Submillimeter Wave Generation by Nonstationary Mixing

V. V. Apollonov and Yu. A. Shakir

General Physics Institute, Russian Academy Sciences, ul. Vavilova 38, Moscow, 117492 Russia
e-mail: shakir@kapella.gpi.ru

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Abstract—Three-wave interaction of picosecond pulses was investigated for difference frequency generation with AgGaSe\(_2\) and AgGa\(_{1-x}\)In\(_x\)Se\(_2\) crystals. Group velocities and radiation absorption were taken into account during spectral analysis realization. Calculations were executed for CO\(_2\) laser parameters: 100–5 ps, 15–150 GW/cm\(^2\). Spectral characteristics and pulse power versus crystal length are represented for difference wavelength 800.5 \(\mu\)m.

INTRODUCTION

We proposed to generate powerful half-cycle pulse by difference frequency generation (DFG) using CO\(_2\) laser pulses with duration \(\geq\)200 ps, nonlinear crystal ZnGeP\(_2\) and both laser semiconductor reflection and absorption switching to shorten produced submillimeter pulse down to half cycle [1]. Later it was found that both AgGaSe\(_2\) and AgGa\(_{1-x}\)In\(_x\)Se\(_2\) crystal properties better correspond to this problem solving [2]. Calculation results promised to obtain the submillimeter pulse with more than 10 MW power at duration \(\sim\)200 ps. Refraction data of these crystals [2,3] allow to present phase-matching characteristics for DFG with AgGa\(_{1-x}\)In\(_x\)Se\(_2\) (for \(x = 0.4\)) and AgGaSe\(_2\) crystals when various combinations of CO\(_2\) lines from both 9-\(\mu\)m and 10-\(\mu\)m branches interact by type ‘oe --- e’ (Fig. 1).

Also we represent investigation results on three-wave interaction in both crystals when laser pulse duration \(\leq\)100 ps. DFG spectral analysis was executed with due account of group velocities and wave absorption. Formulas, figures and tables present the calculation results.

NONSTATIONARY OPTICAL MIXING

We considered plane-wave mixing in approximation of laser fields fixed with frequencies \(\omega_2, \omega_1\). It is necessary to notice that CO\(_2\) laser frequency absorption is much less than one of difference frequency at submillimeter region [2]. We suppose that phase-matching conditions were fulfilled: \(k_1 = k_3 - k_2\). For ultrashort laser pulses process of DFG is described by equation:

\[
\begin{align*}
\partial A_i/\partial z + 1/u_i \partial A_i/\partial t + \delta_i A_i & = -i\gamma_i A_j(t-z/u_3)A_j^\star(t-z/u_2), \\
\end{align*}
\]

where \(\omega_i = \omega_3 - \omega_2, A_j(t, z)\) is complex field amplitude, \(u_i\) is the group velocity, absorption coefficient \(\delta_i\) and refractive index \(n_1\) of crystal at frequency \(\omega_i\), nonlinear coupling coefficient \(\gamma_i = 4\pi\omega_i d_{eff}/(en_1)\), \(c\) is the light velocity, \(d_{eff}\) is the effective nonlinearity.

For spectral analysis solution to Eq. (1) was represented by Fourier spectrum of amplitudes \(A_j(t, z)\). In result

\[
\begin{align*}
(\partial A_1(\Omega_1, z)/\partial z + \delta_1 A_1(\Omega_1, z))\exp(i\Omega_1 z/u_1) & = -i\gamma_1/2\pi \int A_3(\Omega_3, z)A_3^\star(\Omega_3 - \Omega_1, z) \\
& \times \exp(i z(\Omega_2/u_2 - \Omega_3/u_3))d\Omega_3,
\end{align*}
\]

there \(A_j(t, z) = 1/2\pi \int_{-\infty}^{\infty} A_j(\Omega, z)\exp(i\Omega(t-z/u))d\Omega\).

