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Sum-frequency generation in the XUV

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The influence of the non-linear part of the wave-vector mismatch $\Delta\mathbf{k}^{NL}$ on the sum frequency generation in a hollow core waveguide is analysed. According to the value and sign of the respective non-linear susceptibilities, considerable deviation in the signal P_s on the pump powers P_1 and P_2 can be induced, compared to the case when the linear part $\Delta\mathbf{k}^L$ is considered only. Non-linear schemes are proposed for X-ray ultra violet XUV-generation near 60 nm.

1. Introduction

Non-linear mixing of frequencies in hollow waveguides has been discussed theoretically [1–3] and experimentally to increase the efficiency of a number of non-linear processes, for example SRS, CARS and four-wave mixing [4, 5]. The input intensity in these metal or dielectric waveguides can be increased to values comparable with a tightly focused beam, while the interaction length L is much higher than the confocal parameter b in the case of tight focusing.

The non-linear optical mixing in inert gases is an efficient method for generation of XUV coherent radiation. In a recent experiment of 5th-harmonic generation of 248 nm in Ne I and Ar I jet, Rosman *et al.* [6] measured a non-linear component of the wave-vector mismatch $\Delta\mathbf{k}^{NL}$ higher than the linear component $\Delta\mathbf{k}^L$. This has lead to a considerable change in the predicted signal on pump dependence, when only $\Delta\mathbf{k}^L$ was taken into account.

In this paper we theoretically analyse the resonant non-linear mixing of the type $\omega_s = 2\omega_1 + \omega_2$ in a hollow waveguide. The influence of the self-modulation of the pump, and modulation induced by the pump beams on the phase-matching conditions, are studied. Schemes for efficient frequency conversion in the XUV are discussed.

2. Theoretical model

In the non-linear optical frequency mixing process the radiation in the hollow waveguide can be represented by a dumped quasi plane wave with a certain amplitude distribution over the waveguide cross-section. The power P_s generated in this process, assuming a small absorption in the non-linear media and a single mode for each of the interacting waves, is given by [2]

$$P_s = |4\pi^3(\chi^{(3)}(-\omega_s;\omega_1,\omega_1,\omega_2)NL/c|^2 \nu_s^2\langle 112s\rangle^2(P_1^2P_2/(n_1^2n_2n_s)) \times \{\sin(\Delta\mathbf{k}L/2)/(\Delta\mathbf{k}L/2)\}^2 \quad (1)$$

where N is the particle density, c is the velocity of the light, L is the length of the waveguide, $\nu_s = \omega_s/2\pi c$ is the wave-number, $\Delta\mathbf{k} = k_s - (2k_1 + k_2)$ is the wave-vector mismatch, n_i

is the ith refractive index and $\langle 112s\rangle$ is the overlap integral ($\langle 112s\rangle = 1/(\pi a^2)$ for Gaussian modes, where a is the inner radius of the waveguide).

The maximum output power at the sum-frequency can be expected for $\Delta\mathbf{k} = 0$ and in this case P_s is proportional to $(NL)^2$. Let us denote with α_i the normalized deviation of the refractive index from unity for the ith wave

$$\alpha_i = (n_i - 1)/N \tag{2}$$

It follows from Equation 1 that

$$\Delta\mathbf{k} = 2\pi N\beta \tag{3a}$$

$$P_s = |4\pi^3\chi^{(3)}(-\omega_s;\omega_1,\omega_1,\omega_2)/c|^2\{v_s^2\langle 112s\rangle^2(P_1^2P_2/(n_1^2n_2n_s))\}\{\sin(\pi\beta Nl)/(\pi\beta)\}^2, \tag{3b}$$

where

$$\beta = \alpha_s/\lambda_s - 2\alpha_1/\lambda_1 - \alpha_2/\lambda_2 \tag{3c}$$

We can see from (3b) that, in the case of non-zero phase mismatch, P_s depends on (NL) mostly through the term $\{\sin(\Delta\mathbf{k}L/2\}^2$, which is a more rapidly varying function of (NL) than the refractive indices n_1, n_2 and n_s. The optimum non-zero value of $\Delta\mathbf{k}$ can be determined by the requirement $\Delta\mathbf{k}L = \pm\pi$.

