega_m,\omega_n)=\chi_{ikj}^{(2)}(\omega_\sigma;\omega_n,\omega_m)\,.\tag{72.4}$$

If the medium is lossless at all the field frequencies taking part in the nonlinear interaction, then the condition of full permutation symmetry is necessarily valid. This condition states that the pair of indices associated with the Cartesian component and the frequency of the nonlinear polarization [i. e., (i,ω_σ)] may be permuted along with the pairs associated with the applied field components. For example, for the second-order susceptibility, this condition implies that

$$\chi_{ijk}^{(2)}(\omega_\sigma;\omega_m,\omega_n)=\chi_{kji}^{(2)}(-\omega_n;\omega_m,-\omega_\sigma)\,.\tag{72.5}$$

If full permutation symmetry holds, and in addition all the frequencies of interest are well below any of the transition frequencies of the medium, the $\chi^{(n)}$ are invariant upon free permutation of all the Cartesian indices. This condition is known as the Kleinman symmetry condition.

72.1.1 Tensor Properties

The spatial symmetry properties of a material can be used to predict the tensor nature of the nonlinear susceptibility. For example, for a material that possesses inversion symmetry, all the elements of the even-ordered susceptibilities must vanish (i. e., $\chi^{(n)}=0$ for n even). The number of independent elements of the nonlinear susceptibility for many materials can be substantially fewer than than the total number of elements. For example, in general $\chi^{(3)}$ consists of 81 elements, but for the case of isotropic media such as gases, liquids, and glasses, only 21 elements are nonvanishing and only three of these are independent. The nonvanishing elements consist of the following types: $\chi_{iijj}^{(3)}$, $\chi_{ijij}^{(3)}$, and $\chi_{ijji}^{(3)}$, where $i\neq j$. In addition, it can be shown that

$$\chi_{iiii}^{(3)}=\chi_{iijj}^{(3)}+\chi_{ijij}^{(3)}+\chi_{ijji}^{(3)}\,.\tag{72.6}$$

72.1.2 Nonlinear Refractive Index

For many materials, the refractive index n is intensity-dependent such that

$$n=n_0+n_2I\,,\tag{72.7}$$

where n_0 is the linear refractive index, n_2 is the nonlinear refractive index coefficient , and $I=[4\pi\varepsilon_0]n_0c|E|^2/2\pi$ is the intensity of the optical field. For the case of a single, linearly polarized light beam traveling in an isotropic medium or along a crystal axis of a cubic material, n_2 is related to $\chi^{(3)}$ by

$$n_2=\left(\frac{1}{16\pi^2\varepsilon_0}\right)\frac{12\pi^2}{n_0^2c}\chi_{iiii}^{(3)}(\omega;\omega,\omega,\omega,-\omega)\,.\tag{72.8}$$

For the common situation in which n_2 is measured in units of $\mathrm{cm^2/W}$ and $\chi^{(3)}$ is measured in Gaussian units, the relation becomes

$$n_2\left(\frac{\mathrm{cm}^2}{\mathrm{W}}\right)=\frac{12\pi^2\times10^7}{n_0^2c}\chi_{iiii}^{(3)}(\omega;\omega,\omega,\omega,-\omega)\,.\tag{72.9}$$

There are various physical mechanisms that can give rise to a nonlinear refractive index. For the case of induced molecular orientation in $\mathrm{CS_2}$, $n_2=3\times10^{-14}\,\mathrm{cm^2/W}$. If the contribution to the nonlinear refractive index is electronic in nature (e.g., glass), then $n_2\approx2\times10^{-16}\,\mathrm{cm^2/W}$.

72.1.3 Quantum Mechanical Expression for $\chi^{(n)}$

The general quantum mechanical perturbation expression for the $\chi^{(n)}$ in the nonresonant limit is (Under conditions of resonant excitation, relaxation phenomena must be included in the treatment, and the density matrix formalism must be used [72.4]. The resulting equation for the nonlinear susceptibility is then more complicated)

$$\begin{aligned}&\chi^{(n)}_{i_0\cdots i_n}(\omega_\sigma;\omega_1,\dots,\omega_n)\\&=\left[\frac{1}{\varepsilon_0}\right]\frac{N}{\hbar^n}\mathcal{P}_F\sum_{ga_1\cdots a_n}\rho_0(g)\\&\quad\times\frac{1}{(\omega_{a_1g}-\omega_1-\cdots-\omega_n)}\\&\quad\times\frac{\mu^{i_0}_{ga_1}\mu^{i_1}_{a_1a_2}\cdots\mu^{i_{n-1}}_{a_{n-1}a_n}\mu^{i_n}_{a_ng}}{(\omega_{a_2g}-\omega_2-\cdots-\omega_n)\cdots(\omega_{a_ng}-\omega_n)}\end{aligned}\qquad(72.10)$$

where $\omega_\sigma=\omega_1+\cdots+\omega_n$, N is the density of atoms or molecules that compose the material, $\rho_0(g)$ is the probability that the atomic or molecular population is initially in the state g in thermal equilibrium, $\mu^{i_1}_{a_1a_2}$ is the i_1th Cartesian component of the (a_1a_2) dipole matrix element, ω_{a_1g} is the transition frequency between the states a_1 and g, and $\mathcal{P}_F$ is the full permutation operator which is defined such that the expression that follows it is to be summed over all permutations of the pairs (i_0,ω_σ), $(i_1,\omega_1)\cdots(i_n,\omega_n)$ and divided by the number of permutations of the input frequencies. Thus the full expression for $\chi^{(2)}$ consists of six terms and that for $\chi^{(3)}$ consists of 24 terms.

In the limit in which the frequencies of all the fields are much smaller than any resonance frequency of the medium, the value of $\chi^{(n)}$ can be estimated to be

$$\chi^{(n)}\approxeq\left[\frac{1}{\varepsilon_0}\right]\left(\frac{2\mu}{\hbar\omega_0}\right)^n N\mu,\qquad(72.11)$$

where μ is a typical value for the dipole moment and ω_0 is a typical value of the transition frequency between the ground state and the lowest-lying excited state. For the case of $\chi^{(3)}$ in Gaussian units, the predicted value is $\chi^{(3)}=3\times10^{-14}$, which is consistent with the measured values of many materials (e.g., glass) in which the nonresonant electronic nonlinearity is the dominant contribution.