If to consider laser Fourier spectrum with Lorentz form, i.e.,

\[
A_{2,3}(\Omega) = \sqrt{S_{2,3}(0)\Delta\omega_{2,3}/(\Delta\omega_{2,3} + i\Omega)},
\]

where \(\Delta\omega_2\) is halfwidth of spectrum then spectral density of difference frequency \(S_1(\Omega, z) = |A_1(\Omega, z)|^2\) would be presented by

\[
S_1(\Omega, z) = (2\gamma_1/\nu_{13})^2 \times \exp(-\delta_1 z - z/L_{32}) F(\Omega, z) S_2(0) S_3(0) S_1^\star(\Omega),
\]

there length of pulses mismatch is \(L_{jk} = 1/(v_j\Delta\omega_k)\), inverse group-velocity mismatch is \(\nu_{jk} = 1/u_j - 1/u_k\), \(\mu = \nu_{13}/\nu_{32}\),

\[
F(\Omega, z) = (\Delta\omega_2\Delta\omega_3/\Delta\omega)^2 \\
\times [\sin^2(\Omega \nu_{13} z/2) + \sin^2(K z/2)]/(M\Delta\omega_2^2 + \Omega^2),
\]
Standardized spectral density of difference frequency is defined from (2) by expression

\[ S_1^q(\Omega) = \frac{\Delta \Omega^2}{(\Delta \Omega^2 + \Omega^2)^2} \]

Group-velocity values were calculated using crystal refractive index approximation in both submillimeter [2] and infrared [3] ranges. According to these calculations and experimental data [2] we present both used radiation parameters and the crystal characteristics:

- \( l_1 = 800.5 \) mm,
- \( l_2 = 9.24 \) mm,
- \( l_3 = 9.13 \) mm,
- laser beam square 1 cm\(^2\),
- crystal length \( z \geq 1.0 \) cm,
- nonlinear coefficient \( d_{36} = 33 \) pm/V [4].

The wavelength \( \lambda_1 \) was selected because of convenient cycle value (~2.6 ps). Other crystal characteristics are presented in Table 1.

When \( z \ll L_{32}/(\delta_1 L_{32} - 1) \) and \( z \ll L_{13} \) conditions simultaneously are fulfilled we have quasi-static regime of frequencies mixing and obtain from (3) that

\[ S_{1N}(\Omega, z) = S_1^q(\Omega) M \Delta \Omega^2 2/M (\Delta \Omega^2 + \Omega^2)^2 \times \left[ \sin^2(\Omega \nu_{13} z/2) + \sinh^2(K z/2) / \sinh(K z/2)^2 \right] \]

In case of AgGaSe\(_2\) and AgGa\(_{1-x}\)In\(_x\)Se\(_2\) samples we can realize \( z \ll L_{32}/(\delta_1 L_{32} - 1) \) condition when laser pulses are not separated but the second condition (\( z \ll L_{13} \)) is fulfilled only for pulse duration \( \tau_2, 3 \geq 50 \) ps (see Table 2). Thus for \( \tau_2, 3 < 50 \) ps nonstationary regime has to take place and DFG standardized spectrum is described by (3) then halfwidth is

\[ \Delta \Omega_{1r}^q = \Delta \Omega = \Delta \omega_2 + \Delta \omega_3. \]

**Fig. 1.** Phase-matching angle of DFG with AgGaSe\(_2\) and AgGa\(_{1-x}\)In\(_x\)Se\(_2\) by both branches of CO\(_2\) lines.

**Fig. 2.** Standardized spectral density of DFG radiation with AgGaSe\(_2\) crystal for next values of CO\(_2\) laser pulses duration: (1) 100, (2) 50, (3) 25, (4) 10, (5) 5, and (6) 2.5 ps. Crystal length 1 cm.

**Fig. 3.** DFG with (a) AgGaSe\(_2\) and (b) AgGa\(_{1-x}\)In\(_x\)Se\(_2\). Conversion efficiency versus crystal length for the following durations and total intensities of laser pulses: (1) 100 ps, 15 GW/cm\(^2\); (2) 50 ps, 30 GW/cm\(^2\); (3) 25 ps, 60 GW/cm\(^2\); (4) 10 ps, 100 GW/cm\(^2\); and (5) 5 ps, 150 GW/cm\(^2\).
Table 1. Crystal sample characteristics

