NONLINEAR OPTICS OF ULTRASHORT PULSES:THEORETICAL FUNDAMENTALS

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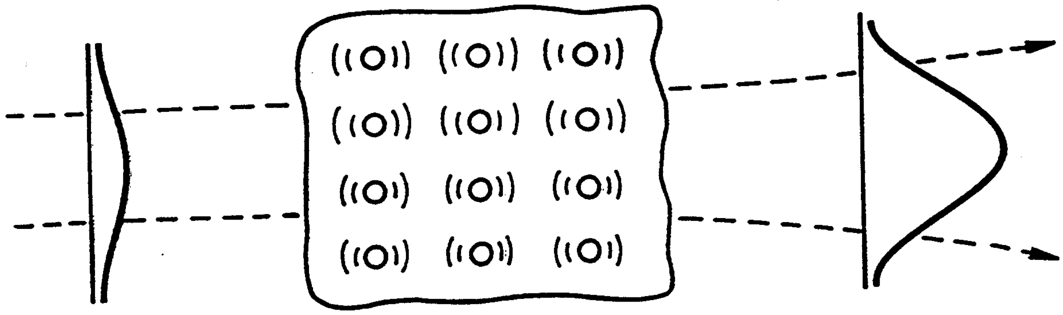
CONTENT:

REFERENCES

J. Herrmann, B. Wilhelmi: Lasers for Ultrashort Pulses
J-C. Diels, W. Rudolph: Ultrashort Laser Pulse Phenomena
S.A.Achmanov, V.A.Vysloukh, A.S.Chirkin: Optics of Femtosecond Laser Pulses

1. Basic equations



$$\vec{P}(\omega,\vec{E}(\omega))$$

Grundgleichung: $\boxed{\vec{\nabla}\times\vec{\nabla}\times\vec{E}+\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\vec{E}=+\mu_0\frac{\partial^2}{\partial t^2}\vec{P}}$

Polarisation: $\vec{P}=\vec{P}_{nr}+\vec{P}_{res}$

$$\vec{P}_{nr(\omega)}=\varepsilon_0\left[\chi^{(1)}_{(\omega)}E(\omega)+\chi^{(3)}_{(\omega=\omega_1+\omega_2+\omega_3)}\vdots E(\omega_1)E(\omega_2)E(\omega_3)+\ldots\right]$$

(isotropic media)

Dispersion:

$$\chi^{(1)}(\omega)=\chi^{(1)}(\omega_l)+(\omega-\omega_l)\left.\frac{d\chi^{(1)}}{d\omega}\right|_{\omega_l}+\frac{(\omega-\omega_l)^2}{2}\left.\frac{d^2\chi^{(1)}}{d\omega^2}\right|_{\omega_l}$$

Kerr - Effekt: $\chi^{(3)}(\omega=\omega_l-\omega_l+\omega_l)$

nichtlineare Brechzahländerung: $n=n_0+n_2|E(\omega)|^2$

$$n_2=\frac{3\chi'^{(3)}}{2n_0}$$

Third Harmonic generation: $\chi^{(3)}(\omega=\omega_1+\omega_1+\omega_1)$

Four-wave mixing $\chi^{(3)}(\omega=\omega_1+\omega_2-\omega_3)$

Slowly varying envelope approximation (SVEA)

$$\vec{E} = \frac{1}{2}\left[A(x,y,z,t)\vec{e}\exp\left[i(\omega_l t - k_l z)\right] + c.c\right]$$

$$\vec{P} = \frac{1}{2}\left[\tilde{P}(x,y,z,t)\vec{e}\exp\left[i(\omega_l t - k_l z)\right] + c.c\right]$$

$$k^2 = \frac{\omega^2}{c^2}\varepsilon(\omega) = \frac{\omega^2}{c^2}\left(1 + \chi^{(1)}_{(\omega)}\right), k_0'' = \left.\frac{d^2 k}{d\omega^2}\right|_{\omega_l}, v_g = \left(\frac{dk}{d\omega}\right)^{-1}$$

$$\eta = t - \frac{z}{v_g}, \quad n_0^2(\omega) = 1 + \chi^{(1)}_{(\omega)},$$

v_g - group velocity

$$\left|\frac{\partial A}{\partial z}\right| << k|A|, \quad \left|\frac{\partial A}{\partial \eta}\right| << \omega|A|,$$

K_0'' - group velocity dispersion (GVD)

$K_0'' > 0$ normal GVD

$K_0'' < 0$ anormal GVD

$$\boxed{\frac{\partial A}{\partial z} + \frac{i}{2k_l}\left(\frac{\partial^2 A}{\partial x^2} + \frac{\partial^2 A}{\partial y^2}\right) - \frac{i}{2}k_0''\frac{\partial^2 A}{\partial \eta^2} = -\frac{i\mu_0\omega_l^2}{2k_l}\tilde{P}_{nr}}$$

2. LINEAR OPTICS OF FEMTOSECOND PULSES

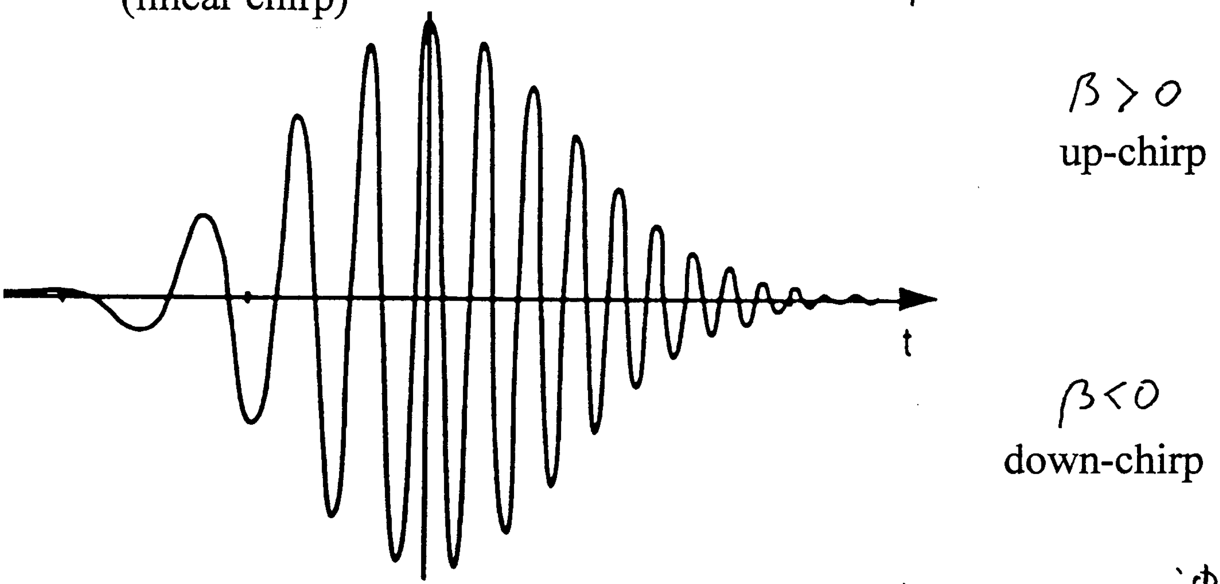
2.1 Characteristics of ultrashort pulses