72.1.4 The Hyperpolarizability

The nonlinear susceptibility relates the macroscopic polarization $\boldsymbol{P}$ to the electric field strength $\boldsymbol{E}$. A related microscopic quantity is the hyperpolarizability, which relates the dipole moment $\boldsymbol{p}$ induced in a given atom or molecule to the electric field $\boldsymbol{E}^{\text{loc}}$ (the Lorentz local field) that acts on that atom or molecule. The relationship between $\boldsymbol{p}$ and $\boldsymbol{E}^{\text{loc}}$ is

$$\begin{aligned}&p_i(\omega_\sigma)\\&=[\varepsilon_0]\Bigg[\sum_j\alpha_{ij}(\omega_\sigma)E^{\text{loc}}_j(\omega_\sigma)\\&\quad+\sum_{jk}\sum_{(mn)}\beta_{ijk}(\omega_\sigma;\omega_m,\omega_n)E^{\text{loc}}_j(\omega_m)E^{\text{loc}}_k(\omega_n)\\&\quad+\sum_{jkl}\sum_{(mno)}\gamma_{ijkl}(\omega_\sigma;\omega_m,\omega_n,\omega_o)\\&\quad\times E^{\text{loc}}_j(\omega_m)E^{\text{loc}}_k(\omega_n)E^{\text{loc}}_l(\omega_o)+\cdots\Bigg],\end{aligned}\qquad(72.12)$$

where α_{ij} is the linear polarizability, β_{ijk} is the first hyperpolarizability, and γ_{ijkl} is the second hyperpolarizability. The nonlinear susceptibilities and hyperpolarizabilities are related by the number density of molecules N and by local-field factors, which account for the fact that the field $\boldsymbol{E}^{\text{loc}}$ that acts on a typical molecule is not in general equal to the macroscopic field $\boldsymbol{E}$. Under many circumstances, it is adequate to relate $\boldsymbol{E}^{\text{loc}}$ to $\boldsymbol{E}$ through use of the Lorentz approximation

$$\boldsymbol{E}^{\text{loc}}(\omega)=\boldsymbol{E}(\omega)+\left[\frac{1}{4\pi\varepsilon_0}\right]\frac{4\pi}{3}\boldsymbol{P}(\omega)\,.\qquad(72.13)$$

To a good approximation, one often needs to include only the linear contribution to $\boldsymbol{P}(\omega)$, and thus the local electric field becomes

$$\boldsymbol{E}^{\text{loc}}(\omega)=\mathcal{L}(\omega)\boldsymbol{E}(\omega)\,,\qquad(72.14)$$

where $\mathcal{L}(\omega)=\left\{\left[\varepsilon_0^{-1}\right]\varepsilon(\omega)+2\right\}/3$ is the local field correction factor and $\varepsilon(\omega)$ is the linear dielectric constant. Since $\boldsymbol{P}(\omega)=N\boldsymbol{p}(\omega)$, (72.3) and (72.12) through (72.14) relate the $\chi^{(n)}$ to the hyperpolarizabilities through

$$\chi^{(1)}_{ij}(\omega_\sigma)=\mathcal{L}(\omega_\sigma)N\alpha_{ij}(\omega_\sigma)\,,\qquad(72.15)$$

$$\begin{aligned}\chi^{(2)}_{ijk}(\omega_\sigma;\omega_m,\omega_n)&=\mathcal{L}(\omega_\sigma)\mathcal{L}(\omega_m)\mathcal{L}(\omega_n)\\&\quad\times N\beta_{ijk}(\omega_\sigma;\omega_m,\omega_n)\,,\end{aligned}\qquad(72.16)$$

$$\begin{aligned}\chi^{(3)}_{ijk}(\omega_\sigma;\omega_m,\omega_n,\omega_o)&=\mathcal{L}(\omega_\sigma)\mathcal{L}(\omega_m)\mathcal{L}(\omega_n)\mathcal{L}(\omega_o)\\&\quad\times N\gamma_{ijkl}(\omega_\sigma;\omega_m,\omega_n,\omega_o)\,.\end{aligned}\qquad(72.17)$$

For simplicity, the analysis above ignores the vector character of the interacting fields in calculating $\mathcal{L}(\omega)$. A generalization that does include these effects is given in [72.5].

72.2 Wave Equation in Nonlinear Optics

72.2.1 Coupled-Amplitude Equations

The propagation of light waves through a nonlinear medium is described by the wave equation

$$\nabla^2\tilde{\boldsymbol{E}}-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\tilde{\boldsymbol{E}}=\left[\frac{1}{4\pi\varepsilon_0}\right]\frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\tilde{\boldsymbol{P}}\,. \tag{72.18}$$

For the case in which $\tilde{\boldsymbol{E}}$ and $\tilde{\boldsymbol{P}}$ are given by (72.1), the field amplitudes associated with each frequency component can be decomposed into their plane wave components such that

$$\begin{aligned}\boldsymbol{E}(\boldsymbol{r},\omega_l)&=\sum_l \boldsymbol{A}_n(\boldsymbol{r},\omega_l)\,\mathrm{e}^{\mathrm{i}\boldsymbol{k}_n\cdot\boldsymbol{r}}\,,\\ \boldsymbol{P}(\boldsymbol{r},\omega_l)&=\sum_l \boldsymbol{P}_n(\boldsymbol{r},\omega_l)\,\mathrm{e}^{\mathrm{i}\boldsymbol{k}_n\cdot\boldsymbol{r}}\,,\end{aligned} \tag{72.19}$$

where $k_n=n(\omega_l)\omega_l/c$ is the magnitude of the wavevector $\boldsymbol{k}_n$. The amplitudes $\boldsymbol{A}_n$ and $\boldsymbol{P}_n$ are next decomposed into vector components whose linear optical properties are such that the polarization associated with them does not change as the field propagates through the material. For example, for a uniaxial crystal these eigenpolarizations could correspond to the ordinary and extraordinary components. In order to describe the propagation and the nonlinear coupling of these eigenpolarizations, the vector field amplitudes are expressed as

$$\begin{aligned}\boldsymbol{A}_n(\boldsymbol{r},\omega_l)&=\hat{\boldsymbol{u}}_{ln}A_n(\boldsymbol{r},\omega_l)\,,\\ \boldsymbol{P}_n(\boldsymbol{r},\omega_l)&=\hat{\boldsymbol{u}}_{ln}\mathcal{P}_n(\boldsymbol{r},\omega_l)\,,\end{aligned} \tag{72.20}$$

where $\hat{\boldsymbol{u}}_{ln}$ is the unit vector associated with the eigenpolarization of the spatial mode n at frequency ω_l. If the fields are assumed to travel along the z-direction, and the slowly-varying amplitude approximation $\partial^2A_n/\partial z^2\ll 2k_n\partial A_n/\partial z$ is made, the change in the amplitude of the field as it propagates through the nonlinear medium with no linear absorption is described by the differential equation

$$\frac{\mathrm{d}A_n(\omega_l)}{\mathrm{d}z}=\pm\left[\frac{1}{4\pi\varepsilon_0}\right]\frac{\mathrm{i}2\pi\omega_l}{n(\omega_l)c}\mathcal{P}_n^{\mathrm{NL}}(\omega_l)\,, \tag{72.21}$$

where $\mathcal{P}_n^{\mathrm{NL}}$ is the nonlinear contribution to the polarization amplitude $\mathcal{P}_n$, $n(\omega_l)$ is the linear refractive index at frequency ω_l, and the plus (minus) sign indicates propagation in the positive (negative) z-direction. Sections 72.3 and 72.4 give expressions for the $\mathcal{P}_n^{\mathrm{NL}}$ for various second- and third-order nonlinear optical processes. Equation (72.21)) is used to determine the set of coupled-amplitude equations describing a particular nonlinear process. For example, for the case of sum-frequency generation , the two fields of frequency ω_1 and ω_2 are combined through second-order nonlinear interaction to create a third wave at frequency $\omega_3=\omega_1+\omega_2$. Assuming full permutation symmetry, the amplitudes of the nonlinear polarization for each of the waves are