<table>
<thead>
<tr>
<th>Crystal</th>
<th>ν₁₃, c/cm</th>
<th>ν₃₂, c/cm</th>
<th>δ₁, cm⁻¹</th>
<th>δ₂ = δ₃, cm⁻¹</th>
</tr>
</thead>
<tbody>
<tr>
<td>AgGaSe₂</td>
<td>1.53 × 10⁻¹¹</td>
<td>9.47 × 10⁻¹³</td>
<td>0.7</td>
<td>0.046</td>
</tr>
<tr>
<td>AgGa₁₋ₓInₓSe₂</td>
<td>1.56 × 10⁻¹¹</td>
<td>5.26 × 10⁻¹³</td>
<td>0.69</td>
<td>0.046</td>
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Table 2. Calculation results for the AgGaSe₂/AgGa₁₋ₓInₓSe₂

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<tr>
<th>Laser pulses duration τ₂,₃, ps</th>
<th>Mismatch length L₃₂, cm</th>
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G(Δω₂) Δω₁'' and coefficient G(Δω₂) for selected values τ₂,₃, when z = 1.0 cm (Table 2). Inverse Fourier-transformation of spectral density (3) allowed to define submillimeter pulse duration τ₁. Appreciably that values τ₁ are neighbor for both crystals. At Table 2 it is possible to observe that DFG is accompanied by submillimeter pulse narrowing in comparison with laser pulse duration τ₂,₃ ≥ 25 ps. But nonstationary mixing of the shorter laser pulses (τ₂,₃ < 10 ps) drives to submillimeter pulse extension and production of these duration pulses will be complicated. Evidently it will be hard to form the half-cycle pulse by means of these crystals without additional shortening of submillimeter pulse.

From these spectra we determined halfwidth values Δω₁'' and coefficient G(Δω₂) for selected values τ₂,₃, when z = 1.0 cm (Table 2). Inverse Fourier-transformation of spectral density (3) allowed to define submillimeter pulse duration τ₁. Appreciably that values τ₁ are neighbor for both crystals. At Table 2 it is possible to observe that DFG is accompanied by submillimeter pulse narrowing in comparison with laser pulse duration τ₂,₃ ≥ 25 ps. But nonstationary mixing of the shorter laser pulses (τ₂,₃ < 10 ps) drives to submillimeter pulse extension and production of these duration pulses will be complicated. Evidently it will be hard to form the half-cycle pulse by means of these crystals without additional shortening of submillimeter pulse.

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where P₂, P₃ from 15 up to 150 GW/cm² (suppose that q = 2). Calculation results are presented in Fig. 3. The efficiency amounts to maximum value at crystal end if 25-ps laser pulses with total intensity 60 GW/cm² interacts with the AgGaSe₂ (η = 0.0059) or with the AgGa₁₋ₓInₓSe₂ (η = 0.0083). Calculation results allow to plan generation of submillimeter pulse with power from 32 MW up to 1 GW depending on both duration and power of laser pulse and crystal type, too. The AgGa₁₋ₓInₓSe₂ application promises better DFG efficiency because it’s inverse group-velocity mismatch between laser pulses ν₃₂ is less and corresponding mismatch length value L₃₂ is more than one for the AgGaSe₂. Apart the AgGa₁₋ₓInₓSe₂ advantage is contributed to the larger parameter d_{eff} = d_{se} sin(2θ) where phase-matching angle θ one can see in Fig. 1. We mark that laser total intensity more than 100 GW/cm² using has meaning if one wants to obtain high DFG power by smaller crystal length. If after radiation mixing ending we’ll shorten the produced pulse down to half-cycle duration by proposed method [1] then evidently pulse peak power would be close to primordial value. Also we note that angular spread of the 800-µm beam will be ~0.8 degrees according to [1].
CONCLUSION

Nonstationary mixing of picosecond pulses was investigated for difference frequency generation with AgGaSe$_2$ and AgGa$_{1-x}$In$_x$Se$_2$ crystals. Calculations were executed for the next parameters of CO$_2$ laser pulses: 100, 50, 25, 10, 5 ps duration and correspondingly 15, 30, 60, 100, 150 GW/cm$^2$ total intensity. Spectral characteristics and pulse power versus crystal length are represented for difference wavelength 800.5 nm. Calculation results discovered that DFG efficiency is saturated for intensity more than 60 GW/cm$^2$. It was revealed that submillimeter pulse shortening is inescapable for half-cycle duration attainment with these crystals using. Our results are comparable with similar values obtained [2] for 200-ps pulse and allow to predict higher power generation (up to 1 GW with the AgGa$_{1-x}$In$_x$Se$_2$) in the submillimeter region.

REFERENCES