Gaussian pulse shape of a phasemodulated (chirped) pulse

$$A(x,y,\eta) = A_0(x,y)\, \exp\{-\eta^2(1-i\beta)/\tau^2\}$$

$\tau_p = (2\ln 2)^{1/2}\tau$ - pulse duration, β- chirp parameter

instantaneous frequency: $\omega(\eta) = \omega_0 + \dfrac{d\varphi(\eta)}{d\eta} = \omega_0 + 2\beta\eta$
(linear chirp)



Fourier transformation $E(\omega) = \int_{-\infty}^{\infty} E(t)\, e^{i\omega t} = |E(\omega)|\, e^{i\Phi(\omega)}$

spectrum: $I(\omega) \sim |E(\omega)|^2 \sim \exp\left(-\dfrac{\omega^2\tau^2}{2(1+\beta^2)}\right)$

spectral phase $\Phi(\omega) = -\dfrac{\beta\tau^2\omega^2}{4(1+\beta^2)} + \dfrac{1}{2}\,\mathrm{arctg}\,\beta$

pulse duration-bandwidth product: $\Delta\nu_p \tau_p = c_B\sqrt{1+\beta^2}$
Gauss: $c_B = 0.441$, sech: $c_B = 0.315$

2.2 Linear propagation: dispersion and diffraction

Basic equation :

$$\frac{\partial A}{\partial z} + \frac{i}{2k_l}\left(\frac{\partial^2 A}{\partial x^2} + \frac{\partial^2 A}{\partial y^2}\right) - \frac{i}{2}k_0''\frac{\partial^2 A}{\partial \eta^2} = 0$$

diffraction dispersion

Input pulse and beam shape: gaussian

$$\boxed{A(x,y,z,\eta) = A_0\alpha(z)\exp\left[-\frac{ik_l r^2}{2q(z)} - \frac{\eta^2}{2}\left(\frac{1}{\tau_l^2(z)} - i\beta(z)\right) + i\varphi(z)\right]}$$

$$\frac{1}{q(z)} = \frac{1}{R(z)} - \frac{i\lambda}{\pi w^2(z)}, \quad \frac{1}{p(z)} = -\beta(z) - i\frac{1}{\tau_l^2(z)}, \quad r^2 = x^2 + y^2$$

$R(z)$ - Krümmungsradius, $w(z)$ - Bündelradius,

$\tau_l(z)$ - Impulsbreite, β - Chirpparameter

$$\frac{dq}{dz} = 1, \quad \frac{dp}{dz} = k_0''$$

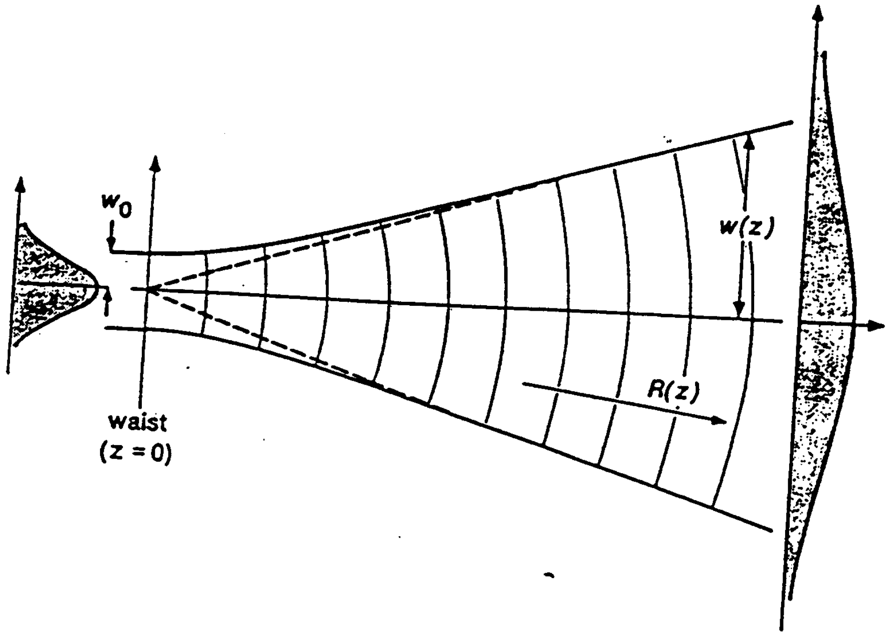
$$q = q_0 + z, \quad q_0^{-1} = -\frac{i\lambda}{\pi w_0^2} \nearrow \quad w^2(z) = w_0^2\left(1 + \frac{1}{2}\left(\frac{\lambda z}{\pi w_0^2}\right)^2\right)$$

$$R(z) = z\left(1 + \frac{1}{2}\left(\frac{\pi w_0^2}{\lambda z}\right)^2\right)$$

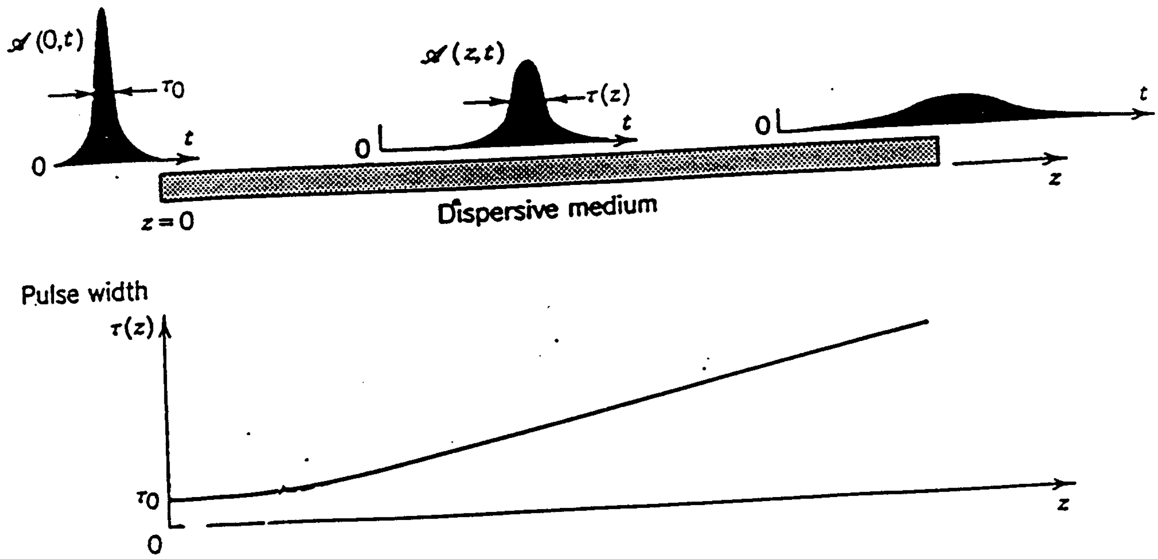
$$p = p_0 + k_0'' z, \quad \frac{1}{p_0} = -i\frac{1}{\tau_0^2} \rightarrow \quad \tau^2(z) = \tau_0^2\left(1 + \left(\frac{k_0'' z}{\tau_0^2}\right)^2\right)$$

$$\beta(z) = \frac{k_0'' z}{\tau_0^4\left(1 + \left(\frac{k_0'' z}{\tau_0^2}\right)^2\right)}$$

