Pico- and femtosecond pulses in the UV and XUV

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ABSTRACT

Ultrashort pulses in the shortwavelength region are indispensable tool for studying of the molecular dynamics. The compression of nanosecond UV pulses from excimer lasers and the four-wave mixing of femtosecond and picosecond pulses, both in the UV, is modeled.

1.THEORETICAL DESCRIPTION

1.1.Compression of excimer laser pulses

In the first method we consider the induced phase modulation (IPM)[1,2] in a gas-filled hollow core waveguide. The sum of the pump ω_P and signal ω_S frequency is near a two-photon resonance on the high frequency side of the transition. In this way the nonlinear susceptibility for induced phase modulation $\chi^{(3)}_{IPM}(\omega_S)$ can be kept negative, thus avoiding the induced focusing of the signal. The refractive index at the signal frequency is influenced by the pump pulse according to the relation

$$n(\omega_S) = n_O(\omega_S) + n^{NL}_{IPM}(\omega_S)\langle A_P\rangle^2 \qquad (1)$$

Since the two-photon resonances in an inert gas are high lying, ω_S and ω_P can be selected far from single-photon resonances, which reduces the absorption. The signal wavelength is selected to coincide with widely used UV, VUV and XUV sources and the respected pump wave can be obtained by a dye or other lasers.

The IPM on the pump from the signal ($I_S \ll I_P$) and pump self-modulation and depletion are considered to be negligible. The evolution of the slowly varying signal is given by the characteristic Schrödinger equation

$$i\frac{\partial\psi_S}{\partial x} + \alpha_S\frac{\partial^2\psi_S}{\partial\tau^2} + k^{IPM}(\omega_S)\,|\psi_P|^2\psi_S = 0 \quad , \qquad (2)$$

where $\alpha_S = (-1/2)[\lambda_S^3/(2\pi c^2)][\partial^2 n/\partial\lambda^2]_{\lambda=\lambda_s}$ is a coefficient, determined by the group velocity dispersion, and $k^{IPM}(\omega_S) = 6\pi^2 N\chi^{(3)}_{IPM}(\omega_S)/[\lambda_S n_O^2(\omega_S)]$ is a nonlinear coefficient (N is the particle density). The pump and probe pulses are approximated by Gaussian functions of the type

$$\psi_s(x,\tau)=A_s(x)\exp\left[[-\tau^2/2a_s^2(x)]+ib_s(x)\tau^2\right] \tag{3}$$

where $a_s(x)$ is the pulse half-width at 1/e level, $b_s(x)$ is the frequency-chirp parameter, A_s is the complex amplitude. Using the variational approach, Eq.(2) is transformed to a set of ordinary differential equations, describing the evolution of the signal pulse[3]

$$\frac{da_s}{dx} = 4\alpha_s a_s b_s \tag{4a}$$

$$a_s\frac{db_s}{dx}=-4\alpha_s a_s b_s^2 + \frac{\alpha_s}{a_s^3} - k^{IPM}(\omega_s)\,|A_p|^2\frac{a_p/a_s}{\sqrt{a_s^2 + a_p^2}}\exp\left\{-\frac{(\tau_D-x\nu_{SP})^2}{a_s^2 + a_p^2}\right\}$$

$$\times\left\{1 - \frac{a_p^2}{a_s^2 + a_p^2} - \frac{2a_s^2(\tau_D-x\nu_{SP})^2}{(a_s^2 + a_p^2)^2}\right\} \tag{4b}$$

In Eq.(4a)-(4b) a_p is the pump pulse duration, τ_D is the initial delay between the pulses and $\nu_{SP}=\left|v_{gs}^{-1}-v_{gP}^{-1}\right|$ is determined by the group velocity mismatch.

In order to illustrate the method, let us consider the specific case of some widely used UV ω_s excimer laser sources at 157 nm, 193 nm, 248 nm (Table 1). The pump source is chosen in such a way, that $\omega_s+\omega_p$ is near the $5p^6[^1S_0]$-6p' two-photon transition in Xe. The waveguide is a hollow core capillary with a length l=16 cm and internal diameter 150 μm. The parameters used in the model of the IPM are presented in Table 1. Included in the model are the single-photon absorption in the medium $\sigma^{(1)}(\omega)$ and the waveguide losses of the beam. The main limitation for the maximum pump power is imposed by multiphoton ionization and breakdown in the gas. As shown in Fig.1, the compression coefficients obtainable for nanosecond input pulses and reasonable gas pressure (1-5 atm) range from 15 to 700.

