Numerical simulation of sum frequency generation spectrum in nonlinear crystals with considering dynamics of generation

A A Ionin\textsuperscript{1}, I O Kinyaevskiy\textsuperscript{1}, Yu M Klimachev\textsuperscript{1}, A A Kotkov\textsuperscript{1}, D S Kryuchkov\textsuperscript{1,3}, A M Sagitova\textsuperscript{1,2}

\textsuperscript{1}P.N. Lebedev Physical Institute of the Russian Academy of Sciences, 53 Leninskiy prospekt, Moscow 119991, Russia
\textsuperscript{2}National research nuclear university MEPhI, 31 Kashirskoe shosse, Moscow 115409, Russia
\textsuperscript{3}Moscow Institute of Physics and Technology, 9 Institutskiy per., Dolgoprudny, Moscow Region, 141701, Russia

Abstract. Numerical simulation of sum frequency generation spectrum method was elaborated by considering dynamics of generation of every Q-switched CO laser rotational-vibrational lines. It resulted in decreasing of total power by 25% and disappearing approximately 15% lines in comparison with calculation proceeded without taking dynamics into account.

1. Introduction
Carbon monoxide laser (CO laser) have an extremely broad spectrum containing about a thousand spectral lines in fundamental (wavelength $\lambda\sim4.7$–$8.7\mu m$) \cite{1, 2} and first overtone ($\lambda\sim2.5$–$4.2\mu m$) \cite{1} spectral bands. This laser can run both on a selected single spectral line and in multi-line mode. Efficiency of an electric discharge CO laser being operated in multi-line mode came up to 30-50% for the fundamental band and exceeded 16% for the overtone band \cite{1}. Due to a rich spectrum, multiline CO laser can be successfully applied for multicomponent gas analysis \cite{3}. Multi-line Q-switched CO laser is able to emit dozens of spectral lines (up to 230 lines \cite{4}) in a microsecond pulse. Rich spectrum and high power of multi-line Q-switched CO laser make it very attractive for covering mid-IR range by frequency conversion of radiation in nonlinear crystals. For example broadband two-stage frequency conversion (including both sum and difference frequency generation) was obtained in a single nonlinear ZnGeP\textsubscript{2} \cite{5, 6} and AgGaSe\textsubscript{2} \cite{7} crystal. As a result the broadband (more than one and half octaves) laser system operating at about 670 lines in the wavelength range from 2.5 to 8.3 $\mu m$ and based on one the ZnGeP\textsubscript{2} crystal and one CO-laser was developed \cite{6}. It should be pointed out that second harmonic generation efficiency of mode-locked CO laser in ZnGeP\textsubscript{2} reached 37% \cite{8}.

In \cite{5-7} it was considered that the multi-line Q-switched CO laser generates on all rotational-vibrational spectral lines simultaneously. However, as it was noted in \cite{4} the total microsecond CO laser pulse consisted of laser pulses on 230 laser lines with pulse duration about 200 ns each. That can be treated as non-simultaneous generation of lines in microsecond multi-line pulse. Besides, multi-line Q-switched RF discharge CO laser emitted a lot of spectral lines with different pulse duration, which depends on small-signal-gain of different CO laser transition in the active medium \cite{9}. Time behavior of laser output emitted by multi-line Q-switched CO laser with DC discharge was investigated in \cite{10}, and a spread of generation beginnings for different spectral lines was 250 ns. However, in \cite{10} only vibrational band 8->7 was considered in details. Time behavior of multi-line Q-switched CO laser on different spectral lines is important for some applications, especially for sum frequency generation...
(SFG). Time behavior of the low-pressure multi-line Q-switched CO laser on each spectral line was studied [11]. In present work the influence of spectral-temporal structure of multi-line Q-switched CO laser radiation on broadband SFG in ZnGeP₂ crystal was studied with applying data from [11].

2. Method of SFG numerical simulation
Calculations were proceeded over experimental data described in detail in [11] when the CO laser generation on each spectral line was measured. For repetition rate 70 Hz the CO laser spectrum consisted of 115 spectral lines in the wavelength range from 4.9 to 6.6 μm (Figure 1a). The laser pulses corresponding to rotational-vibrational transitions of the vibrational band 5→4 are presented on Figure 1(b). It was found that some spectral lines were not simultaneously emitted (see Figure 1(b)). Especially, weak lines with rotational quantum numbers J=6, 7 and J=13 were separated in time. It means that they do not interact with each other under SFG in nonlinear crystals.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure1.png}
\caption{Spectrum of multi-line Q-switched CO laser (a) and time behavior of spectral lines in the vibration band 5→4 (b).}
\end{figure}