Beugung: $q = q_0 + z$



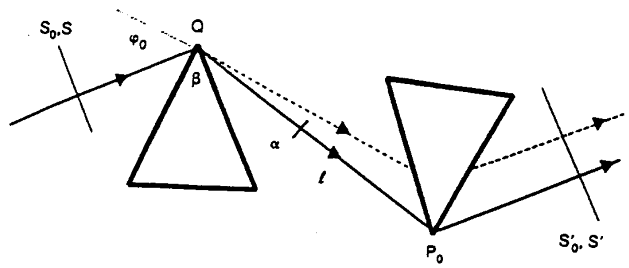
Dispersion: $p = p_0 + zk_0''$



2.3Phase control by optical elements with anomalous dispersion.

$$E_{out}(\omega) = |E_{in}(\omega)|\, e^{i\varphi(\omega)}\, T(\omega)\, e^{i\Psi(\omega)}$$

Prism sequency with anomalous GVD without net angular dispersion



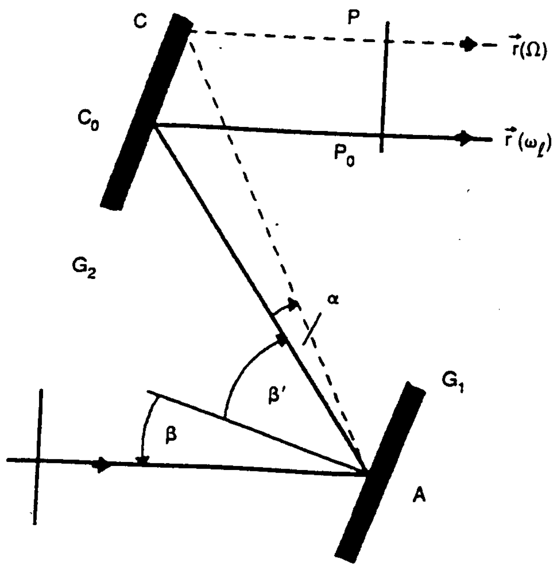
$$\Psi(\omega) = \frac{\omega}{c}\, l \cos\alpha$$

$$\frac{d\alpha}{d\omega} = \frac{d\alpha}{dn}\,\frac{dn}{d\omega}$$

Brewster condition:

—— ray for ω_0 - - - - ray for ω , $\frac{d\alpha}{dn} = -2$

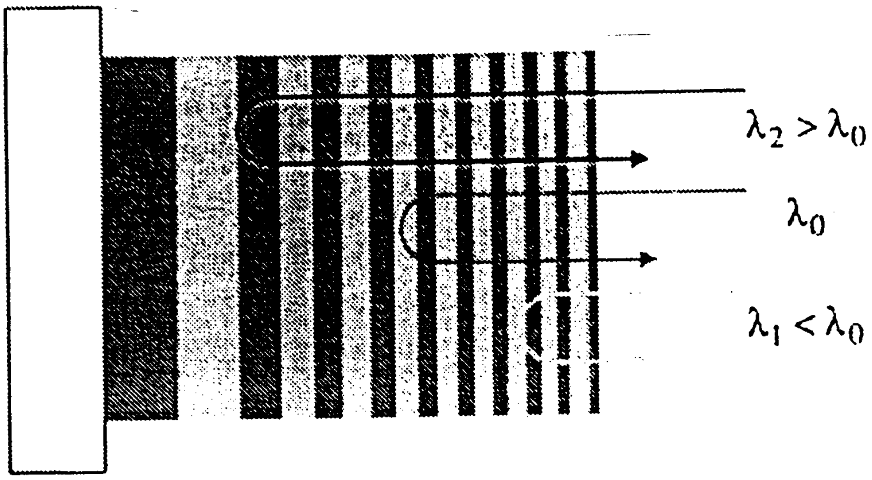
Parallel gratings with anomalous GVD without net angular dispersion

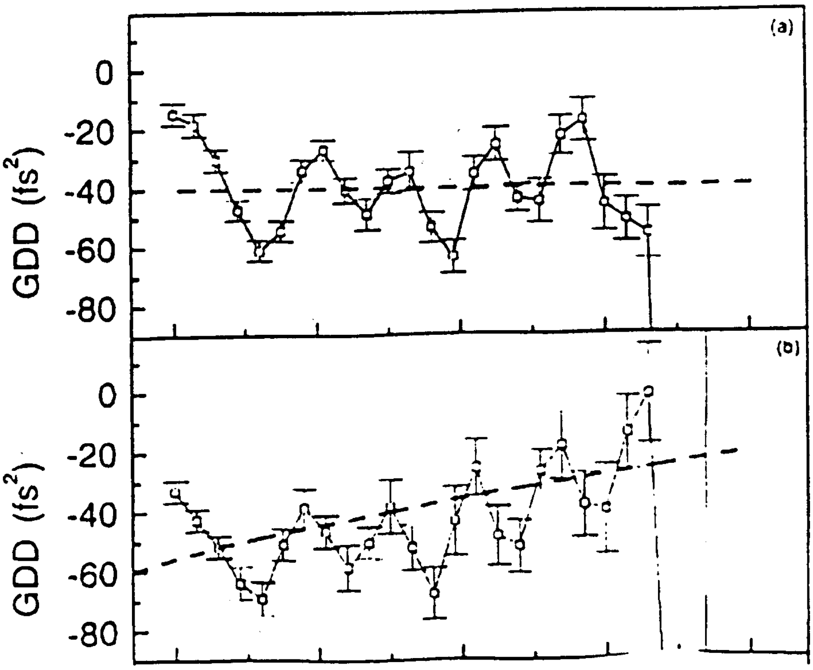


Examples for second and third order dispersion:

Device	λ_ℓ [nm]	ω_ℓ [fs^{-1}]	Ψ'' [fs^{-2}]	Ψ''' [fs^{-3}]
SQ1 ($L = 1$ cm)	620	3.04	550	240
	800	2.36	362	280
Brewster prism pair, SQ1	620	3.04	-760	-1300
$\ell = 50$ cm	800	2.36	-523	-612
grating pair [illegible] $= 20$ cm; $\beta = 0°$	620	3.04	-8.2 10^4	1.1 10^5
$d = 1.2$ μm	800	2.36	-3 10^6	6.8 10^6

Dispersion by chirped multilayer mirrors





Spatial light modulators (SLM):
Phasemodulation and adaptive control

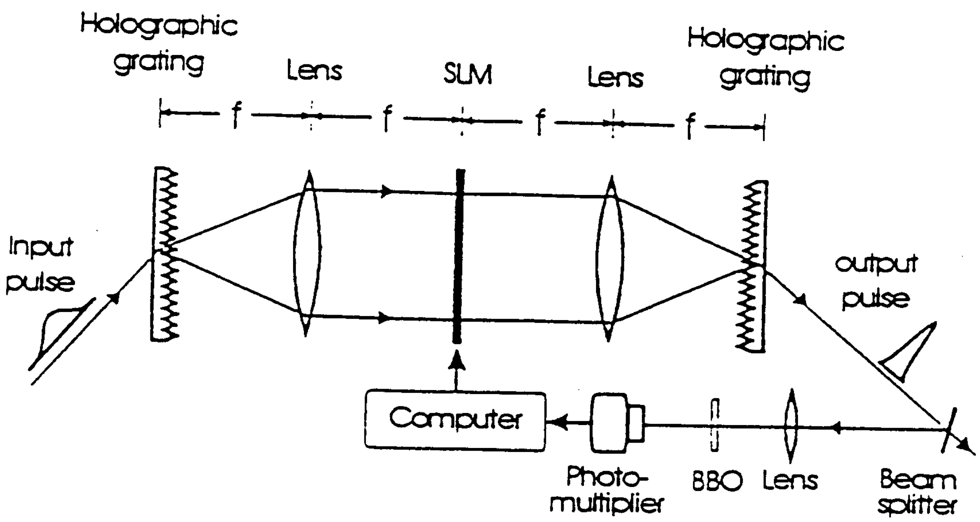


Figure 2. Schematic layout of the experimental setup for adaptive femtosecond pulse compression. The setup is composed of a programmable 4-f pulse shaper, a feedback measurement arrangement and a computer.

3. Nonresonant nonlinear processes

3.1 Optical Kerr effect in isotropic media

$$\boxed{\frac{\partial A}{\partial z}+\frac{i}{2k_l}\left(\frac{\partial^2 A}{\partial x^2}+\frac{\partial^2 A}{\partial y^2}\right)-\frac{i}{2}k_0''\frac{\partial^2 A}{\partial \eta^2} = -i\frac{\omega_l}{c}\Delta n\left(|A|^2\right)A}$$

Kerr - Effekt: $\Delta n\left(|A|^2\right)=n_2|A|^2$

Selbstphasenmodulation

$$i\frac{\partial A(\eta)}{\partial z} = \kappa |A|^2 A(\eta)$$

$$\begin{aligned} A(z,\eta) &= A_0(\eta)\exp\left(-i\kappa|A_0(\eta)|^2 z\right) \\ &\approx A_0(\eta)\exp\left(-i\kappa z|A_0^{max}|^2\left[1-\beta\eta^2+\ldots\right]\right) \end{aligned}$$

spectral broadening

$$\Delta\omega = \Delta\omega_{in} \cdot \kappa\, |A_{0max}|^2 \cdot L$$

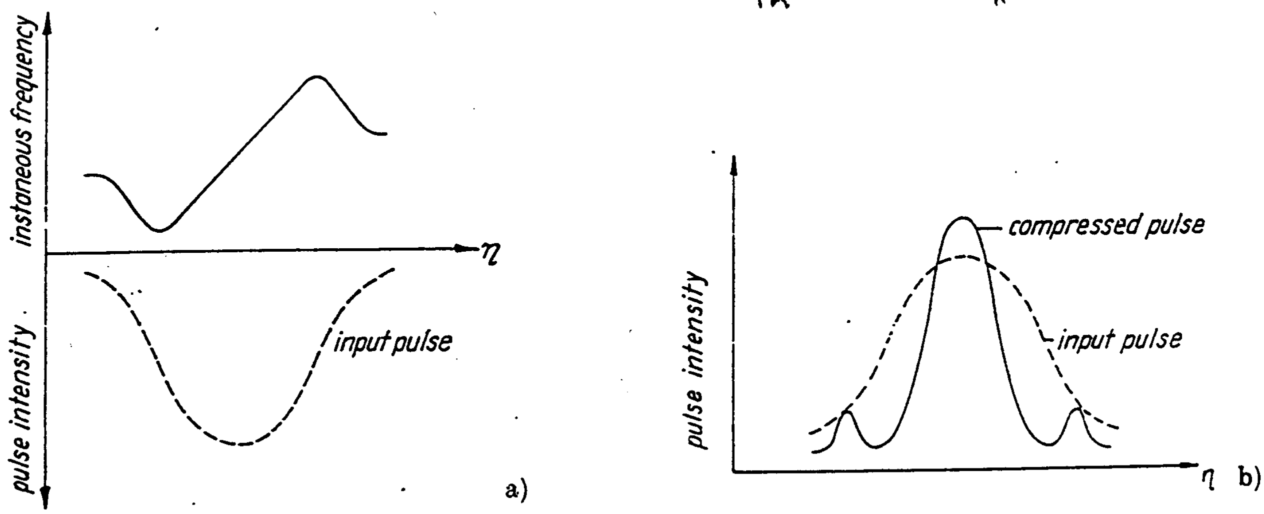


Fig. 8.12.
a) Chirp generation in a nonlinear optical medium
b) Compression of a chirped pulse after passing through a linear dispersive medium

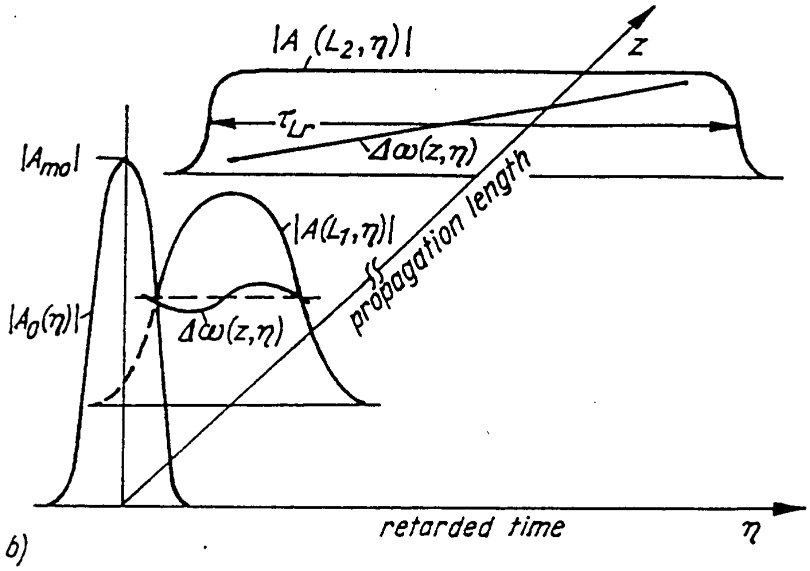
Dispersives nichtlineares Medium ($k_l'' > 0$)

$$i\frac{\partial A}{\partial z} = -\frac{k_l''}{2}\frac{\partial^2 A}{\partial \eta^2} + \kappa |A|^2 A$$

