Modeling the Induced-Phase Modulation and Compression of UV Laser Pulses

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Abstract—**The induced-phase modulation in a gas-filled hollow-core waveguide is modeled as a possible method for pulse compression in the UV using grating pair. The method has been applied to a number of excimer lasers and Raman-shifted wavelengths, predicting compression coefficients from 15 to 700. The limitations posed by system parameters, absorption, waveguide losses, and breakdown are discussed.**

I. Introduction

Alfano and co-workers [1], [2] studied the spectral broadening of the second harmonic of Nd : glass laser in the presence of the primary pulse in BK-7 glass both theoretically and experimentally. Since then, several groups have been studying the cross-phase modulation (XPM and IPM) effects [3]–[8].

In this paper we model the pulse-compression method to the UV region using the effect of the induced-phase modulation (IPM, also commonly called XPM) of a pump pulse and a relatively weak signal pulse at resonance using a grating pair. A gas-filled hollow-core waveguide is modeled where the sum of the pump and signal frequency is near a two-photon resonance. Several excimer laser wavelengths are taken into account, and the system parameters, including capillary length, gas pressure, detuning from resonance, absorption, waveguide losses, and breakdown, are discussed. A large compression coefficients ranging from 15 to 700 are predicted for nanosecond pulses.

II. Description of the Proposed Method

Let us consider the propagation of two pulses, pump I_P and probe signal I_S in a waveguide with cubic nonlinearity. The refractive index at the signal frequency can be presented in the form [9]

$$n(\omega_S) = n_0(\omega_S) + n^{\mathrm{NL}}_{\mathrm{SPM}}(\omega_S)\langle A_S\rangle^2 + n^{\mathrm{NL}}_{\mathrm{IPM}}(\omega_S)\langle A_P\rangle^2 \quad (1)$$

where

$$n^{\mathrm{NL}}_{\mathrm{SPM}}(\omega_S) = \frac{3\pi N\chi^{(3)}_{\mathrm{SPM}}(-\omega_S;\ \omega_S,\ -\omega_S,\ \omega_S)}{n_0(\omega_S)} \quad (2a)$$

and

$$n^{\mathrm{NL}}_{\mathrm{IPM}}(\omega_S) = \frac{6\pi N\chi^{(3)}_{\mathrm{IPM}}(-\omega_S;\ \omega_S,\ \omega_P,\ -\omega_P)}{n_0(\omega_S)}. \quad (2b)$$

N is particle density. Our calculation of the nonlinear susceptibility for self-phase modulation $\chi^{(3)}_{\mathrm{SPM}}$ in xenon in the VUV and XUV is shown in Fig. 1. Using the existing coherent sources [9], [10] with relatively low peak powers, the bandwidth of the chirp obtained is not enough for compression of pulses in the ns and ps range [11]. The waveguide length usable in this part of the spectrum [12], [13] is limited by technical reasons to several centimeters. Therefore, high values of the nonlinear susceptibility are required for pulse compression.

In the vicinity of $\omega_S + \omega_P$ two-photon resonance $\chi^{(3)}_{\mathrm{IPM}}$ is determined mostly by terms of the type

$$\frac{r^2_{\mathrm{GA}}r^2_{\mathrm{AF}}}{(\omega_{\mathrm{GF}} - \omega_S - \omega_P - i\Gamma)}\{(\omega_{\mathrm{GA}} - \omega_S)^{-2} + (\omega_{\mathrm{GA}} - \omega_P)^{-2} + [(\omega_{\mathrm{GA}} - \omega_P)(\omega_{\mathrm{GA}} - \omega_S)]^{-1}\} \quad (3)$$

where r_{ij} denotes the corresponding dipole matrix element and Γ is the damping constant of the transition. Choosing $\omega_S + \omega_P > \omega_{\mathrm{GF}}$ and $\omega_S, \omega_P < \omega_{\mathrm{GA}}$, the nonlinear coefficient $\chi^{(3)}_{\mathrm{IPM}}(\omega_S) < 0$, thus avoiding induced focusing. Fig. 2 shows the wavelength λ_P dependence of $\chi^{(3)}_{\mathrm{IPM}}$ for $\lambda_S = 193$ nm in a part of the negative sign region of XeI.