$$\mathcal{P}^{\mathrm{NL}}(z,\omega_1)=[\varepsilon_0]2\chi_{\mathrm{eff}}^{(2)}A(z,\omega_3)A^*(z,\omega_2)\,\mathrm{e}^{-\mathrm{i}\Delta kz}\,, \tag{72.22}$$

$$\mathcal{P}^{\mathrm{NL}}(z,\omega_2)=[\varepsilon_0]2\chi_{\mathrm{eff}}^{(2)}A(z,\omega_3)A^*(z,\omega_1)\,\mathrm{e}^{-\mathrm{i}\Delta kz}\,, \tag{72.23}$$

$$\mathcal{P}^{\mathrm{NL}}(z,\omega_3)=[\varepsilon_0]2\chi_{\mathrm{eff}}^{(2)}A(z,\omega_1)A(z,\omega_2)\,\mathrm{e}^{\mathrm{i}\Delta kz}\,, \tag{72.24}$$

where $\Delta k=k_1+k_2-k_3$ is the wavevector mismatch (see Sect. 72.2.2) and $\chi_{\mathrm{eff}}^{(2)}$ is given by

$$\chi_{\mathrm{eff}}^{(2)}=\sum_{ijk}\chi_{ijk}^{(2)}(\hat{\boldsymbol{u}}_1^*)_i(\hat{\boldsymbol{u}}_2)_j(\hat{\boldsymbol{u}}_3)_k\,, \tag{72.25}$$

where $(\hat{\boldsymbol{u}}_l)_i=\hat{\boldsymbol{u}}_l\cdot\hat{\boldsymbol{\imath}}$. For simplicity, the subscripts on each of the field amplitudes have been dropped, since only one spatial mode at each frequency contributed. The resulting coupled amplitude equations are

$$\frac{\mathrm{d}A(\omega_1)}{\mathrm{d}z}=\left[\frac{1}{4\pi}\right]\frac{\mathrm{i}4\pi\omega_1\chi_{\mathrm{eff}}^{(2)}}{n(\omega_1)c}A(\omega_3)A^*(\omega_2)\,\mathrm{e}^{-\mathrm{i}\Delta kz}\,, \tag{72.26}$$

$$\frac{\mathrm{d}A(\omega_2)}{\mathrm{d}z}=\left[\frac{1}{4\pi}\right]\frac{\mathrm{i}4\pi\omega_2\chi_{\mathrm{eff}}^{(2)}}{n(\omega_2)c}A(\omega_3)A^*(\omega_1)\,\mathrm{e}^{-\mathrm{i}\Delta kz}\,, \tag{72.27}$$

$$\frac{\mathrm{d}A(\omega_3)}{\mathrm{d}z}=\left[\frac{1}{4\pi}\right]\frac{\mathrm{i}4\pi\omega_3\chi_{\mathrm{eff}}^{(2)}}{n(\omega_3)c}A(\omega_1)A(\omega_2)\,\mathrm{e}^{\mathrm{i}\Delta kz}\,. \tag{72.28}$$

72.2.2 Phase Matching

For many nonlinear optical processes (e.g., harmonic generation) it is important to minimize the wave vector mismatch in order to maximize the efficiency. For example, if the field amplitudes $A(\omega_1)$ and $A(\omega_2)$ are constant, the solution to (72.28) yields for the output intensity

$$I(L,\omega_3) = \left[\frac{1}{64\pi^3\varepsilon_0}\right] \times \frac{32\pi^3\left[\chi^{(2)}_{\text{eff}}\right]^2 \omega_3^2 I(\omega_1) I(\omega_2) L^2}{n(\omega_1)n(\omega_2)n(\omega_3)c^3} \times \operatorname{sinc}^2(\Delta k L/2)\,, \tag{72.29}$$

in terms of sinc $x = (\sin x)/x$, where $I(L,\omega_3) = (4\pi\varepsilon_0)n(\omega_3)c|A(L,\omega_3)|^2/2\pi$, and $I(\omega_1)$ and $I(\omega_2)$ are the corresponding input intensities. Clearly, the effect of the wavevector mismatch is to reduce the efficiency of the generation of the sum frequency wave. The maximum propagation distance over which efficient nonlinear coupling can occur is given by the coherence length

$$L_c = \frac{2}{\Delta k}\,. \tag{72.30}$$

As a result of the dispersion in the linear refractive index that occurs in all materials, achieving phase matching over typical interaction lengths (e.g., 5 mm) is nontrivial. For the case in which the nonlinear material is birefringent, it is sometimes possible to achieve phase matching by insuring that the interacting waves possess some suitable combination of ordinary and extraordinary polarization. Other techniques for achieving phase matching include quasiphase matching [72.5] and the use of the mode dispersion in waveguides [72.6].

However, the phase matching condition is automatically satisfied for certain nonlinear optical processes, such as two-photon absorption (see Sect. 72.4.6) and Stokes amplification in stimulated Raman scattering (see Sect. 72.5.1). One can tell when the phase matching condition is automatically satisfied by examining the frequencies that appear in the expression for the nonlinear susceptibility. For a nonlinear susceptibility of the sort $\chi^{(3)}(\omega_1;\omega_2,\omega_3,\omega_4)$ the wave vector mismatch is given in general by $\Delta \boldsymbol{k} = \boldsymbol{k}_2 + \boldsymbol{k}_3 + \boldsymbol{k}_4 - \boldsymbol{k}_1$. Thus, for the example of Stokes amplification in stimulated Raman scattering, the nonlinear susceptibility is given by $\chi^{(3)}(\omega_1;\omega_1,\omega_0,-\omega_0)$ where $\omega_0(\omega_1)$ is the frequency of the pump (Stokes) wave, and consequently the wave vector mismatch vanishes identically.

72.2.3 Manley–Rowe Relations

Under conditions of full permutation symmetry, there is no flow of power from the electromagnetic fields to the medium, and thus the total power flow of the fields is conserved. The flow of energy among the fields can be described by the Manley–Rowe relations. For example, for the case of sum-frequency generation, one can deduce from (72.26, 27, 28) that

$$\frac{\mathrm{d}}{\mathrm{d}z}\left[\frac{I(\omega_1)}{\omega_1}\right] = \frac{\mathrm{d}}{\mathrm{d}z}\left[\frac{I(\omega_2)}{\omega_2}\right] = -\frac{\mathrm{d}}{\mathrm{d}z}\left[\frac{I(\omega_3)}{\omega_3}\right]. \tag{72.31}$$

The expressions in square brackets are proportional to the flux of photons per unit area per unit time, and imply that the creation of a photon at ω_3 must be accompanied by the annihilation of photons at both ω_1 and ω_2. Similar relations can be formulated for other nonlinear optical processes that are governed by a nonlinear susceptibility that satisfies full permutation symmetry. Since this behavior occurs at the photon level, nonlinear optical processes can lead to the generation of light fields that have esoteric quantum statistical properties (Chapt. 78 and Chapt. 80).