$\tau_L \approx 2.9 \sqrt{k_l'' \varkappa |A_0|^2}$

$\beta \to \frac{1}{k_0'' z}$

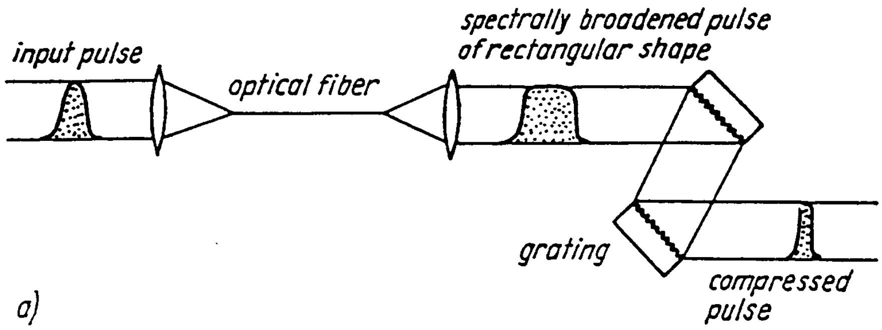
(linear chirp)



b)

Fig. 8.13.
a) Scheme of an experimental arrangement for pulse shortening
b) Pulse evolution

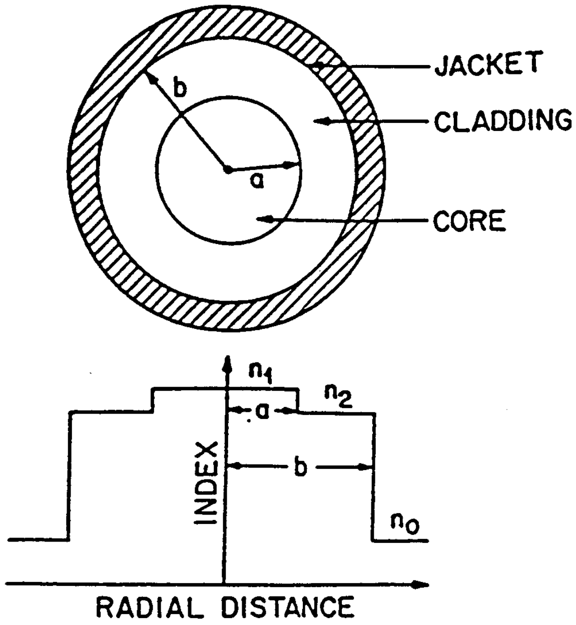
Impulskompression

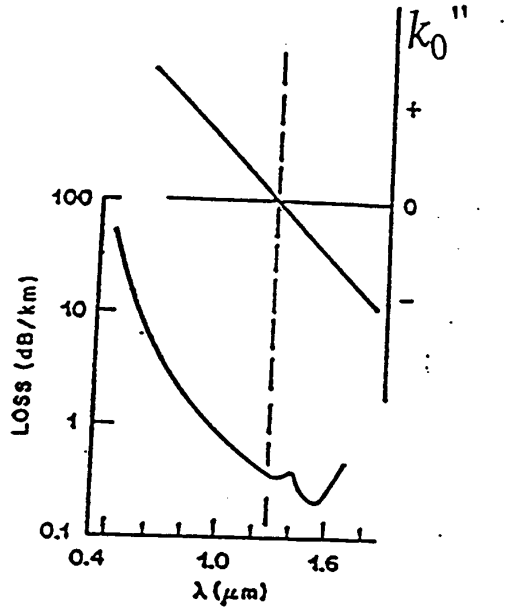


a)

$z_{opt} \approx 2 \frac{\tau_{L0}}{\sqrt{\varkappa |A_0|^2 k_l''}}$, $(\tau_L)_{comp.} \approx 2 \sqrt{\frac{k_l''}{\varkappa |A_0|^2}}$

Optische Solitonen





Hasegawa, Tappert (1973), Mollenauer u.a. (1981)

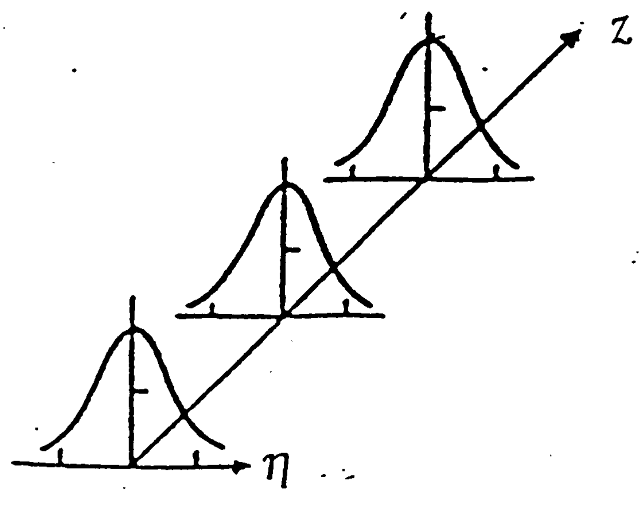
$\Delta n = n_2 |A|^2$, $k_0'' < 0$: $A(z,\eta) = e^{i\beta z}\varphi(\eta)$

$$\boxed{\varphi = A_s \operatorname{sech}\left(\frac{\eta}{\tau_l}\right)}$$

Soliton ($N = 1$)

$$A_s = \frac{1}{\tau_l}\sqrt{\frac{|k_0''|n_0}{n_2 k_l}}$$

$\tau_l = 10\,ps, P_s \approx 1W$



Self-Focusing

Quasi-steady state self-focusing for ps and ns-pulses:

$$\frac{\partial A}{\partial z}+\frac{i}{2k_l}\left(\frac{\partial^2 A}{\partial x^2}+\frac{\partial^2 A}{\partial y^2}\right)=-i\kappa|A|^2 A$$

Analytical approximation: $A(r,z)=\frac{A_0 w_0^2}{w^2(z)}\exp\left[-\frac{r^2}{2}\left(\frac{1}{w^2(z)}+i g(z)\right)\right]$

Beam radius: $w^2(z)=w_0^2\left[1+\frac{z^2\lambda^2}{2\pi^2 w_0^4}\left(1-P_0/P_{cr}\right)\right]$