The characteristic equation describing the evolution of the slowly varying signal is the Schrödinger equation (SE)

$$i\frac{\partial\psi_S}{\partial x} = \alpha_S\frac{\partial^2\psi_S}{\partial\tau^2} + k^{\mathrm{IPM}}(\omega_S)|\psi_P|^2\psi_S \quad (4)$$

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

$$k^{\mathrm{IPM}}(\omega_S) = -3\pi^2 N\chi^{(3)}_{\mathrm{IPM}}(\omega_S)/(\lambda_S n_0^2(\omega_S))$$

is a nonlinear coefficient. The Gaussian pulses ψ_S and ψ_P are considered with complex amplitudes $A_S(x)$ and $A_P(x)$, with the half-width at $1/e$ level $a_S(x)$ and $a_P(x)$, respectively, and frequency chirp parameter $b_S(x)$. The following conditions are used:

1) The self-phase modulation is negligible.
2) $I_S << I_P$, i.e., the phase modulation, induced by

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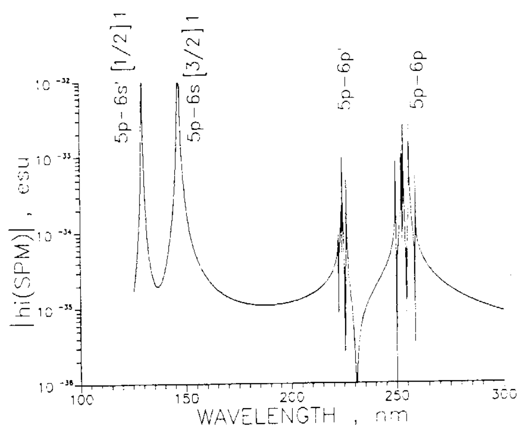

Fig. 1. Nonlinear susceptibility $\chi^{(3)}_{\mathrm{SPM}}$ versus wavelength for XeI.

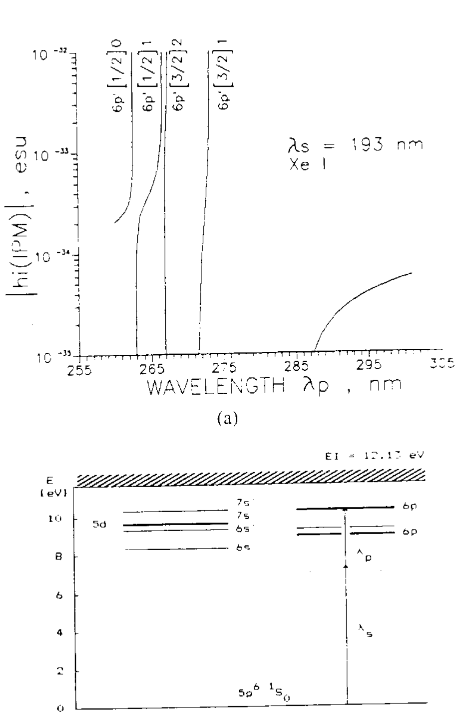

Fig. 2. (a) Nonlinear susceptibility $\chi^{(3)}_{\mathrm{IPM}}(\lambda_S = 193$ nm) versus pump wavelength λ_P for XeI (part of the region with negative sign only). (b) Simplified energy-level diagram for XeI.

the signal on the pump wave, is not to be considered.

3) The duration of the input pulses is approximately equal and the delay between the pulses is approximately zero.
4) The group-velocity mismatch between λ_S and λ_P is negligible and a single laboratory coordinate system can be used.
5) The four-wave mixing efficiency and the efficiency of the four-photon parametric processes is low, i.e., the pulse energy does not change.
6) The nonlinearity is fast compared to the pulse duration.

It has been shown [14] that in the case of single-pulse propagation in a waveguide both the variational method and the method of the NLSE invariants lead to the same characteristic equation describing the pulse evolution. Our analysis is based on the variational functional, corresponding to the SE [14]–[16]. Equation (4) can be restated as a variational problem corresponding to the Lagrangian L given by

$$L = (i/2)\left[\psi_S \frac{\partial \psi_S^*}{\partial x} - \psi_S^* \frac{\partial \psi_S}{\partial x}\right] - \alpha \left|\frac{\partial \psi_S}{\partial \tau}\right|^2 + k^{\mathrm{IPM}}(\omega_S)\,|\psi_S|^2 \cdot |\psi_P|^2 \tag{5}$$

and the Lagrange equation

$$\frac{\partial}{\partial x}\frac{\partial L}{\partial\left[\dfrac{\partial \psi_S^*}{\partial x}\right]} + \frac{\partial}{\partial \tau}\frac{\partial L}{\partial\left[\dfrac{\partial \psi_S^*}{\partial \tau}\right]} - \frac{\partial L}{\partial \psi_S^*} = 0 \tag{6}$$

