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The induced phase modulation in the UV

S G Dinev and A A Dreischuh

Department of Physics, Sofia University, BG-1126 Sofia, Bulgaria

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Abstract. The method of induced phase modulation has been extended to the UV. Pump and signal waves are involved, the sum of their frequencies lying near a two-photon resonance in gas media. The variational method for solving the Schrödinger equation for two waves has been applied.

The techniques for pulse compression have been extensively developed in recent years. The introduction of self-phase modulation (SPM) in single-mode fibres (Fork *et al* 1987) and the following compression have allowed the generation of pulses, the shortest being 6 fs in the visible. Pulses of duration 43 fs at 310 nm are obtained by intracavity frequency doubling in a colliding pulse-mode-locked laser (Edelstein *et al* 1988). Because of the relatively low peak powers of the existing coherent sources in the VUV and XUV regions, the SPM does not offer the possibility of obtaining the desired dispersion for pulse compression. Time compression of x-ray pulses from electron-storage rings using equitemporal optics and x-ray pulse lengths down to 100 fs also seem feasible (Gsonka 1988). The method of the induced phase modulation (IPM) in optical fibres has been studied both theoretically and experimentally by Manassah (1987, 1988), Yamashita *et al* (1988), Alfano *et al* (1989) and Agrawal *et al* (1989a, b). In this work we report a new approach to the IPM method in a gas media near two-photon resonant conditions. The phase modulation is induced by a pump pulse on a relatively weak signal pulse. The sum of the two frequencies coincides with a two-photon transition, while both waves are far from single-photon resonance. Changing the pump power and other system parameters, a controllable compression could be achieved in the UV region. The variational method for solving the Schrödinger equation in the case of two waves was applied.

Let us consider the case of two pulses, signal I_s and pump I_p, propagating in the waveguide with cubic non-linearity. We assume that:

(i) the self-phase modulation is negligible;

(ii) $I_s \ll I_p$, i.e. the phase modulation induced by the signal I_s on the pump pulse can be neglected;

(iii) the pulses are of different, but nearly equal durations and the delay is approximately zero;

(iv) the group-velocity mismatch between λ_s and λ_p is negligible and a single laboratory coordinate system can be used;

(v) the efficiency of the four-wave frequency mixing and parametric processes is low, i.e. the energy of the pulses does not change, and

(vi) the non-linear response time is negligible compared with the pulse durations.

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The characteristic equation determining the evolution of the slowly varying signal pulse is the Schrödinger equation (SE):

$$\mathrm{i}\,\frac{\partial\psi_s}{\partial x}=\alpha\,\frac{\partial^2\psi_s}{\partial\tau^2}+k(\omega_s)|\psi_p|^2\psi_s. \tag{1}$$

We assume that the pulses have a Gaussian form:

$$\psi_s(x,\tau)=A_s(x)\exp\{[-\tau^2/2a_s^2(x)]+\mathrm{i}b_s(x)\tau^2\} \tag{2a}$$

$$\psi_p(x,\tau)=A_p(x)\exp[-\tau^2/2a_p^2(x)] \tag{2b}$$

where $A_s(x)$ and $A_p(x)$ are the complex amplitudes, $a_s(x)$ and $a_p(x)$ are the half pulse widths on 1/e level and $b_s(x)$ is the frequency-chirp parameter for the induced phase modulation. In (1) $\alpha=(1/2)[\lambda_s^3/(2\pi c^2)](\partial^2 n/\partial\lambda^2)_{\lambda=\lambda_s}$ is a coefficient determined by the dispersion and $k(\omega_s)=-3\pi^2 N\chi^{(3)}_{\mathrm{IPM}}(\omega_s)/[\lambda_s n_0^2(\omega_s)]$ is the non-linear coefficient for the IPM $(\chi^{(3)}_{\mathrm{IPM}}(\omega_s)=\chi^{(3)}(-\omega_s,\omega_s,\omega_p,-\omega_p))$.