In the previous works [5-7], where SFG simulation was carried out, the time behavior of multi-line Q-switched CO laser on different spectral lines was not considered. By other words it was considered that SFG power was proportional to the composition of pulse’s amplitudes on two wavelengths \( F_1 \) and \( F_2 \):

\[
P_{SF}^{(1)} \sim \max( P_{F_1} ) \cdot \max( P_{F_2} )
\]  

(1)

It caused significant mistake for separated in the time pulses (as in Figure 1b). For correct SFG simulation, the time behavior of multi-line Q-switched CO laser must be considered. To do it SFG power must be considered as amplitude of composition of pulse’s on two wavelengths \( F_1 \) and \( F_2 \):

\[
P_{SF}^{(2)} \sim \max( P_{F_1} , P_{F_2} )
\]  

(2)

Influence of time behavior of multi-line Q-switched CO laser generation on SFG spectrum can be characterized by \( K \) coefficient, evaluated with:

\[
K = \frac{P_{SF}^{(2)}}{P_{SF}^{(1)}} = \frac{\max( P_{F_1} , P_{F_2} )}{\max( P_{F_1} ) \cdot \max( P_{F_2} )}
\]  

(3)

\( K \) has a physical meaning of SFG lines power decreasing due to taking non-simultaneous lasing of different lines into account. Simulation of \( K \) coefficient for every possible SFG line (for CO laser spectrum Figure 1a) is presented on Figure 2.
Figure 2. Coefficient K, simulated for all possible SFG lines of CO laser spectrum (Fig. 1a).

Figure 2 clearly shows that power of SFG lines significantly decreases, sometimes by more than 10 times, due to non-simultaneous emitting. For second harmonic generation coefficient $K$ equals 1. A spread of coefficient $K$ for SFG lines with higher wavelength is going less. Average $K$ was about 0.75 that is to say integral power of SFG multiline pulse will be reduced by approximately 25%.

To carry out the numerical simulation of SFG in ZnGeP$_2$ for usual experiment conditions, we applied simulation model presented in [12]. This model is based on a plane wave and low conversion efficiency approximation, where the SFG radiation power $P_{SF}$ can be calculated by expression [13]:

$$P_{SF} = \frac{8\pi^2 d_{eff}^2 L^2 P_{F1} P_{F2}}{\varepsilon_0 cn_{F1} n_{F2} n_{SF}^2} \sin^2\left(\frac{\Delta k L}{2}\right)$$

where $d_{eff}$ is the effective nonlinear coefficient; $n$ is refractive index; $\lambda_{SF}$ is SFG wavelength; $\varepsilon_0$ is the dielectric constant; $P_{F1}$ and $P_{F2}$ are the pump laser radiation power for two CO laser lines; $\Delta k$ is the wave mismatch, $L$ is the crystal length; $A$ is a cross-section of the laser beam. Dispersion equations and effective nonlinear coefficient were taken from [13].

The simulation was proceeded for phase-matching angle 47.4 degree and crystal length of 7 mm. The simulation results with (red bars) and without (blue bars) considering the time behavior of multi-line Q-switched CO laser generation is presented in Figure 3.

Figure 3 shows that the influence of spectral-temporal structure of multi-line Q-switched CO laser radiation on general shape of broadband SFG spectrum was insignificant. Calculated SFG spectrum with considering the time behavior of multi-line Q-switched CO laser generation consisted of 1291 spectral lines at 0.01 maximal power level from 6670 possible SFG spectral lines. Calculated SFG spectrum without considering consisted of 1487 spectral lines at 0.01 maximal power level. That means that time separation of interacting pulses decreased a number of SFG lines on 13%.
The integrated over spectrum power of SFG radiation decreased by 18% due to nonsimultaneous generation of CO laser pulses for different spectral lines. Figure 4 presents the enlarged part of SFG spectrum in the region of 2.5μm wavelength.

Figure 4 shows that the decrease of most powerful spectral lines was small. Significant drop of SFG lines power was observed for weak SFG lines (Figure 4). This fact can be explained as weak CO laser lines at generation were separated significantly on time while powerful CO laser lines generated simultaneously.
3. Conclusion
In the first time the influence of time form and duration of Q-switched CO laser rotational-vibrational lines generation was carefully explored. It resulted in elaboration of numerical simulation method used for calculating CO laser SFG spectrum which leaded to decreasing of calculated integral pulse power by 25%. The SFG experiment simulation showed that time separation of interacting pulses of CO laser lines decreased a number of SFG lines by 13% and total spectrum power of SFG radiation by 18%. Future aim is to clarify process of SFG spectrum simulation even more by taking into account atmospheric absorption and compare with upcoming experiments.

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