Critical power of self-focusing: $P_{cr}=c\lambda^2/16\pi^2 n_2$

P_0 - input power

$P_0 < P_{cr}$ diffraction dominates

$P_0 > P_{cr}$ self-focusing dominates

Catastrofic self-focusing: $w(z)\rightarrow 0$

Fokal length: $z_f=\sqrt{2}\pi\lambda^{-1}\left(P_0/P_{cr}-1\right)^{-1/2}$

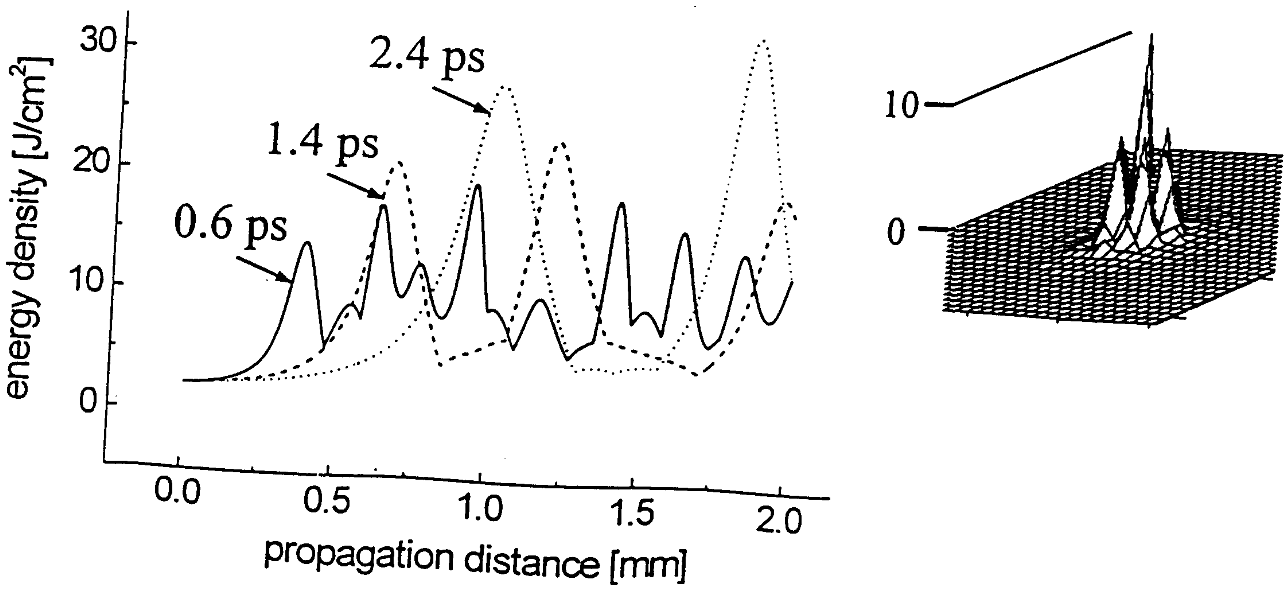
Moving Fokus model: $P_0=P_0(\eta)$ (long pulses)

Self-focusing depends on time:

leads to pulse compression and phasemodulation

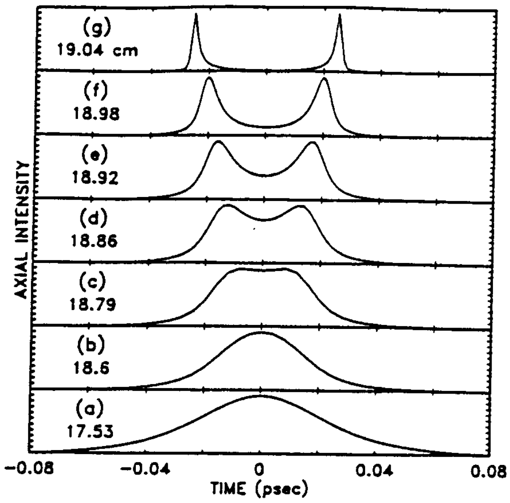
Filamentation and spontaneous symmetry-breaking

(Real physical behavior determined by transverse instabilities)



Self-focusing of fs -pulses

Influence of dispersion: temporal broadening, increase of the critical power of self-focusing and pulse splitting in dependence on the pulse duration



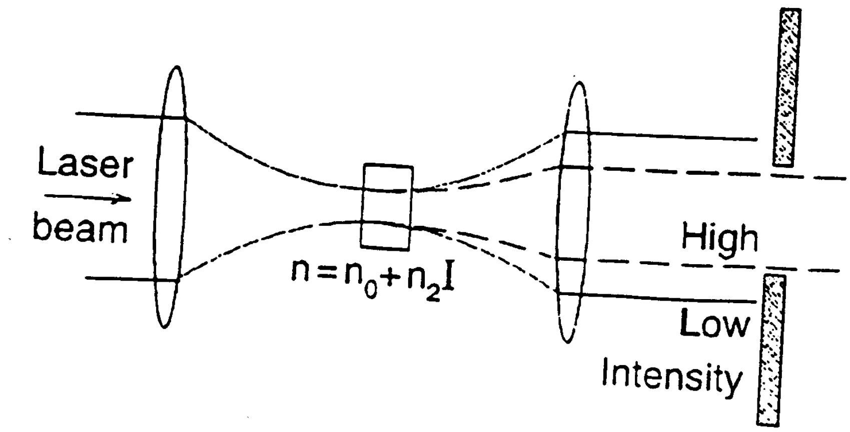
Kerr Lens Effect

radial dependence of the intensity leads to an intensity-depending lens effect: $\Delta n = n_2 I_0 (1 - r^2/2w^2)$

Focal length of a Kerr-lens: $\frac{1}{f_K} = \alpha\, n_2 I_0 L / w^2$

α numerical factor : $\alpha \sim 2 \ldots 8$

Physical basis for Kerr-lens modelocking: Intensity-depending (saturable) loss at an aperture (or intensity-depending gain due to gain guiding): $\alpha(I) = \alpha_0 - \gamma I(r)$



Self-focusing-induced changes in the transmittivity

3.2 Second harmonic generation

Anisotropic media: second order nonlinearity ($\chi^{(2)} \neq 0$)

Basic equations:

$$\left(\frac{\partial}{\partial z} + \frac{1}{v_1}\frac{\partial}{\partial t}\right) A_1 = -i\Delta^{(2)}(\omega_1, 2\omega_1, -\omega_1)\, A_2 A_1^* \, e^{-i\Delta k z}$$

$$\left(\frac{\partial}{\partial z} + \frac{1}{v_2}\frac{\partial}{\partial t}\right) A_2 = -i\Delta^{(2)}(2\omega_1, \omega_1, \omega_1)\, A_1^2 \, e^{i\Delta k z}$$