is equivalent to the SE. Using this method we obtain a set of ordinary differential equations for $a_S(x)$ and $b_S(x)$:

$$a_S \frac{db_S}{dx} = 4\alpha_S a_S b_S^2 - \frac{\alpha_S}{a_S^3} + 2k^{\mathrm{IPM}}(\omega_S)\,|A_P|^2 \cdot \frac{a_P/a_S}{\sqrt{a_S^2 + a_P^2}}\left(1 - \frac{a_P^2}{a_S^2 + a_P^2}\right) \tag{7a}$$

$$\frac{da_S}{dx} = -4\alpha_S a_S b_S \tag{7b}$$

$$a_P \frac{db_P}{dx} = 4\alpha_P a_P b_P^2 - \frac{\alpha_P}{a_P^3} + k^{\mathrm{SPM}}(\omega_P)\,|A_P|^2/(2^{3/2}a_P) \tag{7c}$$

$$\frac{da_P}{dx} = -4\alpha_P a_P b_P. \tag{7d}$$

Equations (7c) and (7d) describe the pump pulse propagation in the waveguide [14]. In this way the pump pulse evolution can be described, proving that the pump self-modulation is negligible (condition 1)). The evolution of the probe pulse is given by (7a) and (7b) [16].

Calculations are performed at some widely used UV λ_S sources: 157 nm, 193 nm, 193.4 nm, 248 nm excimer laser; 206 nm – second anti-Stokes from 249 nm; 210 nm – first Stokes from 193 nm. λ_S and λ_P are chosen in a way that $\omega_S + \omega_P$ is near the two-photon resonance with the transitions $5p^6[^1S_0] - 6p'$ in XeI (see Fig. 2(a) and (b)). The waveguide modeled is a hollow-core capillary with a length $l = 16$ cm and internal diameter 150 μm, filled with 1 atm XeI. The signal and pump frequencies ω_S and ω_P are far from single-photon resonances in XeI. The $\chi^{(3)}_{\mathrm{SPM}}$ values are of the order of 10^{-35} esu; therefore, the self-phase modulation is negligible. The parameters used in the model of the induced phase modulation are presented in Table I(a)–(c). The nonlinear susceptibilities are calculated according to the single-sided Feynman diagrams [17]. The respective matrix elements for bound-

TABLE I
PARAMETERS FOR THE INDUCED-PHASE MODULATION AND COMPRESSION MODEL

Medium XeI: $N = 2.5 \times 10^{19}$ cm^{-3}				
Wavelength (nm)	$\lambda_S = 157$	$\lambda_P = 404.75$	$\lambda_S = 193$	$\lambda_P = 262.69$
Pulse duration (FWHM)	6 ns	9 ns	11 ns	18 ns
P_{MAX} (W)	540	2.4×10^6	250	1.7×10^5
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}
b (dB/cm)	1.08×10^{-3}	7.18×10^{-3}	1.63×10^{-3}	3.1×10^{-3}
Confocal parameter (cm)	9.2	3.6	7.5	5.4
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	
Wavelength (nm)	$\lambda_S = 193.4$	$\lambda_P = 266$	$\lambda_S = 248$	$\lambda_P = 207.99$
Pulse duration (FWHM)	11 ns	11 ns	20 ns	15 ns
P_{MAX} (W)	1550	1.2×10^5	4.4×10^4	3.1×10^6
Resonance	$5p^6 - 6p'(1/2)_1$		$5p^6 - 6p'(3/2)_1$	
$\chi^{(3)}_{IPM}$ (esu)	-2.0×10^{-32}		-1.49×10^{-32}	
$\chi^{(3)}_{SPM}$ (esu)	2.4×10^{-35}	3.1×10^{-35}	-6.6×10^{-35}	-1.4×10^{-35}
$\sigma^{(1)}(\omega)$ (cm^2)	4.44×10^{-22}	8.66×10^{-22}	7.37×10^{-22}	5.07×10^{-22}
b (dB/cm)	1.64×10^{-3}	3.10×10^{-3}	2.70×10^{-3}	1.90×10^{-3}
Confocal parameter (cm)	7.5	5.4	5.8	6.9
α_S (s^2/cm)	-1.47×10^{-28}		-3.57×10^{-28}	
α_P (s^2/cm)	-4.56×10^{-28}		-1.95×10^{-28}	
$k_{IPM}(\omega_S)$ (esu)	7.7×10^{-7}		4.4×10^{-7}	
$k_{SPM}(\omega_P)$ (esu)	-4.3×10^{-10}		2.6×10^{-10}	
$P(\lambda_P)$ (W)	1.2×10^5		10^5	
Compression ratio F	26		18	
Wavelength (nm)	$\lambda_S = 206$	$\lambda_P = 241.91$	$\lambda_S = 210$	$\lambda_P = 239.92$
Pulse duration (FWHM)	15 ns	13 ns	14 ns	13 ns
P_{MAX} (W)	10^4	1.4×10^5	2×10^3	1.3×10^5
Resonance	$5p^6 - 6p'(1/2)_0$		$5p^6 - 6p'(1/2)_1$	
$\chi^{(3)}_{IPM}$ (esu)	-1.1×10^{-32}		-5.3×10^{-32}	
$\chi^{(3)}_{SPM}$ (esu)	-1.4×10^{-35}	-2.3×10^{-35}	-1.5×10^{-35}	-1.9×10^{-35}
$\sigma^{(1)}(\omega)$ (cm^2)	4.98×10^{-22}	6.97×10^{-22}	5.16×10^{-22}	6.84×10^{-22}
b (dB/cm)	1.86×10^{-3}	2.57×10^{-3}	1.93×10^{-3}	2.52×10^{-3}
Confocal parameter (cm)	7.0	6.0	6.9	6.0
α_S (s^2/cm)	-1.89×10^{-28}		-2.02×10^{-28}	
α_P (s^2/cm)	-3.28×10^{-28}		-3.19×10^{-28}	
$k_{IPM}(\omega_S)$ (esu)	4.0×10^{-7}		1.9×10^{-6}	
$k_{SPM}(\omega_P)$ (esu)	3.6×10^{-10}		2.9×10^{-10}	
$P(\lambda_P)$ (W)	1.4×10^5		1.3×10^5	
Compression ratio F	15		69	