1.2. Up-conversion of femtosecond laser pulses to the XUV

In the second approach we have studied the generation of femtosecond pulses in the XUV using a noncolinear phase-matching, taking into account the influence of the IPM on the different aspects of the up-conversion[4]. A noncolinear phase-matched process $\omega_s=2\omega_1+\omega_2$ is considered. Using again the variational method, we find an equation for the temporal history of the pulses

$$i\frac{\partial\psi_S}{\partial x} + \alpha_S\frac{\partial^2\psi_S}{\partial\tau^2} + k^{IPM}(\omega_S)\,|\psi_P|^2\psi_S + i\beta\frac{\partial}{\partial\tau}\left[|\psi_P|^2\psi_S\right] = 0 \quad , \qquad (5)$$

where the term comprising β reflects the shock-wave of the envelope. In a model calculations we consider femtosecond pulses from excimer amplifiers as a last stage at λ_2=248 nm (τ_2(z=0)=60 fs) and λ_1=249.6 nm (second harmonic of a dye laser). Some important parameters, included in the model, are presented in Table 2. The maximum pump powers are limited by selffocusing and/or breakdown in the gas. The phase-matching angles ϑ_1 and ϑ_2 satisfy the conditions for waveguide propagation. The signal pulse at λ_s=83nm has a duration of τ_s= 200 fs and intensity $I_s=1.9.10^5$ W/cm^2. Fig.2 shows the influence of the pumping femtosecond pulse on the signal. The trailing edge of the signal pulse is accelerated and steepened.

2.CONCLUSION

In conclusion, the induced phase modulation can influence to a great extend not only the temporal, but also the spatial characteristics of the signal.

3.REFERENCES

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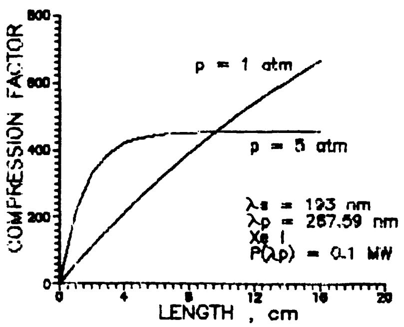

Fig.1 Dependence of the compression factor on the length for dîfferent densities of the Xe I - gas

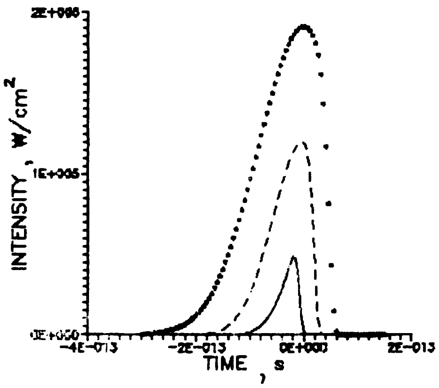

Fig.3 Signal intensity vs. time including pump-induced steepening (z=1 cm /solid line/, 2 cm /dashed line/ and 3 cm /dotted line/)

Table 1. Parameters for the induced phase modulation and compression model

Medium Xe I ; N = $2.5.10^{19}$ cm^{-3}				
Wavelength [nm]	λ_s = 157	λ_p=404.75	λ_s= 193	λ_p=262.69
Pulse duration (FWHM)	6 ns	9 ns	11 ns	18 ns
Resonance	$5p^6$ − 6p'$[3/2]_1$		$5p^6$ − 6p'$[3/2]_2$	
$\chi^{(3)}_{IPM}$ [esu]	$-7.8.10^{-32}$		$-8.1.10^{-32}$	
$\chi^{(3)}_{SPM}$ [esu]	$1.1.10^{-35}$	$3.5.10^{-36}$	$2.4.10^{-35}$	$2.7.10^{-35}$
$\sigma^{(1)}(\omega)$ [cm^2]	$6.14.10^{-22}$	$2.73.10^{-21}$	$4.43.10^{-22}$	$8.79.10^{-22}$
α_s [s^2/cm]	$9.9.10^{-28}$		$-1.46.10^{-28}$	
α_p [s^2/cm]	$-2.55.10^{-27}$		$-4.66.10^{-28}$	
$k_{IPM}(\omega_s)$ [esu]	$3.7.10^{-6}$		$1.7.10^{-6}$	
$k_{SPM}(\omega_p)$ [esu]	$-2.9.10^{-11}$		$-3.0.10^{-10}$	
$P(\lambda_p)$ [W]	10^6		$1.7.10^5$	
Compression ratio F	766		664	

Table 2. Model parameters for conversion of femtosecond pulses in the XUV (λ =83.02 nm) and the induced shock wave

Parameter	Unit	Value	
		λ_1 = 249.6 nm	λ_2 = 248 nm
τ (FWHM)	s	2.10^{-11}	6.10^{-14}
$\sigma^{(1)}$	cm^2	$7.47.10^{-22}$	$7.37.10^{-22}$
I	W/cm^2	$4.7.10^6$	10^8
$\chi^{(3)}_{SPM}$	esu	$8.5.10^{-34}$	$-6.6.10^{-35}$
$\chi^{(3)}_{IPM}(\omega_s,\omega_p)$	esu	$-1.5.10^{-30}$	-9.10^{-29}
$\chi^{(3)}_{MIX}$	esu	$5.4.10^{-33}$	
λ_s	nm	83.02	
z	cm	3	
N	cm^{-3}	$2.5.10^{19}$	
I_s	W/cm^2	$1.91.10^5$	
τ_s (FWHM)	s	2.10^{-13}	
ϑ_1	deg	4.3	
ϑ_2	deg	1.5	