Let us consider the non-linear component of the wave-vector mismatch $\Delta\mathbf{k}^{NL}$. The complex amplitude of the field and the induced polarization can be considered to be of the form

$$E_j(r,z,t) = (1/2)\{E_j(r,z)\exp[-i(\omega_j t - k_j z)] + c.c.\},\quad j = 1, 2, s \tag{4a}$$

$$P_j(r,z) = N\{\chi^{(1)}E_j(r,z) + (1/4)\chi^{(3)}E_j^3(r,z) + \ldots\} \tag{4b}$$

Included in our model are the phase self-modulation of the pump beams and the phase modulation induced by the pump both on signal and pump waves [15], that is,

$$P^{NL}(\omega_1) = (3/4)N\{\chi^{(3)}(-\omega_1;\omega_1,-\omega_1,\omega_1)E_1|E_1|^2 + 2\chi^{(3)}(-\omega_1;\omega_1,\omega_2,-\omega_2)E_1|E_2|^2\} \tag{5a}$$

$$P^{NL}(\omega_2) = (3/4)N\{\chi^{(3)}(-\omega_2;\omega_2,-\omega_2,\omega_2)E_2|E_2|^2 + 2\chi^{(3)}(-\omega_2;\omega_2,\omega_1,-\omega_1)E_2|E_1|^2\} \tag{5b}$$

$$P^{NL}(\omega_s) = (3/2)N\{\chi^{(3)}(-\omega_s;\omega_s,-\omega_1,\omega_1)E_s|E_1|^2 + \chi^{(3)}(-\omega_s;\omega_s,\omega_2,-\omega_2)E_s|E_2|^2\} \tag{5c}$$

Taking into account the relations [15]

$$\varepsilon_{tot}(\omega_i) = 1 + 4\pi[P^L(\omega_i)/E_i] + 4\pi[P^{NL}(\omega_i)/E_i] \tag{6a}$$

$$n_{tot}^2(\omega_i) = \varepsilon_{tot}(\omega_i),\quad i = 1, 2, s \tag{6b}$$

$$n_{tot}^2(\omega_1) = [n^L(\omega_1) + (1/2)n_0^{NL}(\omega_1)E_1^2 + (1/2)n_{i2}^{NL}(\omega_1)E_2^2 + \ldots]^2 \tag{7a}$$

$$n_{tot}^2(\omega_2) = [n^L(\omega_2) + (1/2)n_0^{NL}(\omega_2)E_2^2 + (1/2)n_{i1}^{NL}(\omega_2)E_1^2 + \ldots]^2 \tag{7b}$$

$$n_{tot}^2(\omega_s) = [n^L(\omega_s) + (1/2)n_{i1}^{NL}(\omega_s)E_1^2 + (1/2)n_{i2}^{NL}(\omega_s)E_2^2 + \ldots]^2 \tag{7c}$$

we get expressions for the respective non-linear refractive indexes. In this way, the non-linear part of the wave-vector mismatch is

$$\begin{aligned}\Delta\mathbf{k}^{NL} = {} & [6\pi^2 N]\{[\chi^{(3)}(-\omega_s;\omega_s,\omega_1,-\omega_1)E_1^2 + \chi^{(3)}(-\omega_s;\omega_s,\omega_2,-\omega_2)E_2^2] \\ & \times [n^L(\omega_s)\lambda_s]^{-1} - [\chi^{(3)}(-\omega_1;\omega_1,-\omega_1,\omega_1)E_1^2 + 2\chi^{(3)}(-\omega_1;\omega_1,\omega_2,-\omega_2)E_2^2] \\ & \times [n^L(\omega_1)\lambda_1]^{-1} - [(1/2)\chi^{(3)}(-\omega_2;\omega_2,-\omega_2,\omega_2)E_2^2 + \chi^{(3)}(-\omega_2;\omega_2,\omega_1,-\omega_1)E_1^2] \\ & \times [n^L(\omega_2)\lambda_2]^{-1}\} \qquad (8)\end{aligned}$$

where the microscopic non-linear susceptibilities are in $m^5\ V^{-2}$, N is in m^{-3}, the amplitudes of the fields are in Vm^{-1}.