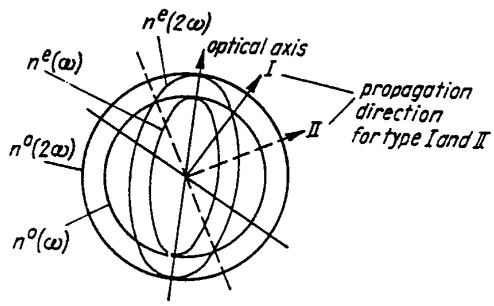
Phase mismatch

$$\Delta k = k_2 - k_1 = \frac{2\omega}{c}\left(n(\omega_2) - n(\omega_1)\right) \approx 0$$

Bire-fringent crystal

$$n^e(\theta, 2\omega_1) = n^o(\omega_1)$$

$$n^o(2\omega_1) = n^e(\theta, \omega_1)$$



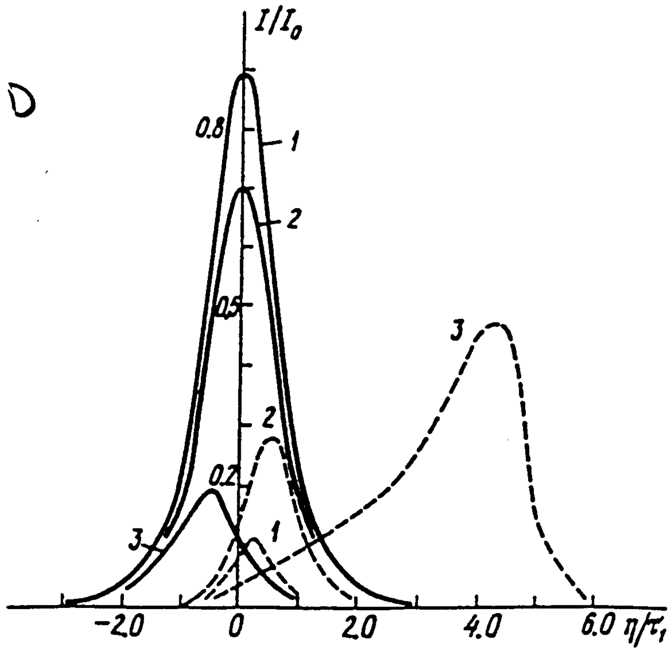
Solution for small signal amplification

$$A_2\left(t - \frac{z}{v_2}, z\right) = -i\Delta^{(2)} \int_0^z A_1^2\left(t - \frac{z}{v_2} + D\frac{z'}{c}\right) e^{i\Delta k z'}\, dz'$$

group delay $D = c\left(\frac{1}{v_2} - \frac{1}{v_1}\right)$

walk-off length $L_w = c\tau_1 / D$

Numerical solution



Shapes of the pulses I_1/I_0 of fundamental frequency (solid lines) and I_2/I_0 (dashed lines) of SH for $L_w = L_{conv}$ and different lengths z/L_{conv} : [1]

3.3 Freqency mixing, parametric three- and four-photon interaction

Second-order nonlinear crystal: $\chi^{(2)}(\omega, \omega_1, \omega_2) \neq 0$

Sum- and difference frequency generation

input pulses with frequencies ω_1, ω_2, output pulse with $\omega_3 = \omega_1 \pm \omega_2$
phase matching condition: $\Delta k = k_3 - k_1 - k_2 \approx 0$

Parametric amplification:

ω_p- input pump pulse, ω_s- weak signal (or noise), ω_I- idler pulse:

$$\frac{\partial A_s}{\partial z} + \frac{1}{v_s}\frac{\partial A_s}{\partial t} = -i\,\Delta_s^{(2)} A_p A_I^* e^{i\Delta k z}$$

$$\frac{\partial A_I}{\partial z} + \frac{1}{v_I}\frac{\partial A_I}{\partial t} = -i\,\Delta_I^{(2)} A_p A_s^* e^{i\Delta k z}$$

$$\frac{\partial A_p}{\partial z} + \frac{1}{v_p}\frac{\partial A_p}{\partial t} = -i\,\Delta_p^{(2)} A_I A_s e^{-i\Delta k z}$$

phase matching determines the direction of idler:

$$\vec{k}_p = \vec{k}_s + \vec{k}_I - \Delta\vec{k}$$

group velocity mismatch effect

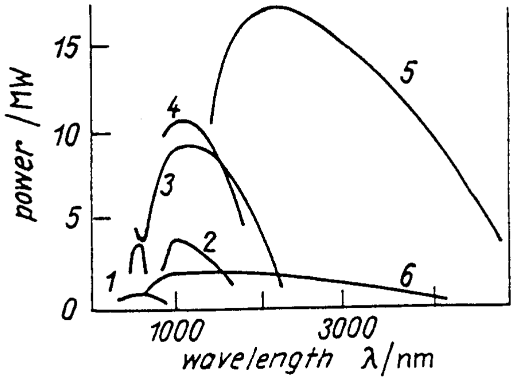
$$D_s = c\left(\frac{1}{v_p} - \frac{1}{v_s}\right), \quad D_I = c\left(\frac{1}{v_p} - \frac{1}{v_I}\right)$$

signal wave:

$$A_p(z,t) \approx A_{p0}(\eta_p), \quad \eta_p = t - \frac{z}{v_p}$$

$$A_s(\eta_s, z) = A_{s0}(\eta_s) \cosh\left[\Delta^{(2)} \int_0^z A_{p0}\left(\eta_s + D_s \frac{z'}{c}\right) dz'\right]$$

Frequency detuning by parametric amplification



Tuning range of a parametric generator

Four-wave mixing

third-order nonlinearity: $\chi^{(3)} \neq 0,\quad \chi^{(2)} = 0$

third harmonic: $\chi^{(3)}(3\omega_1, \omega_1, \omega_1, \omega_1)$: $\omega_1 + \omega_1 + \omega_1 \rightarrow 3\omega_1$

four-photon interaction: $\chi^{(3)}(\omega_1, -\omega_2, \omega_3, \omega_4)$: $\omega_1 + \omega_2 \rightarrow \omega_3 + \omega_4$

degenerate four-wave mixing: $2\omega_1 \rightarrow \omega_3 + \omega_4$

colinear and noncolinear interaction:

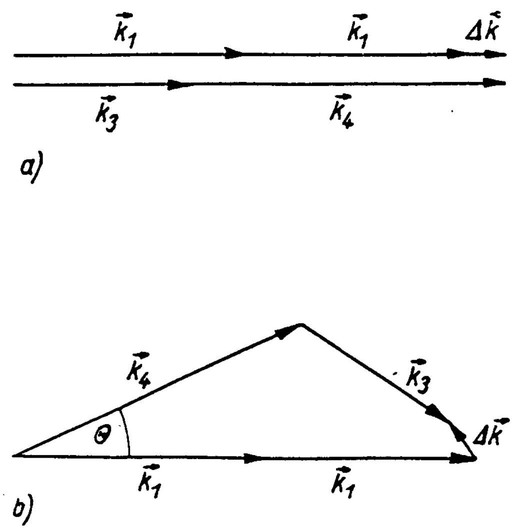


Fig. 8.6. Phase matching condition of the parametric four photon interaction

3.4 Ionization and damage

Strong-field nonlinear optics (non-perturbativ)
Perturbative Taylor expansion of the polarization becomes invalid for high intensities ($I > 10^{13}\,\mathrm{W/cm^2}$) due to ionization

