

New potentialities of a broadband femtosecond optical parametric oscillator for remote sensing multicomponent aerosol and gaseous atmospheric pollutions

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Abstract. An analysis is made of a lidar designed on the basis of a broadband repetitively pulsed IR parametric oscillator for monitoring multicomponent aerosol and gaseous atmospheric pollutions. It is shown that efficient parametric generation of femtosecond IR radiation can be obtained in a scheme using the properties of group velocity matching.

1. Introduction

IR absorption and scattering spectrometry offers promise as an active analytical method for remote sensing multicomponent aerosol and gaseous atmospheric pollutions. Remote sensing of chemical atmospheric pollutions is aimed at determining discharge of chemical substances from industrial enterprises into the atmosphere and rapid mapping of pollutions on large areas for determining the general ecological situation and finding clouds of toxic substances. Note that discharge can be both one- and multicomponent, and its composition may be unknown *a priori*.

Methods of IR spectrometry are realised in practice using differential-absorption lidars (DA lidars), lidars based on differential absorption of scattered radiation (DAS lidars), and IR Fourier gas analysers (FGAs) [1, 2]. In IR FGAs and DA lidars, the backward signal is formed by reflection or scattering of probing radiation from a special retroreflector, a topographic reflector, or an underlying surface in the course of probing from an aircraft. In DAS lidars, the backward signal is formed by scattering from an atmospheric aerosol, which makes probing with spatial resolution possible.

Upon passing a path in the atmosphere, radiation of a DAS lidar carries information on specific spectral features of its transmission caused by selective absorption of pollutants and background molecular components of the atmosphere in the region of their characteristic ‘fingerprints’. To identify each component of the mixture, it should be probed by radiation at least at two frequencies characteristic of this component.

The main problem at the current stage of using DAS lidars for the analysis of multicomponent mixtures consists

in the difficulty of both the technical realisation and the simultaneous spectral and time analysis of the multicomponent response of a medium being studied. Because of this, DA lidars [3, 4] and IR FGA lidars [5] are the most commonly used nowadays, whereas DAS lidars are used predominantly for remote sensing and measuring the concentration of one or two predetermined components under the assumption that other possible components of a mixture do not interfere with the diagnostics.

However, this assumption often fails for mixtures of polyatomic molecules with complex overlapping absorption spectra, and one should increase the number of probe frequencies for the sake of reliable identification, which naturally leads to a further complication of lidars. Nowadays, the greatest promise for the multicomponent analysis with spatial resolution is offered by the DAS lidar based on a chemical HF(DF) laser, because this laser can emit simultaneously 100 lines with energies up to 1 J in the 2.7–4.2- μm region, which overlaps the 3- μm atmospheric transparency window [6]. Using this lidar, one can obtain, owing to the so-called spectral resonances in backscattering [7], information with spatial resolution not only on gas components, but also on the chemical composition of aerosol pollutants.

However, in spite of a relatively large number of frequencies, a large frequency spacing between laser lines (10–40 cm^{-1}) and the accidental character of their coincidence with the absorption bands of possible atmospheric pollutants considerably limit the number of different molecules that can be analysed. Because of this, remote IR FGAs with thermal sources emitting radiation with a continuous spectrum used as probing radiation are the only systems that are currently suitable for multicomponent analysis. However, because of the low spectral brightness of a thermal source, such gas analysers can operate only on a path using special retroreflectors mentioned above, with the path being no longer than several hundred meters [8].

Thus, to carry out efficient remote sensing of multicomponent atmospheric pollutions, one should combine in one device spectral potentialities of an IR gas analyser and the feasibility of spatial localisation of pollutions that is inherent in pulsed lidars operating in the mode of detection and analysis of radiation scattered by an atmospheric aerosol as a distributed retroreflector.

In this paper, we study the feasibility of designing a new class of IR lidars – femtosecond spectroscopic IR lidars based on super-broadband optical parametric oscillators (OPOs) using the properties of group velocity matching [9] and pumped by high-power femtosecond pulses from a repetitively pulsed laser. In our opinion, such a lidar offers much

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promise for expanding potentialities of remote sensing of multicomponent mixtures and approaches spectroscopic capabilities of IR FGAs, with a high brightness of coherent IR radiation produced in it.