It should be noted that, by opposite signs of $\Delta\mathbf{k}^L$ and $\Delta\mathbf{k}^{NL}$, the sum-frequency signal power can be increased by an appropriate choice of both NL and the pump powers.

3. Discussion

Let us consider a sum-frequency mixing in the particular case of $\lambda_1 = 157$ nm (F_2 laser) and $\lambda_2 = 318$ nm or 310.7 nm (second harmonic of a dye laser). The sum-frequency ω_s would be in a three-photon resonance with the $2p^6 - 4s[3/2]_1$ ($\delta\lambda_s = 0.013$ nm) or $2p^6 - 4s'[1/2]_1$ ($\delta\lambda_s = 0.011$ nm) in Ne I, respectively. The two-photon detuning for λ_1 from the $3p[1/2]_1$ level is $\delta\lambda_1 = 12.86$ nm. Shown in Table I are some of the input and output parameters and the respective non-linear susceptibilities. The non-linear coefficients are calculated according to the single-sided diagrams, extensively discussed in references [7, 8]. In the vicinity of resonance the homogeneous linewidths of the transitions are taken into account. The matrix elements are derived by the respective transition probabilities [9, 10], while their signs are obtained by the Bates and Damgaard approach [11], together with the correction of Bebb [12]. The energy levels are from Bashkin and Stoner [13]. The matrix elements for the bound-free transitions are calculated by Peach [14]. The overall accuracy in the calculation of the non-linear susceptibilities is within 50%.

TABLE I Parameters for the sum-frequency model

Parameter	Unit	Value	
		$\lambda_s = 62.96$ nm	$\lambda_s = 62.67$ nm
λ_1	nm	157	157
λ_2	nm	318	310.7
P_1	MW	5	5
P_2	MW	0.1 to 1	0.1 to 1
$\Delta\mathbf{k}^L/N$	cm^2	1.61×10^{-16}	1.27×10^{-16}
$\chi^{(3)}(\omega_s;\omega_1,\omega_1,\omega_2)$	$m^5\ V^{-2}$	-2.8×10^{-47}	-5.6×10^{-47}
$\chi^{(3)}(\omega_1;\omega_1,-\omega_1,\omega_1)$	$m^5\ V^{-2}$	4.2×10^{-51}	4.2×10^{-51}
$\chi^{(3)}(\omega_2;\omega_2,-\omega_2,\omega_2)$	$m^5\ V^{-2}$	1.3×10^{-51}	1.3×10^{-51}
$\chi^{(3)}(\omega_s;\omega_s,\omega_1,-\omega_1)$	$m^5\ V^{-2}$	-1.4×10^{-44}	-2.8×10^{-45}
$\chi^{(3)}(\omega_s;\omega_s,\omega_2,-\omega_2)$	$m^5\ V^{-2}$	-7.0×10^{-44}	-1.3×10^{-44}
$\chi^{(3)}(\omega_1;\omega_1,\omega_2,-\omega_2)$	$m^5\ V^{-2}$	8.4×10^{-51}	8.4×10^{-51}
$\chi^{(3)}(\omega_2;\omega_2,\omega_1,-\omega_1)$	$m^5\ V^{-2}$	8.4×10^{-51}	8.4×10^{-51}
$\sigma^{(1)}(\omega_s)$	cm^2	5.3×10^{-21}	1.6×10^{-20}
NL	cm^{-2}	1.95×10^{15}	1.95×10^{17}
a	μm	100	100
P_s	W	~70	~70
$\eta = P_s/P_1$	%	1.4×10^{-3}	1.4×10^{-3}