A nonlinear optical process that satisfies the Manley–Rowe relations is called a parametric process. Conversely, a process for which field energy is not conserved, and thus Manley–Rowe relations cannot be formulated, is said to be nonparametric. Thus, parametric processes are described by purely real $\chi^{(n)}$, whereas nonparametric proceses are described by complex $\chi^{(n)}$.

72.2.4 Pulse Propagation

If the optical field consists of ultrashort (<100 ps) pulses, it is more convenient to work with the temporally varying amplitude, rather than with the individual frequency components. Thus, for a linearly polarized plane wave pulse propagating along the z-axis, the field is decomposed into the product of a slowly varying amplitude $A(z,t)$ and a rapidly varying oscillatory term such that

$$\tilde{E}(\boldsymbol{r},t) = A(z,t)\,\mathrm{e}^{\mathrm{i}(k_0 z - \omega_0 t)} + \text{c.c.}\,, \tag{72.32}$$

where $k_0 = n_0\omega_0/c$. For a pulse propagating in a material with an intensity-dependent refractive index, the propagation can be described by the nonlinear Schrödinger equation

$$\frac{\partial A}{\partial z} + \frac{\mathrm{i}\beta_2}{2}\frac{\partial^2 A}{\partial \tau^2} = \mathrm{i}\gamma|A|^2 A\,, \tag{72.33}$$

where $\beta_2 = \left(\mathrm{d}^2k/\mathrm{d}\omega^2\right)|_{\omega=\omega_0}$ is the group velocity dispersion parameter, $\tau = t - z/v_g$ is the local time for the pulse, $v_g = [(\mathrm{d}k/\mathrm{d}\omega)|_{\omega=\omega_0}]^{-1}$ is the group velocity, and $\gamma = [4\pi\varepsilon_0]n_2 n_0\omega_0/2\pi$ is the nonlinear refractive index parameter.

72.3 Second-Order Processes

Second-order nonlinear optical processes occur as a consequence of the second term in expression (72.3), i. e., processes whose strength is described by $\chi^{(2)}(\omega_\sigma; \omega_m, \omega_n)$. These processes entail the generation of a field at frequency $\omega_\sigma = \omega_m + \omega_n$ in response to applied fields at (positive and/or negative) frequencies ω_m and ω_n. Several examples of such processes are described in this Section.

72.3.1 Sum Frequency Generation

Sum frequency generation produces an output field at frequency $\omega_3 = \omega_1 + \omega_2$ for ω_1 and ω_2 both positive. It is useful, for example, for the generation of tunable radiation in the uv if ω_1 and/or ω_2 are obtained from tunable lasers in the visible range. Sum frequency generation is described in detail in Sects. 72.2.1–72.2.3.

72.3.2 Second Harmonic Generation

Second harmonic generation is routinely used to convert the output of a laser to a higher frequency. It is described by $\chi^{(2)}(2\omega; \omega, \omega)$. Let η be the power conversion efficiency from frequency ω to 2ω. Assuming that phase matching is perfect, and the pump wave at frequency ω is undepleted by the interaction, a derivation analogous to that for (72.29) yields

$$\eta = \tanh^2\left(z/l\right) , \tag{72.34}$$

where the characteristic conversion length l is given by

$$l = \left[4\pi\right] \frac{c\sqrt{n(\omega)n(2\omega)}}{4\pi\omega\chi^{(2)}|A_1(0)|} . \tag{72.35}$$

Note that the conversion efficiency asymptotically approaches unity. In practice, conversion efficiencies exceeding 80% can be achieved.

72.3.3 Difference Frequency Generation

Difference frequency generation can be used to create light in the infrared and far infrared by generating the difference frequency $\omega_2 = \omega_3 - \omega_1$ (where ω_3 and ω_1 are positive and $\omega_3 > \omega_1$) of two incident lasers. Consider the case in which a strong (undepleted) pump wave at frequency ω_3 and a weak (signal) wave at ω_1 are incident on a nonlinear medium described by $\chi^{(2)}(\omega_2; \omega_3, -\omega_1) = \chi^{(2)}(\omega_1; \omega_3, -\omega_2)$. The amplitude $A(\omega_3)$ of the strong wave can be taken as a constant, and thus the interaction can be described by finding simultaneous solutions to (72.26) and (72.27) for $A(\omega_1)$ and $A(\omega_2)$. In the limit of perfect phase matching (i. e., $\Delta k = 0$), the solutions are

$$A(z, \omega_1) = A(0, \omega_1) \cosh \kappa z , \tag{72.36}$$

$$A(z, \omega_2) = \mathrm{i}\sqrt{\frac{n_1\omega_2}{n_2\omega_1}} \frac{A(\omega_3)}{|A(\omega_3)|} A^*(0, \omega_1) \sinh \kappa z , \tag{72.37}$$

where

$$\kappa^2 = \left[\frac{1}{16\pi^2}\right] \frac{16\pi^2\left[\chi^{(2)}\right]^2\omega_1^2\omega_2^2}{k_1k_2c^4} |A(\omega_3)|^2 . \tag{72.38}$$

Equation (72.37) describes the spatial growth of the difference frequency signal.

72.3.4 Parametric Amplification and Oscillation

For the foregoing case of a strong wave at frequency ω_3 and a weak wave with $\omega_1 < \omega_3$ incident on a second-order nonlinear optical material, the lower frequency input wave is amplified by the nonlinear interaction; this process is known as parametric amplification. Difference frequency generation is a consequence of the Manley–Rowe relations, as described above in Sect. 72.2.3. Since $\omega_3 = \omega_1 + \omega_2$, the annihilation of an ω_3 photon must be accompanied by the simultaneous creation of photons ω_1 and ω_2.

An optical parametric oscillator can be constructed by placing the nonlinear optical material inside an optical resonator that provides feedback at ω_1 and/or ω_2. When such a device is excited by a wave at ω_3, it can produce output frequencies ω_1 and ω_2 that satisfy $\omega_1 + \omega_2 = \omega_3$. Optical parametric oscillators are of considerable interest as sources of broadly tunable radiation [72.7].

72.3.5 Focused Beams

For conceptual clarity, much of the discussion so far has assumed that the interacting beams are plane waves. In practice, the incident laser beams are often focused into the nonlinear material to increase the field strength within the interaction region and consequently to increase the nonlinear response. However, it is undesirable to focus too tightly, because doing so leads to a decrease in the effective length of the interaction region. In particular, if w_0 is the radius of the laser beam at the beam waist, the beam remains focused only over a distance of the order $b = 2\pi w_0^2/\lambda$ where λ is the laser wavelength measured in the nonlinear material. For many types of nonlinear optical processes, the optimal nonlinear response occurs if the degree of focusing is adjusted so that b is several times smaller than the length L of the nonlinear optical material.

72.4 Third-Order Processes

A wide variety of nonlinear optical processes are possible as a result of the nonlinear contributions to the polarization that are third-order in the applied field. These processes are described by $\chi^{(3)}(\omega_\sigma; \omega_m, \omega_n, \omega_o)$ (72.3) and can lead not only to the generation of new field components (e.g., third-harmonic generation) but can also result in a field affecting itself as it propagates (e.g., self-phase modulation). Several examples are described in this section.