Our analysis is based on the variational functional corresponding to the SE (Anderson 1983, Anderson *et al* 1988). It can be shown that (1) can be restated as a variational problem corresponding to the Lagrangian L given by

$$L=(\mathrm{i}/2)\left(\psi_s\frac{\partial\psi_s^*}{\partial x}-\psi_s^*\frac{\partial\psi_s}{\partial x}\right)-\alpha\left|\frac{\partial\psi_s}{\partial\tau}\right|^2+k(\omega_s)|\psi_s|^2\cdot|\psi_p|^2. \tag{3}$$

The variational problem can be reduced to the form (Anderson *et al* 1988)

$$\delta\int_{-\alpha}^{+\alpha}\langle L\rangle\,\mathrm{d}x=0$$

where

$$\langle L\rangle=\int L_G\,\mathrm{d}\tau$$

and L_G denotes the result of inserting equations (2*a*–*b*) into the Lagrangian (3). In the case considered

$$\begin{aligned}\langle L\rangle=(\mathrm{i}/2)&\left(A_s(x)\frac{\mathrm{d}A_s^*}{\mathrm{d}x}-A_s^*(x)\frac{\mathrm{d}A_s}{\mathrm{d}x}\right)a_s(x)\pi^{1/2}+|A_s(x)|^2\frac{\mathrm{d}b_s}{\mathrm{d}x}\frac{a_s^3(x)\pi^{1/2}}{2}\\&-\left(\frac{\alpha|A_s(x)|^2}{a_s^4(x)}+4\alpha|A_s(x)|^2b_s^2(x)\right)\tfrac{1}{2}a_s^3(x)\pi^{1/2}\\&+k|A_s(x)|^2\times|A_p(x)|^2\frac{a_s(x)a_p(x)\pi^{1/2}}{[a_s^2(x)+a_p^2(x)]^{1/2}}.\end{aligned} \tag{4}$$

In this way we obtain a set of two coupled ordinary differential equations for $a_s(x)$ and $b_s(x)$:

$$a_s\frac{\mathrm{d}b_s}{\mathrm{d}x}=4\alpha a_s b_s^2-\frac{\alpha}{a_s^3}+2k(\omega_s)|A_p|^2\frac{a_p/a_s}{(a_s^2+a_p^2)^{1/2}}\left(1-\frac{a_p^2}{a_s^2+a_p^2}\right) \tag{5}$$

$$\frac{\mathrm{d}a_s}{\mathrm{d}x}=-4\alpha a_s b_s. \tag{6}$$

In order to describe correctly the evolution of the pulsewidth of the pump pulse $a_p(x)$ and to prove that the SPM of this pulse is negligible, we have included the variational-approach results (Anderson 1983, Anderson *et al* 1988), concerning single-pulse propagation in a waveguide.

As an illustration of the induced phase modulation in the UV we have chosen $\lambda_s = 193$ nm (ArF excimer laser) and $\lambda_p = 262.69$ nm (second harmonic of a dye laser). In a possible experiment, the synchronization of pulses would be relatively simple in a double-channel excimer laser (e.g. EMG150E). The pulses of the first one are to be compressed and the second one to pump a dye laser ($\lambda_{DYE} = 2\lambda_p$). In Xe I, $(\omega_s + \omega_p)$ is near the two-photon resonance $5p^6[^1S_0]$-$6p'[1/2]_0$ and, at the same time, ω_s and ω_p are far from the single-photon resonances (see figure 1). The waveguide considered is a hollow-core capillary of length $l = 16$ cm and inner diameter 150 μm containing Xe I under a pressure of 1 atm. The pump and the signal power are limited by the two- and three-photon ionization up to $P_{s,max} = 1.2 \times 10^4$ W and $P_{p,max} = 1.2 \times 10^5$ W. The non-linear susceptibilities for phase modulation, calculated according to the single-sided Feynman diagrams (Prior 1984) and the corresponding matrix elements (Radzig and Smirnov 1986), are found to be

$$\chi^{(3)}_{SPM}(\lambda_s = 193\text{ nm}) = 2.4 \times 10^{-35}\text{ esu}$$

$$\chi^{(3)}_{SPM}(\lambda_p = 262.69\text{ nm}) = 4.4 \times 10^{-35}\text{ esu.}$$

The non-linear susceptibility for induced phase modulation is calculated to be

$$\chi^{(3)}_{IPM}(-\omega_s, \omega_s, \omega_p, -\omega_p) = -1.7 \times 10^{-32}\text{ esu.}$$

Therefore, the self-phase modulation will be negligible. According to the accuracy of the matrix elements, the accuracy of the $\chi^{(3)}$ coefficients is within 40%. Assuming initial pulse durations of $a_s(0) = 11$ ns (FWHM) and $a_p(0) = 14$ ns (FWHM), the maximum achievable compression factor will be $F = 46$. The group-velocity mismatch is calculated to be less than 20 ps m^{-1} at $p = 1$ atm and less than 80 ps m^{-1} at $p = 5$ atm, which is negligible compared with the nanosecond duration and capillary length used (condition (iv)). Therefore, if we neglect the pump absorption, the increase of medium pressure would enhance the chirp-bandwidth, i.e. the compression factor. The influence of the two pulse durations towards IPM is discussed by Manassah (1988). The results show that at $a_p \gg a_s$ the chirp is nearly linear at the expense of a reduced chirp-bandwidth at the same pump power. Time shortening of the signal pulses might be done using a grating compressor with positive group-velocity dispersion (Martinez 1987). The possible initial delay τ_d between the nanosecond signal and pump pulse will influence