The maximum value of NL is determined from the calculated single-photon absorption cross-section $\sigma^{(1)}(\omega_s)$ of the signal, according to the relation

$$(I/I_0)_{min} = \exp[-\sigma^{(1)}(\omega_s)(NL)_{max}] \geqslant 0.95$$

The NL values discussed satisfy this relation. Generally, one can use a two-photon resonance, thereby maintaining large values of $\chi^{(3)}(\omega_s; \omega_1, \omega_1, \omega_2)$ and $\chi^{(3)}(\omega_s; \omega_s, \omega_2, -\omega_2)$ but allowing increased NL.

As seen from Table I, the main contribution to $\Delta\mathbf{k}^{NL}$ is from the signal phase modulation, induced by the pump, $\chi^{(3)}(\omega_s; \omega_s, \omega_p, -\omega_p) \approx (-10^{-44}$ to $-10^{-45})\text{m}^5\,\text{V}^{-2}$. The negative sign of $\Delta\mathbf{k}^{NL}$ allows, in principle, by appropriate choice of P_1, P_2 and NL, to compensate for the wave-vector mismatch and to satisfy the condition $\Delta\mathbf{k}L = 0, \pm\pi$. The absence of signal focusing, induced by the pump, is an additional appropriate condition.

The dependence of signal power P_s on pump power P_2 and NL is plotted in Fig. 1 for the first case ($\lambda_s = 62.96\,\text{nm}$). The left maximum of the signal corresponds to $NL = 6.0 \times 10^{16}\,\text{cm}^{-2}$ and the right to $NL = 1.8 \times 10^{17}\,\text{cm}^{-2}$. At fixed pump power the oscillating dependence of P_s on NL is evident (see Equation 3). For an optimum NL value the dependence of P_s on P_2 is non-linear. For the maximum $NL = 6.0 \times 10^{16}\,\text{cm}^{-2}$, $P_s \approx P_2^3$ instead of $P_s \approx P_2$, as would be expected if $\Delta\mathbf{k}^L$ was considered only (see Fig. 2). Fixing $P_2 = 10^6\,\text{W}$ and changing P_1 from $10^6\,\text{W}$ to $5 \times 10^6\,\text{W}$ for the same values of NL, the dependence of P_s on P_1 is much faster compared to the quadratic dependence expected (see Equation 1).

The waveguide intensities of the beams are at least an order of magnitude lower than the saturation intensities. The level splitting due to the optical Stark effect [15] is negligible compared to the level linewidths, hence its negative contribution to the resonant non-linear

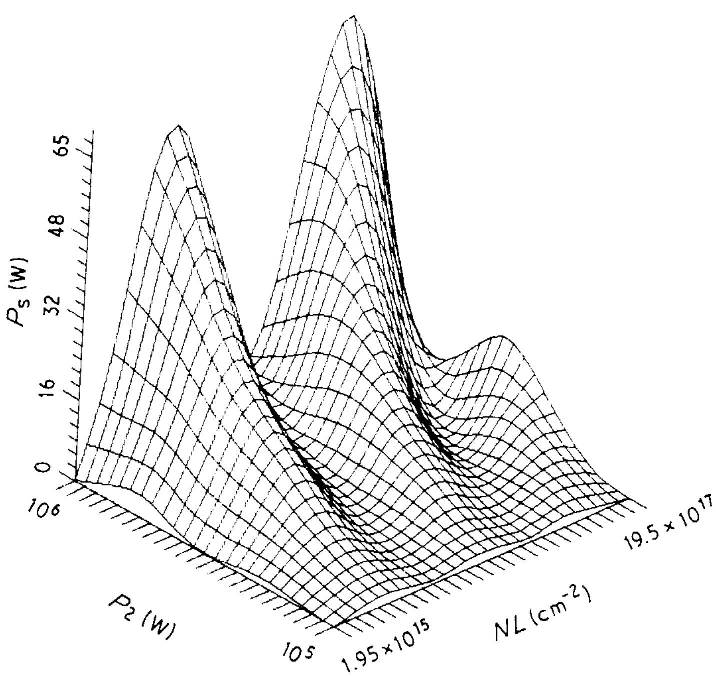

Figure 1 Signal power P_s dependence on pump power P_2 and density-length product NL for $\lambda_s = 62.96\,\text{nm}$.