Note that the use of broadband picosecond pulses for absorption spectroscopy was discussed earlier in [10] and potentialities of employing high-power femtosecond laser systems for probing parameters of the atmosphere were recently demonstrated in [11].

2. Spectral and energy characteristics of IR radiation produced by efficient parametric generation using the properties of group velocity matching

Today, the most popular femtosecond lasers are Ti:sapphire solid-state lasers producing 10-fs pulses. Investigations aimed at the development of Cr:forsterite lasers of the same class are also carried out [12]. Unique spectroscopic, optical, and physical properties of Ti:sapphire and Cr:forsterite single crystals and modern technologies of fabrication of active laser elements offer considerable promise for generating ultrashort pulses tunable in the 0.75–1.0- μm (Ti:sapphire) and 1.2–1.32- μm (Cr:forsterite) spectral ranges. In our opinion, femtosecond parametric oscillators pumped by Ti:sapphire and Cr:forsterite lasers and using the properties of group velocity matching [9] are of considerable interest and open up new opportunities for scientists working in the field of IR spectroscopy. It is evident that the group length or group velocity detuning are uniquely related to the spectral width of nonlinear conversion, which reaches a maximum in the case of group velocity matching.

In the case of nondegenerate three-frequency interaction, the equality of group velocities is possible only for a pair of interacting pulses. However, in a parametric oscillator pumped by femtosecond pulses and using a long (nano- or subnanosecond) pulse injected at the frequency of the short-wavelength component of a parametric oscillator, one can realise phase matching (the zero group velocity mismatch) for pump pulses and the long-wavelength component of parametric emission. This makes possible high-efficiency conversion of pump radiation into the long-wavelength IR component in the regime when the latter has the maximum spectral width.

As follows from our calculations, by choosing an appropriate nonlinear crystal and the type of parametric interaction for the given pump source, one can propose a scheme for parametric generation of radiation of the prescribed spectral width in the desired spectral range. The calculations were carried out for collinear parametric interaction on the basis of Sellmeyer dispersion equations [13]. For the given pump wavelength, we calculated the angular tuning curves ($k_p = k_s + k_i$) and determined, for each point, the difference of group velocities for the pump pulse and the pulse of long-wavelength parametric emission

$$\Delta u_{\text{gr}} = \left(\frac{\partial k_p}{\partial \omega_p} \right)^{-1} - \left(\frac{\partial k_i}{\partial \omega_i} \right)^{-1}.$$

We also calculated the effective nonlinearity of a crystal.

To estimate the spectral width of IR radiation produced in a parametric oscillator, we use the expression for parametric gain obtained in the approximation of the given field [10]

$$K = \Gamma^2 Z^2 \sinh^2 \left(\Gamma^2 Z^2 - \frac{\Delta k^2 Z^2}{4} \right)^{1/2} \left(\Gamma^2 Z^2 - \frac{\Delta k^2 Z^2}{4} \right)^{-1},$$

where $\Gamma = \sigma_1 \sigma_2 |A_p|^2$; Δk is the wave detuning; Z is the length of a nonlinear medium; $\sigma_{1,2}$ are nonlinear-coupling coefficients; and A_p is the pump amplitude. Results of our numerical calculation are illustrated in Fig. 1, which presents normalised profiles of the parametric gain under group-velocity matching conditions for different schemes and $\Gamma Z = 10$.