bound transitions [18] are corrected for sign [19], [20]. The matrix elements for the bound-free transitions are from [21]. With the accuracy of the matrix elements used, the accuracy in the calculation of the nonlinear susceptibility is estimated to be 40%. In the vicinity of resonance we have taken into account the collision ($\sim 6.9 \cdot 10^9$ Hz) and Doppler ($\sim 2.3 \times 10^{10} - 4.3 \times 10^{10}$ Hz) broadening of the transitions. The upper limit for the signal power, according to assumption 1), is $P_S < ((\lambda_S A)/(\pi n_2^{IPM} L))$, where A is the core area. According to our estimation, the maximum pump power is restricted by multiphoton ionization and breakdown in the gas ($P_{P_{MAX}} = 1.7 \cdot 10^5$ W for $\lambda_P = 262.69$ nm—Table I). The maximum signal power $P_{S_{MAX}}$ is limited by the ionization ($\lambda_S = 157$, 193.4, and 248 nm) or by the approximation used ($\lambda_S = 193$, 206, and 210 nm).

The overall losses can be taken into account [21] by including a damping nonlinear coupling in the SE (4):

$$k(\omega_S) \rightarrow k(\omega_S, x) \equiv k(\omega_S) \exp[-2\gamma x] \tag{8}$$

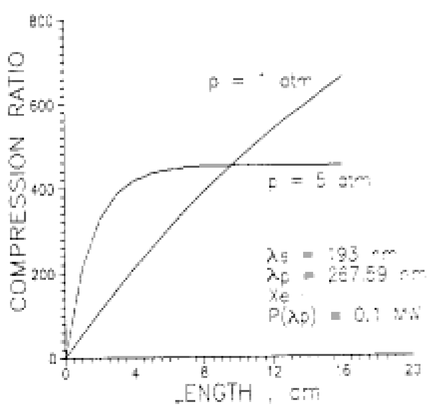

Fig. 3. Dependence of the compression factor on the length for different densities of the Xel-gas.