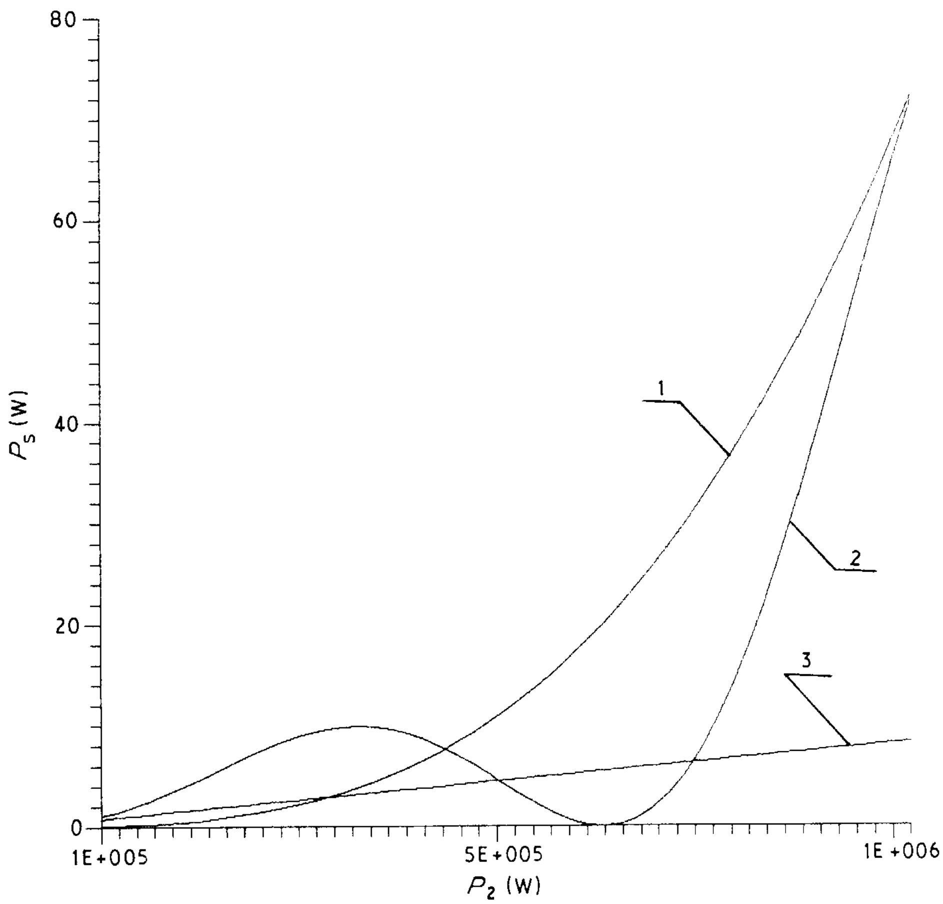

Figure 2 Signal power P_s versus pump power P_2 in the case $\Delta \mathbf{k} = \Delta \mathbf{k}^L + \Delta \mathbf{k}^{NL}$, (1) $NL = 6.0 \times 10^{16}$ cm^{-2}, (2) $NL = 1.8 \times 10^{17}$ cm^{-2} and (3) $\Delta \mathbf{k} = \Delta \mathbf{k}^L$.