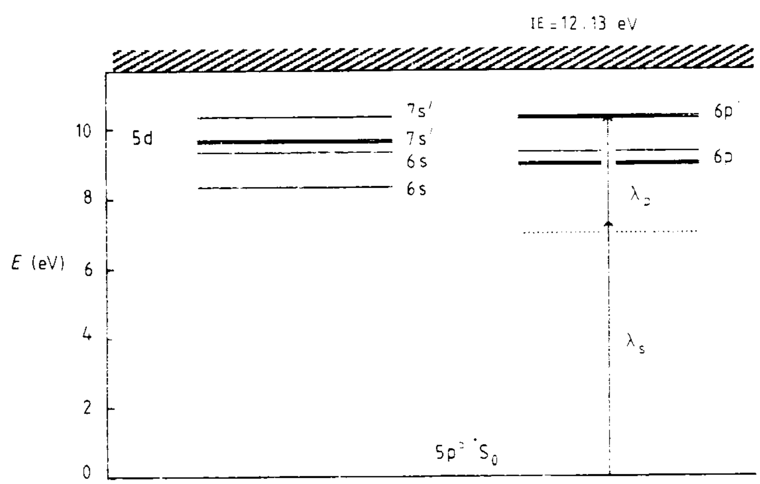

Figure 1. Simplified energy level diagram for Xe I.

negatively the compression factor. This situation can be included in our model by a coordinate system, connected with the signal pulse. In this case the pump pulse will have the form

$$\psi_p(x, \tau) = A_p(x) \exp(-(\tau - \tau_d)^2 / 2a_p^2(x)) \tag{7}$$

and the chirp-rate can be determined by

$$a_s \frac{db_s}{dx} = 4\alpha a_s b_s^2 - \frac{\alpha}{a_s^3} + \frac{2k(\omega_s)|A_p|^2 a_s a_p}{(a_s^2 + a_p^2)^{3/2}} \exp\left(-\frac{\tau_d^2}{a_s^2 + a_p^2}\right)\left(1 - \frac{2\tau_d^2}{a_s^2 + a_p^2}\right) \tag{8}$$

and (6). Figure 2 (curve 1) shows the strong dependence of the compression ratio $F = a_s \Delta\omega^{IPM}/2$ on the initial delay τ_d. Using picosecond pulses in a region of normal dispersion, the delay of the pump pulse in respect to the signal could be compensated by the group-velocity mismatch. In the nanosecond range considered, however, the effect is negligible. As seen from figure 2 (curve (2)), within the range considered, the compression ratio decreases monotonically with the duration of the transform-limited pulse.

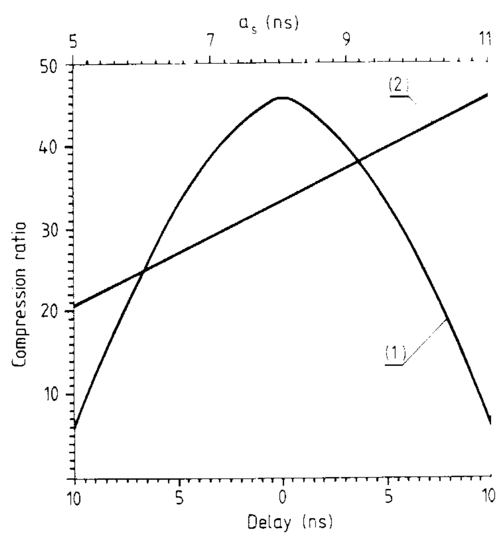

Figure 2. Compression ratio against (1) initial delay between the signal and pump pulses; (2) duration of the input transform-limited signal pulse.

The inherent limitation of the resonance becomes significant by a two-photon detuning of the order of the chirp-bandwidth and the induced wavelength shift from the carrier frequency has to be considered carefully (Alfano *et al* 1989) in this case. The spectral broadening $\Delta\lambda$ of a transform-limited pulse of fixed duration decreases with increasing of the carrier frequency. Therefore, the IPM seems to be an attractive alternative to the SPM in the VUV and the XUV regions.

In conclusion the induced phase modulation in a gas-filled hollow-core waveguide is proposed to be a possible approach for pulse compression in the UV. Further calculations are under way to optimize the system parameters as well as to obtain the limitation of this approach in the VUV and XUV.

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