72.4.1 Third-Harmonic Generation

Assuming full-permutation symmetry, the nonlinear polarization amplitudes for the fundamental and third-harmonic beams are

$$\begin{aligned}\mathcal{P}^{\mathrm{NL}}(z,\omega) &= [\varepsilon_0]3\chi_{\mathrm{eff}}^{(3)}A(z,3\omega)[A^*(z,\omega)]^2\,\mathrm{e}^{-\mathrm{i}\Delta kz}\,,\\ \mathcal{P}^{\mathrm{NL}}(z,3\omega) &= [\varepsilon_0]\chi_{\mathrm{eff}}^{(3)}[A(z,\omega)]^3\,\mathrm{e}^{\mathrm{i}\Delta kz}\,,\end{aligned} \tag{72.39}$$

where $\Delta k = 3k(\omega) - k(3\omega)$ and $\chi_{\mathrm{eff}}^{(3)}$ is the effective third-order susceptibility for third-harmonic generation and is defined in a manner analogous to the $\chi_{\mathrm{eff}}^{(2)}$ in (72.25). If the intensity of the fundamental wave is not depleted by the nonlinear interaction, the solution for the output intensity $I(L, 3\omega)$ of the third-harmonic field for a crystal of length L is

$$\begin{aligned}I(L,3\omega) = {}&\left[\frac{1}{256\pi^4\varepsilon_0^2}\right]\frac{48\pi^2\omega^2\left[\chi_{\mathrm{eff}}^{(3)}\right]^2}{n(3\omega)n(\omega)^3c^4}\\ &\times I(\omega)^3L^2\sinh^2[\Delta kL/2]\,,\end{aligned} \tag{72.40}$$

where $I(\omega)$ is the input intensity of the fundamental field. As a result of the typically small value of $\chi_{\mathrm{eff}}^{(3)}$ in crystals, it is generally more efficient to generate the third harmonic by using two $\chi^{(2)}$ crystals in which the first crystal produces second harmonic light and the second crystal combines the second harmonic and the fundamental beams via sum-frequency generation. It is also possible to use resonant enhancement of $|\chi^{(3)}|$ in gases to increase the efficiency of third-harmonic generation [72.8].

72.4.2 Self-Phase and Cross-Phase Modulation

The nonlinear refractive index leads to an intensity-dependent change in the phase of the beam as it propagates through the material. If the medium is lossless, the amplitude of a single beam at frequency ω propagating in the positive z-direction can be expressed as

$$A(z,\omega) = A(0,\omega)\,\mathrm{e}^{\mathrm{i}\phi^{\mathrm{NL}}(z)}\,, \tag{72.41}$$

where the nonlinear phase shift $\phi^{\mathrm{NL}}(z)$ is given by

$$\phi^{\mathrm{NL}}(z) = \frac{\omega}{c}n_2Iz\,, \tag{72.42}$$

and $I = [4\pi\varepsilon_0]n_0c|A(0,\omega)|^2/2\pi$ is the intensity of the laser beam. If two fields at different frequencies ω_1 and ω_2 are traveling along the z-axis, the two fields can affect each other's phase; this effect is known as cross-phase modulation. The nonlinear phase shift $\phi_{1,2}^{\mathrm{NL}}(z)$ for each of the waves is given by

$$\phi_{1,2}^{\mathrm{NL}}(z) = \frac{\omega_{1,2}}{c}n_2(I_{1,2} + 2I_{2,1})z\,. \tag{72.43}$$

For the case of a light pulse, the change in the phase of the pulse inside the medium becomes a function of time. In this case the solution to (72.33) for the time-varying amplitude $A(z,\tau)$ shows that in the absence of group-velocity dispersion (GVD) (i.e., $\beta_2 = 0$) that the solution for $A(z,\tau)$ is of the form of (72.41), except that the temporal intensity profile $I(\tau)$ replaces the steady-state intensity I in (72.42). As the pulse propagates through the medium, its frequency becomes time dependent, and the instantaneous frequency shift from the central frequency ω_0 is given by

$$\delta\omega(\tau) = -\frac{\partial\phi^{\mathrm{NL}}(\tau)}{\partial\tau} = -\frac{\omega n_2z}{c}\frac{\partial I}{\partial t}\,. \tag{72.44}$$

This time-dependent self-phase modulation leads to a broadening of the pulse spectrum and to a frequency chirp across the pulse.

If the group velocity dispersion parameter β_2 and the nonlinear refractive index coefficient n_2 are of opposite sign, the nonlinear frequency chirp can be compensated by the chirp due to group velocity dispersion, and (72.33) admits soliton solutions . For example, the fundamental soliton solution is

$$A(z,t)=\sqrt{\frac{1}{L_\mathrm{D}}}\operatorname{sech}\left(\frac{\tau}{\tau_p}\right)\mathrm{e}^{\mathrm{i}z/2L_\mathrm{D}}\,, \tag{72.45}$$

where τ_p is the pulse duration and $L_\mathrm{D}=\tau_p^2/|\beta_2|$ is the dispersion length. As a result of their ability to propagate in dispersive media without changing shape, optical solitons show a great deal of promise in applications such as optical communications and optical switching. For further discussion of optical solitons see [72.9].

72.4.3 Four-Wave Mixing

Various types of four-wave mixing processes can occur among different beams. One of the most common geometries is backward four-wave mixing used in nonlinear spectroscopy and optical phase conjugation. In this interaction, two strong counterpropagating pump waves with amplitudes A_1 and A_2 and with equal frequencies $\omega_{1,2}=\omega$ are injected into a nonlinear medium. A weak wave, termed the probe wave, (with frequency ω_3 and amplitude A_3) is also incident on the medium. As a result of the nonlinear interaction among the three waves, a fourth wave with an amplitude A_4 is generated which is counterpropagating with respect to the probe wave and with frequency $\omega_4=2\omega-\omega_3$. For this case, the third-order nonlinear susceptibilities for the probe and conjugate waves are given by $\chi^{(3)}(\omega_{3,4};\,\omega,\,\omega,\,-\omega_{4,3})$. For constant pump wave intensities and full permutation symmetry, the amplitudes of the nonlinear polarization for the probe and conjugate waves are given by

$$\begin{aligned}\mathcal{P}^{\mathrm{NL}}(z,\omega_{3,4})=\pm[\varepsilon_0]6\chi^{(3)}\big[&\big(|A_1|^2+|A_2|^2\big)A_{3,4}\\ &+A_1A_2A^*_{4,3}\,\mathrm{e}^{\mathrm{i}\Delta kz}\big]\,,\end{aligned} \tag{72.46}$$

where $\Delta k=k_1+k_2-k_3-k_4$ is the phase mismatch, which is nonvanishing when $\omega_3\neq\omega_4$. For the case of optical phase conjugation by degenerate four-wave mixing (i. e., $\omega_3=\omega_4=\omega$ and $A_4(L)=0$), the phase conjugate reflectivity R_PC is

$$R_\mathrm{PC}=\frac{|A_4(0)|^2}{|A_3(0)|^2}=\tan^2(\kappa L)\,, \tag{72.47}$$

where $\kappa=\big[1/16\pi^2\varepsilon_0\big]\big[24\pi^2\omega\chi^{(3)}/(n_0c)^2\big]\sqrt{I_1I_2}$ and $I_{1,2}$ are the intensities of the pump waves. Phase-conjugate reflectivities greater than unity can be routinely achieved by performing four-wave mixing in atomic vapors or photorefractive media.