Free electron density N_e(plasma)

$$\frac{\partial N_e}{\partial t} = \underbrace{(N_a - N_e)\, W(E(t))}_{\text{(optical field ionization)}} + \underbrace{N_e f |E|^2}_{\text{(avalanche ionization)}}$$

$W(E(t))$-ionization rate, E_I-ionization energy, N_a - atom density
electron collision time - τ_c , $f = \frac{e^2 \tau_c}{2 m e E_I} (1+\omega^2 \tau_c^2)^{-1}$

neglect of avalanche ionization for short pulses:

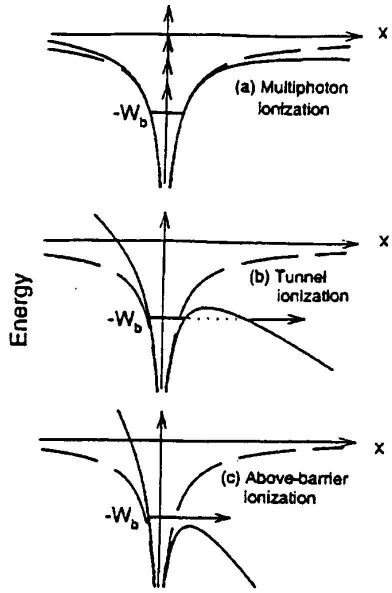
$$N_e = N_a \left(1 - \exp\left[-\int_{-\infty}^{t} dt'\, W(E(t'))\right]\right)$$

Regimes of optical ionization

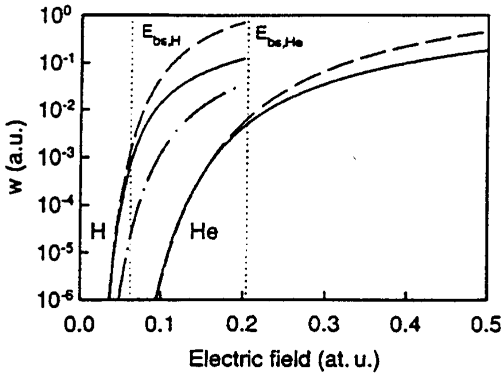
Keldysh parameter: $\gamma = \dfrac{\omega \sqrt{2m E_I}}{e E_0}$

$\gamma > 1$: n-photon ionization (Keldysh formula)
$\gamma < 1$ quasi-static tunneling ionization (Amosov-Delone-Krainev formula)
$\gamma \ll 1$ above barrier ionization (suppression of the Coulomb barrier below the ground state)

regimes of optical ionization



ionization rates in different models

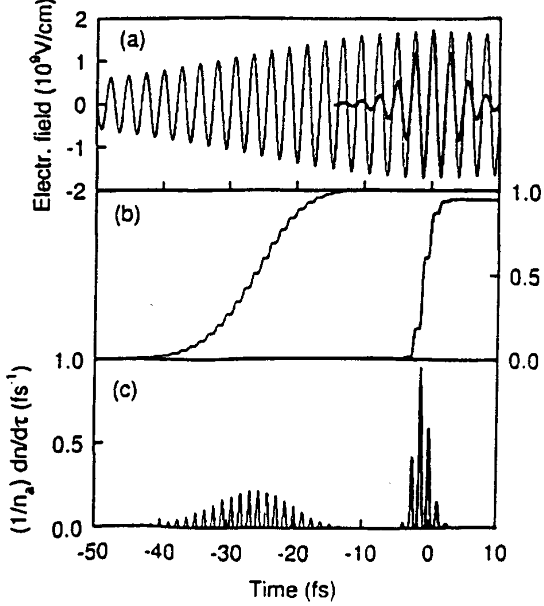


numerical

Amosov - Delone

Keldysh

fraction of ionized electrons and ionization rate



$E(t)$

$N_e(t)/N_a$

$W(E(t))$

plasma contribution to polarization

Free electron motion: $\frac{d^2x}{dt^2} + \frac{1}{\tau_c}\frac{dx}{dt} = \frac{e}{m}E(t)$

Polrization $\dot{P}_{free} = J_{free} = e\,N_e(E(t))\,\dot{x}$

Wave equation $2ik\frac{\partial A}{\partial z} + kk''\frac{\partial^2 A}{\partial t^2} - \left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right)A - 2\,\frac{k^2 n_2}{n_0}|A|^2 A$

$$+\frac{k^2}{n_0}(1 + i\omega_0\tau_c)\frac{N_e}{N_{cr}}A = 0$$

$N_{cr} = \frac{\omega_0^2 m^* \varepsilon_0}{e^2}$

N_{cr}-critical plasma density, ω_0 central frquency

Propagation effects of plasma electrons :

contribution to the refractive index: $\Delta n_{pl} = -\frac{1}{2}\,N_e(E(t))/N_{cr}$

Linear dispersive plasma effects : $N_e = N_e^0 = const$

shift of group velocity and GVD: $\Delta n_{pl} = -\frac{1}{2}\,\omega_{pl}^2/\omega^2$

$\frac{\partial \Delta k}{\partial \omega}\Big|_{\omega_0} = \omega_{pe}^2/\omega_0^2 c$

$\omega_{pl}^2 = e^2 N_e^0/m^* \varepsilon_0$

anomalous GVD contribution

$$k''_{pl} = \frac{\partial^2(\Delta k)}{\partial \omega^2}\Big|_{\omega_0} = -\,\omega_{pe}^2/c\,\omega_0^3$$

Nonlinear plasma effects : $\Delta\varphi = -L\omega_0 N_e(E(t))/c$

$N_e(t) \approx N_a \int^t W(E(t'))dt'$

(weak ionization : $N_e \ll N_a$)

blue-shift $\omega(t) = \omega_0 - L\frac{\omega_0}{c} N_a \cdot W(E(t))$

self-phase modulation (opposite to Kerr-effect)

$\Delta\varphi = \Delta\varphi(t)$

self-defocusing (opposite to Kerr-effect) : $\Delta\varphi = \Delta\varphi(t,x,y)$

self-channeling of fs-pulses: $\Delta n_{Kerr} \approx \Delta n_{pl}$

Optical damage : $N_e \approx N_{cr} = 1.6 \cdot 10^{21}\ cm^3$

→ damage fluence