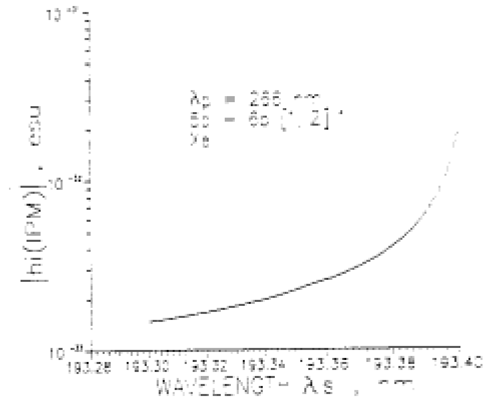

Fig. 4. Nonlinear susceptibility $\chi_{\rm IPM}^{(3)}$ for $\lambda_P = 266$ nm/fourth harmonic of an Nd:YAG laser/versus λ_S.

where γ is the decay constant. In this constant we have included the single-photon absorption in the medium $\chi^{(1)}(\omega)$, the multiphoton ionization and the waveguide losses of the beam b, taking $\nu = (\epsilon/\epsilon_0)^{1/2} < 2.02$ [13]. Fig. 3 illustrates the importance of choosing a proper medium density. Due to the stronger absorption at higher density, the bandwidth of the chirp is narrower, and the compression ratio is lower in a higher pressure gas waveguide.

In a possible experiment, the synchronization of pulses would be relatively simple in a double-channel excimer laser (e.g., EMG 150E). The pulses of the first one to be compressed λ_S(157, 193, and 248 nm) and the second one pumping a dye laser ($\lambda_{\rm DYE} = 2\lambda_P$). More difficult seems to be the synchronization of an ArF and YAG lasers. The jitter will negatively affect the compression of the probe signal [4], [6]. In the example considered (see Table I) λ_S is tuned near a two-photon resonance via $\omega_S + \omega_P$ within 1 Å. Fig. 4 shows the strong dependence of $\chi_{\rm IPM}^{(3)}$ (respectively, compression ratio $F = a_S\, \Delta\omega^{\rm IPM}/2$, where $\Delta\omega^{\rm IPM}$ is the chirp bandwidth) on the detuning from the resonance. As seen from Table I, the compression ratio F predicted is between 15 and 700. If a narrowband tunable ArF laser is used (e.g., EMG 150 MSC) with a 1 nm tuning range and a bandwidth of 0.005 nm FWHM, higher compression coefficients seem to be obtainable.

The calculation shows that the group-velocity mismatch between λ_S and λ_P is less than 20 ps/m at $p = 5$ atm, which is negligible, compared to the nanosecond duration and capillary length used (condition 4)).

An important condition for a successful pulse compression by a pair of gratings is the absence of a nonlinear frequency chirp. In all cases considered, the ratio $(1/2)n_2^{\rm IPM}|A_P|^2/((1/4)n_4^{\rm IPM}|A_P|^4)$ is higher than 10^4. In our considerations we have neglected the change in the pulses shapes, described by the term $(n^{\rm NL}/c)[\partial(|A_i|^2A_j)/\partial\tau]$. ($i$, $j = s, p$); however, the dependence of the group velocity on the intensity of the pulses may play an important role in the spectral broadening of the signal pulse [22]. Since $k_{\rm IPM} < 0$, the signal pulse compression should be obtainable with a grating compressor with positive group velocity dispersion [23].

Condition 4), i.e., low efficiency of the four-wave mixing and the four-photon parametric processes, is important in deriving (7a) and (7b). The dependence of the sum-frequency signal from the bandwidth, Doppler, and collision broadening and detuning from resonance of the pump laser has been investigated [24], however, for the case considered, a special study is needed.

It is interesting to evaluate the range of applicability of the IPM approach in the short wavelength region. The higher values of $\chi_{\rm IPM}^{(3)}$ are due to the vicinity of $\omega_S + \omega_P$ to a two-photon resonance. It seems, that the method would be applicable in inert gases for λ_S in the region between XUV and the soft X-ray. The inherent limitation of the resonance become significant by a detuning of the order of the chirp bandwidth. Therefore, the induced wavelength shift from the carrier frequency [14] of the signal pulse at different delays has to be considered carefully. Our estimation has shown that the method will be applicable to compressed-pulsewidths of the order of several picoseconds, taking into account the limitations imposed on the pump power, waveguide length ~ 10 cm, and $\chi_{\rm IPM}^{(3)} \sim 10^{-31}$ esu.

III. Conclusion

An approach is proposed for extension of the method for pulse compression in the UV, based on the induced-phase modulation. In the calculations we have used the variational method for solving the Schrödinger equation for propagation of two pulses in a waveguide filled with a low pressure inert gas as a nonlinear medium. High compression coefficients using grating pair seem to be attainable for the widely used nanosecond excimer laser pulses. The peak power limits ranging from $2.5 \cdot 10^2$ to $4 \cdot 10^4$ W are posed by multiphoton ionization and plasma formation. A study is under way to evaluate the influence of the induced phase modulation on the process of four-wave mixing.

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