susceptibilities can be neglected. From the energy level diagram of Ne I, a three-photon ionization from the F_2 laser, as well as a two-photon ionization from the signal, enhanced by an intermediate resonance, are to be expected. The free electrons obtained can contribute to the wave-vector mismatch [15]. The ionization would negatively influence the efficiency of the process, depleting the ground state atoms. The plasma generated would lead to line broadening, given by the expression [16]

$$\delta\nu \approx 1.4(n_u^2 - n_l^2)Z_i N_i^{2/3}/S_e, \qquad (9)$$

for transitions originating from a shell of principal quantum number n_u, at which electrons experience an effective nuclear charge S_e. In Equation 9 n_l is the principal quantum number of the lower shell and Z_i is the ion charge. The plasma density N_i can be calculated from the photo-ionization rate $W_{kg}^{(n)}$ [12, 17]

$$N_i = W_{kg}^{(n)} \tau_{imp} N, \qquad (10)$$

where τ_{imp} is the laser pulse duration. Our estimate for the Ne I gives $W_{kg}^{(2)}(\lambda = 62.96\,\text{nm}) = 10^{-44} F^2$, where $F = P_s/(\pi a^2 h\nu_s)$ is the photon flux. When the line broadening in the plasma is comparable with the detuning from the resonance, a decrease in the non-linear coefficients, respectively process efficiency, is to be expected. The limitation obtained from Equation 9 and Equation 10 at $\tau_{imp} \approx 10$ ns and $N_{NeI} \approx 10^{17}$ cm^{-3} is signal power P_s of the order of tens of Watts. Possibly, the use of picosecond pulses and the increase of P_2 up to 2.1 MW would allow an exact phase matching and a signal power of the order of several kW to be achieved. The group velocity mismatch of the pulses for a waveguide length $L = 8$ cm causes a shift of about 8 fs, which is negligible, compared to the pulse duration considered.

The second case (see Table I) is qualitatively similar. Due to the smaller $\Delta\mathbf{k}^L$, within the range of NL discussed, the period of oscillation is smaller. From the other side, the non-linear coefficients $\chi^{(3)}(-\omega_s; \omega_s, \omega_j, -\omega_j)$ $(j = 1, 2)$ are smaller, leading to weaker deviation of P_s from the linear, respectively quadratic, dependence on P_1 and P_2.

The case of frequency mixing $\omega_s = 2\omega_1 + \omega_2$ with $\lambda_1 = 120.3$ nm (non-linear frequency conversion source with $P_1 = 300$ W [18]), $\lambda_2 = 497.8$ nm (dye laser) and $\lambda_s = 53.67$ nm in He I was also considered. The high non-linear coefficient $\chi^{(3)}(-\omega_s; \omega_1, \omega_1, \omega_2) = 5.6 \times 10^{-46}$ m^5 V^{-2} is due to the two-photon resonance He(1s – 2s) ($\delta\lambda_1 = 0.007$ nm) and the three-photon resonance He(1s – 3p) ($\delta\lambda_s = 0.035$ nm). Since $\chi^{(3)}(\omega_s; \omega_s, \omega_1, -\omega_1) = -7 \times 10^{-45}$ m^5 V^{-2} and $\chi^{(3)}(\omega_s; \omega_s, \omega_2, -\omega_2) = 4.2 \times 10^{-43}$ m^5 V^{-2}, an uncontrolled self-focusing is to be expected still above $P_{2cr} \approx 20$ W, which strongly limits the interaction length. From the other side, an appropriate choice of P_1, P_2 and NL can optimize the wave-vector mismatch in a small range. All this would limit the signal power to $< 1\,\mu$W.

4. Conclusions

We have analysed the influence of the non-linear part of the wave-vector mismatch $\Delta\mathbf{k}^{NL}$ on the sum-frequency generation in hollow waveguides. According to the value and the sign of the respective non-linear coefficient, considerable deviations in the sum-frequency power P_s dependence on pump powers P_1 and P_2 can be induced, compared to the case, when the linear part $\Delta\mathbf{k}^L$ only is accounted for. Two cases are considered for XUV frequency generation with peak power of several tens of Watts.

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