72.4.4 Self-Focusing and Self-Trapping

Typically a laser beam has a transverse intensity profile that is approximately Gaussian. In a medium with an intensity-dependent refractive index, the index change at the center of the beam is different from the index change at the edges of the beam. The gradient in the refractive index created by the beam can allow it to self-focus for $n_2>0$. For this condition to be met, the total input power of the beam must exceed the critical power P_cr for self-focusing which is given by

$$P_\mathrm{cr}=\frac{\pi(0.61\lambda)^2}{8n_0n_2}\,, \tag{72.48}$$

where λ is the vacuum wavelength of the beam. For powers much greater than the critical power, the beam can break up into various filaments, each with a power approximately equal to the critical power. For a more extensive discussion of self-focusing and self-trapping see [72.10, 11].

72.4.5 Saturable Absorption

When the frequency ω of an applied laser field is sufficiently close to a resonance frequency ω_0 of the medium, an appreciable fraction of the atomic population can be placed in the excited state. This loss of population from the ground state leads to an intensity-dependent saturation of the absorption and the refractive index of the medium (see Sect. 69.2 for more detailed discussion) [72.4]. The third-order susceptibility as a result of this saturation is given by

$$\chi^{(3)}=\left[\frac{1}{\varepsilon_0}\right]\frac{|\mu|^2T_1T_2\alpha_0c}{3\pi\omega_0\hbar^2}\,\frac{\delta T_2-i}{\big[1+(\delta T_2)^2\big]^2}\,, \tag{72.49}$$

where μ is the transition dipole moment, T_1 and T_2 are the longitudinal and transverse relaxation times, respectively (see Sect. 68.4.3), α_0 is the line-center weak-field intensity absorption coefficient, and $\delta=\omega-\omega_0$ is the detuning. For the 3s ↔ 3p transition in atomic sodium vapor at 300 °C, the nonlinear refractive index $n_2\approx10^{-7}\,\mathrm{cm}^2/\mathrm{W}$ for a detuning $\delta T_2=300$.

72.4.6 Two-Photon Absorption

When the frequency ω of a laser field is such that 2ω is close to a transition frequency of the material, it is possible for two-photon absorption (TPA) to occur. This process leads to a contribution to the imaginary part of $\chi^{(3)}(\omega;\omega,\omega,-\omega)$. In the presence of TPA, the intensity $I(z)$ of a single, linearly polarized beam as a function of propagation distance is

$$I(z)=\frac{I(0)}{1+\beta I(0)z}\,, \tag{72.50}$$

where $\beta=\left[1/16\pi^2\varepsilon_0\right]24\pi^2\omega\,\mathrm{Im}\left[\chi^{(3)}\right]/(n_0c)^2$ is the TPA coefficient. For wide-gap semiconductors such as ZnSe at 800 nm, $\beta\approx10^{-8}\,\mathrm{cm/W}$.

72.4.7 Nonlinear Ellipse Rotation

The polarization ellipse of an elliptically polarized laser beam rotates but retains its ellipticity as the beam propagates through an isotropic nonlinear medium. Ellipse rotation occurs as a result of the difference in the nonlinear index changes experienced by the left- and right-circular components of the beam, and the angle θ of rotation is

$$\begin{aligned}\theta&=\frac{1}{2}\Delta n\omega z/c\\&=\left[\frac{1}{16\pi^2\varepsilon_0}\right]\frac{12\pi^2}{n_0^2c}\chi^{(3)}_{xyyx}\\&\quad\times(\omega;\omega,\omega,-\omega)(I_+-I_-)z\,,\end{aligned} \tag{72.51}$$

where $I_\pm$ are the intensities of the circularly polarized components of the beam with unit vectors $\hat{\boldsymbol{\sigma}}_\pm=(\hat{\boldsymbol{x}}\pm i\hat{\boldsymbol{y}})/\sqrt{2}$. Nonlinear ellipse rotation is a sensitive technique for determining the nonlinear susceptibility element $\chi^{(3)}_{xyyx}$ for isotropic media and can be used in applications such as optical switching.

72.5 Stimulated Light Scattering

Stimulated light scattering occurs as a result of changes in the optical properties of the material that are induced by the optical field. The resulting nonlinear coupling between different field components is mediated by some excitation (e.g., acoustic phonon) of the material that results in changes in its optical properties. The nonlinearity can be described by a complex susceptibility and a nonlinear polarization that is of third order in the interacting fields. Various types of stimulated scattering can occur. Discussed below are the two processes that are most commonly observed.

72.5.1 Stimulated Raman Scattering

In stimulated Raman scattering (SRS), the light field interacts with a vibrational mode of a molecule. The coupling between the two optical waves can become strong if the frequency difference between them is close to the frequency ω_v of the molecular vibrational mode. If the pump field at ω_0 and another field component at ω_1 are propagating in the same direction along the z-axis, the steady-state nonlinear polarization amplitudes for the two field components are given by

$$\begin{aligned}\mathcal{P}^{\mathrm{NL}}(z,\omega_{0,1})&=[\varepsilon_0]6\chi_{\mathrm{R}}(\omega_{0,1})\\&\quad\times|A(z,\omega_{1,0})|^2A(z,\omega_{0,1})\,,\end{aligned} \tag{72.52}$$

where $\chi_{\mathrm{R}}(\omega_{0,1})\equiv\chi^{(3)}(\omega_{0,1};\omega_{0,1},\omega_{1,0},-\omega_{1,0})$, the Raman susceptibility, actually depends only on the frequency difference $\Omega=\omega_0-\omega_1$ and is given by

$$\chi_{\mathrm{R}}(\omega_{0,1})=\left[\frac{1}{\varepsilon_0}\right]\frac{N(\partial\alpha/\partial q)_0^2}{6\mu_M}\frac{1}{\omega_v^2-\Omega^2\mp2\mathrm{i}\gamma\Omega}\,, \tag{72.53}$$

where the minus (plus) sign is taken for the ω_0 (ω_1) susceptibility, μ_M is the reduced nuclear mass, and $(\partial\alpha/\partial q)_0$ is a measure of the change of the polarizability of the molecule with respect to a change in the intermolecular distance q at equilibrium. If the intensity of the pump field is undepleted by the interaction with the ω_1 field and is assumed to be constant, the solution for the intensity of the ω_1 field at $z=L$ is given by

$$I(L,\omega_1)=I(0,\omega_1)\,\mathrm{e}^{G_{\mathrm{R}}}\,, \tag{72.54}$$

where the SRS gain parameter G_{R} is

$$\begin{aligned}G_{\mathrm{R}}&=\left[\frac{1}{16\pi^2\varepsilon_0}\right]48\pi^2\frac{\omega_1}{(n_1c)^2}\mathrm{Im}[\chi_{\mathrm{R}}(\omega_1)]I_0L\\&=g_{\mathrm{R}}I_0L\,,\end{aligned} \tag{72.55}$$

g_{R} is the SRS gain factor, and I_0 is the input intensity of the pump field. For $\omega_1<\omega_0$ ($\omega_1>\omega_0$), the ω_1 field is termed the Stokes (anti-Stokes) field, and it experiences exponential amplification (attenuation). For sufficiently large gains (typically $G_{\mathrm{R}}\gtrsim25$), the Stokes wave can be seeded by spontaneous Raman scattering and can grow to an appreciable fraction of the pump field. For a complete discussion of the spontaneous initiation of SRS see [72.12]. For the case of CS$_2$, $g_\mathrm{R} = 0.024\,\mathrm{cm/MW}$.

Four-wave mixing processes that couple a Stokes wave having $\omega_1 < \omega_0$ and an anti-Stokes wave having $\omega_2 > \omega_0$, where $\omega_1 + \omega_2 = 2\omega_0$, can also occur [72.4]. In this case, additional contributions to the nonlinear polarization are present and are characterized by a Raman susceptibility of the form $\chi^{(3)}(\omega_{1,2}; \omega_0, \omega_0, -\omega_{2,1})$. The technique of coherent anti-Stokes Raman spectroscopy is based on this four-wave mixing process [72.13].

72.5.2 Stimulated Brillouin Scattering

In stimulated Brillouin scattering (SBS), the light field induces and interacts with an acoustic wave inside the medium. The resulting interaction can lead to extremely high amplification for certain field components (i. e., Stokes wave). For many optical media, SBS is the dominant nonlinear optical proccess for laser pulses of duration > 1 ns. The primary applications for SBS are self-pumped phase conjugation and pulse compression of high-energy laser pulses.

If an incident light wave with wave vector $\boldsymbol{k}_0$ and frequency ω_0 is scattered from an acoustic wave with wave vector $\boldsymbol{q}$ and frequency Ω, the wave vector and frequency of the scattered wave are determined by conservation of momentum and energy to be $\boldsymbol{k}_1 = \boldsymbol{k}_0 \pm \boldsymbol{q}$ and $\omega_1 = \omega_0 \pm \Omega$, where the (+) sign applies if $\boldsymbol{k}_0 \cdot \boldsymbol{q} > 0$ and the (−) applies if $\boldsymbol{k}_0 \cdot \boldsymbol{q} < 0$. Here, Ω and $\boldsymbol{q}$ are related by the dispersion relation $\Omega = v|\boldsymbol{q}|$ where v is the velocity of sound in the material. These Bragg scattering conditions lead to the result that the Brillouin frequency shift $\Omega_\mathrm{B} = \omega_1 - \omega_0$ is zero for scattering in the forward direction (i. e., in the $\boldsymbol{k}_0$ direction) and reaches its maximum for scattering in the backward direction given by

$$\Omega_\mathrm{B} = 2\omega_0 v n_0 / c\,, \tag{72.56}$$

where n_0 is the refractive index of the material.

The interaction between the incident wave and the scattered wave in the Brillouin-active medium can become nonlinear if the interference between the two optical fields can coherently drive an acoustic wave, either through electrostriction or through local density fluctuations resulting from the absorption of light and consequent temperature changes. The following discussion treats the more common electrostriction mechanism.

Typically, SBS occurs in the backward direction (i. e., $\boldsymbol{k}_0 = k_0\hat{z}$ and $\boldsymbol{k}_1 = -k_1\hat{z}$), since the spatial overlap between the Stokes beam and the laser beam is maximized under these conditions and, as mentioned above, no SBS occurs in the forward direction. The steady-state nonlinear polarization amplitudes for backward SBS are

$$\begin{aligned}\mathcal{P}^{\mathrm{NL}}(z, \omega_{0,1}) &= [\varepsilon_0] 6 \chi_\mathrm{B}(\omega_{0,1}) \\ &\quad \times |A(z, \omega_{1,0})|^2 A(z, \omega_{0,1})\,,\end{aligned} \tag{72.57}$$

where $\chi_\mathrm{B}(\omega_{0,1}) \equiv \chi^{(3)}(\omega_{0,1}; \omega_{0,1}, \omega_{1,0}, -\omega_{1,0})$, the Brillouin susceptibility, depends only on $\Omega = \omega_0 - \omega_1$ and is given by

$$\chi_\mathrm{B}(\omega_{0,1}) = \left[\frac{1}{\varepsilon_0}\right] \frac{\omega_0^2 \gamma_\mathrm{e}^2}{24\pi^2 c^2 \rho_0} \frac{1}{\Omega_\mathrm{B}^2 - \Omega^2 \mp \mathrm{i}\Gamma_\mathrm{B}\Omega}\,, \tag{72.58}$$

where the minus (plus) sign is taken for the ω_0 (ω_1) susceptibility, γ_e is the electrostrictive constant, ρ_0 is the mean density of the material, and Γ_B is the Brillouin linewidth given by the inverse of the phonon lifetime. If the pump field is undepleted by the interaction with the ω_1 field and is assumed to be constant, the solution for the output intensity of the ω_1 field at $z = 0$ is given by

$$I(0, \omega_1) = I(L, \omega_1)\,\mathrm{e}^{G_\mathrm{B}}\,, \tag{72.59}$$

where the Brillouin gain coefficient G_B is given by

$$\begin{aligned}G_\mathrm{B} &= \left[\frac{1}{16\pi^2\varepsilon_0}\right] 48\pi^2 \frac{\omega_1}{(n_0 c)^2} \mathrm{Im}[\chi_\mathrm{B}(\omega_1)] I_0 L, \\ &= g_0 \frac{\Omega\Omega_\mathrm{B}\Gamma_\mathrm{B}^2}{\left[\Omega_\mathrm{B}^2 - \Omega^2\right]^2 + (\Omega\Gamma_\mathrm{B})^2} I_0 L \\ &= g_\mathrm{B} I_0 L\,,\end{aligned} \tag{72.60}$$

g_B is the SBS gain factor, I_0 is the input intensity of the pump field, and

$$g_0 = \left[\frac{1}{\varepsilon_0^2}\right] \frac{\omega_0^2 \gamma_\mathrm{e}^2}{n_0 c^3 \rho_0 v \Gamma_\mathrm{B}} \tag{72.61}$$

is the line-center (i. e., $\Omega = \pm\Omega_\mathrm{B}$) SBS gain factor. For $\Omega > 0$ ($\Omega < 0$), the ω_1 field is termed the Stokes (anti-Stokes) field, and it experiences exponential amplification (attenuation). For sufficiently large gains (typically $G_\mathrm{B} \gtrsim 25$), the Stokes wave can be seeded by spontaneous Brillouin scattering and can grow to an appreciable fraction of the pump field. For a complete discussion of the spontaneous initiation of SBS see [72.14]. For CS$_2$, $g_0 = 0.15\,\mathrm{cm/MW}$.

72.6 Other Nonlinear Optical Processes

72.6.1 High-Order Harmonic Generation

If full permutation symmetry applies and the fundamental field ω is not depleted by nonlinear interactions, then the intensity of the qth harmonic is given by

$$\begin{aligned} I(z, q\omega) = &\left[\frac{1}{4\pi(4\pi\varepsilon_0)^{(q-1)/2}}\right] \\ &\times \frac{2\pi q^2\omega^2}{n^2(q\omega)c}\left[\frac{2\pi I(\omega)}{n(\omega)c}\right]^q \\ &\times \left|\chi^{(q)}(q\omega; \omega, \ldots, \omega) J_q(\Delta k, z_0, z)\right|^2 , \end{aligned} \tag{72.62}$$

where $\Delta k = [n(\omega) - n(q\omega)]\omega/c$,

$$J_q(\Delta k, z_0, z) = \int_{z_0}^{z} \frac{\mathrm{e}^{\mathrm{i}\Delta k z'}\,\mathrm{d}z'}{(1 + 2\mathrm{i}z'/b)^{q-1}} , \tag{72.63}$$

$z = z_0$ at the input face of the nonlinear medium, and b is the confocal parameter Sect. 72.3.5 of the fundamental beam. Defining $L = z - z_0$, the integral J_q can be easily evaluated in the limits $L \ll b$ and $L \gg b$. The limit $L \ll b$ corresponds to the plane-wave limit in which case

$$|J_q(\Delta k, z_0, z)|^2 = L^2 \mathrm{sinc}^2\left(\frac{\Delta k L}{2}\right) . \tag{72.64}$$

The limit $L \gg b$ corresponds to the tight-focusing configuration in which case

$$J_q(\Delta k, z_0, z) = \begin{cases} 0, & \Delta k \leq 0 , \\ \dfrac{\pi b}{(q-2)!}\left(\dfrac{b\Delta k}{2}\right)^{q-2} \mathrm{e}^{-b\Delta k/2}, & \\ & \Delta k > 0 . \end{cases} \tag{72.65}$$

Note that in this limit, the qth harmonic light is only generated for positive phase mismatch. *Reintjes* et al. [72.15, 16] observed both the fifth and seventh harmonics in helium gas which exhibited a dependence on $I(\omega)$ which is consistent with the $I^q(\omega)$ dependence predicted by (72.62). However, more recent experiments in gas jets have demonstrated the generation of extremely high-order harmonics which do not depend on the intensity in this simple manner (see Chapt. 74 for further discussion of this nonperturbative behavior).

72.6.2 Electro-Optic Effect

The electro-optic effect corresponds to the limit in which the frequency of one of the applied fields approaches zero. The linear electro-optic effect (or Pockels effect) can be described by a second-order susceptibility of the form $\chi^{(2)}(\omega; \omega, 0)$. This effect produces a change in the refractive index for light of certain polarizations which depends linearly on the strength of the applied low-frequency field. More generally, the linear electro-optic effect induces a change in the amount of birefringence present in an optical material. This electrically controllable change in birefringence can be used to construct amplitude modulators, frequency shifters, optical shutters, and other optoelectronic devices. Materials commonly used in such devices include KDP and lithium niobate [72.17]. If the laser beam is propagating along the optic axis (i.e., z-axis) of the material of length L and the low-frequency field E_z is also applied along the optic axis, the nonlinear index change $\Delta n = n_y - n_x$ between the components of the electric field polarized along the principal axes of the crystal is given by

$$\Delta n = \left[\frac{1}{4\pi}\right] n_0^3 r_{63} E_z \tag{72.66}$$

where r_{63} is one of the electro-optic coefficients.

The quadratic electro-optic effect produces a change in refractive index that scales quadratically with the applied dc electric field. This effect can be described by a third-order susceptibility of the form $\chi^{(3)}(\omega; \omega, 0, 0)$.

72.6.3 Photorefractive Effect

The photorefractive effect leads to an optically induced change in the refractive index of a material. In certain ways this effect mimics that of the nonlinear refractive index described in Sect. 72.1.2, but it differs from the nonlinear refractive index in that the change in refractive index is independent of the overall intensity of the incident light field, and depends only on the degree of spatial modulation of the light field within the nonlinear material. In addition, the photorefractive effect can occur only in materials that exhibit a linear electro-optic effect, and contain an appreciable density of trapped electrons and/or holes that can be liberated by the application of a light field. Typical photorefractive materials include lithium niobate, barium titanate, and strontium barium niobate.

A typical photorefractive configuration might be as follows: two beams interfere within a photorefractive crystal to produce a spatially modulated intensity distribution. Bound charges are ionized with greater probability at the maxima than at the minima of the distribution and, as a result of the diffusion process, carriers tend to migrate away from regions of large light intensity. The resulting modulation of the charge distribution leads to the creation of a spatially modulated electric field that produces a spatially modulated change in refractive index as a consequence of the linear electro-optic effect. For a more extensive discussion see [72.18].

72.6.4 Ultrafast and Intense-Field Nonlinear Optics

Additional nonlinear optical processes are enabled by the use of ultrashort (< 1 ps) or ultra-intense laser pulses. For reasons of basic laser physics, ultra-intense pulses are necessarily of short duration, and thus these effects normally occur together. Ultrashort laser pulses possess a broad frequency spectrum, and therefore the dispersive properties of the optical medium play a key role in the propagation of such pulses. The three-dimensional nonlinear Schrödinger equation must be modified when treating the propagation of these ultrashort pulses by including contributions that can be ignored under other circumstances [72.19,20]. These additional terms lead to processes such as space-time coupling, self-steepening, and shock wave formation [72.21, 22]. The process of self-focusing is significantly modified under short-pulse (pulse duration shorter than approximately 1 ps) excitation. For example, temporal splitting of a pulse into two components can occur; this pulse splitting lowers the peak intensity, and can lead to the arrest of the usual collapse of a pulse undergoing self-focusing [72.23]. Moreover, optical shock formation, the creation of a discontinuity in the intensity evolution of a propagating pulse, leads to supercontinuum generation, the creation of a light pulse with an extremely broad frequency spectrum [72.24]. Shock effects and the generation of supercontinuum light can also occur in one-dimensional systems, such as a microstructure optical fiber. The relatively high peak power of the ultrashort pulses from a mode-locked laser oscillator and the tight confinement of the optical field in the small ($\approx 2\,\mu$m) core of the fiber yield high intensities and strong self-phase modulation, which results in a spectral bandwidth that spans more than an octave of the central frequency of the pulse [72.25]. Such a coherent octave-spanning spectrum allows for the stabilization of the underlying frequency comb of the mode-locked oscillator, and has led to a revolution in the field of frequency metrology [72.26]. Multiphoton absorption [72.27] constitutes an important loss process that becomes important for intensities in excess of $\approx 10^{13}\,\mathrm{W/cm^2}$. In addition to introducing loss, the electrons released by this process can produce additional nonlinear effects associated with their relativistic motion in the resulting plasma [72.28, 29]. For very large laser intensities (greater than approximately $10^{16}\,\mathrm{W/cm^2}$), the electric field strength of the laser pulse can exceed the strength of the Coulomb field that binds the electron to the atomic core, and nonperturbative effects can occur. A dramatic example is that of high-harmonic generation [72.30–32]. Harmonic orders as large as the 341-st have been observed, and simple conceptual models have been developed to explain this effect [72.33]. Under suitable conditions the harmonic orders can be suitably phased so that attosecond pulses are generated [72.34].

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