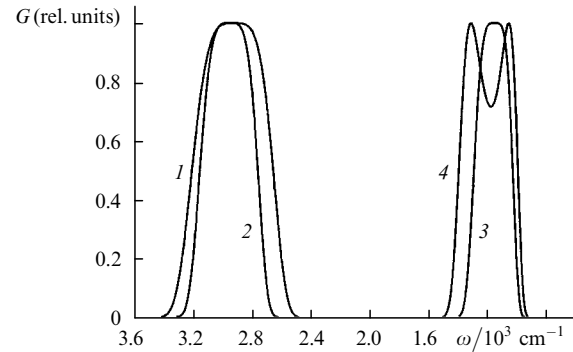


Figure 1. Profiles of parametric gain G for the collinear interaction under conditions of group velocity matching for copropagating waves: (1) parametric gain (type II interaction) in an RTA crystal ($\theta = 45.8^\circ$, $\varphi = 90^\circ$) pumped by a Cr:forsterite laser for a 1.96- μm injected signal; (2) parametric gain (type I interaction) in a CTA crystal ($\theta = 49.9^\circ$, $\varphi = 45^\circ$) pumped by a Ti:sapphire laser for a 1.048- μm injected signal; (3, 4) parametric gain (type II interaction) in a ZnGeP₂ crystal ($\theta = 66.4^\circ$, $\varphi = 0$) pumped by radiation with $\lambda = 2.96 \mu\text{m}$ for a 4.5- μm injected signal and $L_{\text{gr}} = \infty$ (3) and (4) 0.5 cm.

If one has margin for the probe signal energy, inaccurate tuning to group velocity matching is possible. On the one hand, this decreases the parametric-conversion efficiency but, on the other hand, enables a controllable change of the spectrum of probing radiation, which may be useful in some cases. For example, Fig. 1 (curve 4) presents the profile of parametric gain for the scheme characterised by curve 3 but for detuning from the direction of group velocity matching ($L_{\text{gr}} = 0.5 \text{ cm}$).

Thus, to obtain broadband coherent emission with the spectrum almost completely overlapping the 3- μm atmospheric transparency window, one can use OPOs with an RTA crystal (pumped by a Cr:forsterite laser, type II interaction) or with a CTA crystal (pumped by a Ti:sapphire laser, type I interaction) in which group velocity matching is fulfilled for the long-wavelength component of parametric emission produced in the 3000- cm^{-1} region.

Note that systems based on a forsterite laser are preferable because group velocity matching of IR and pump pulses is obtained for type II interaction whose efficiency in crystals belonging to the KTP group is higher by an order of magnitude than that of type I interaction. The addition of the second parametric conversion cascade based on ZnGeP₂ or AgGaSe₂ crystals pumped by OPO radiation in the 3- μm region and using the second harmonic of a CO₂ laser as an injection source enables one to carry out probing in the 10- μm atmospheric transparency window. As an example of spectra typical of hydrocarbon atmospheric pollutants, we present the spectrum of tributylamine (C₄H₉)₃N measured

by us (Fig. 2). Here, the absorption band in the 2800–3000- cm^{-1} region is determined by stretching vibrations of methyl groups CH_3 and the methylene group CH_2 , and the band in the 1000–1400- cm^{-1} region is determined by deformation vibrations of these groups and by skeletal vibrations of C–C and C–N bonds of the tributylamine molecule.

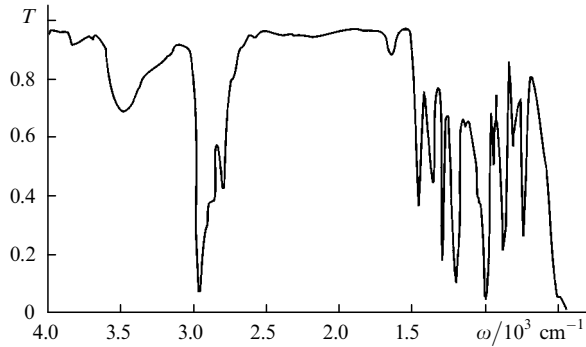


Figure 2. Transmission spectrum of tributylamine.

As noted above, nonlinear optical conversion of ultra-short-pulse frequency under group velocity matching conditions enables one to obtain a high energy conversion. In particular, it follows from Ref. [9] that the efficiency of conversion to the 3- μm region can reach 25%. This gives an energy of the order of 50 μJ in a single pulse of a femtosecond OPO for a quite realistic Cr:forsterite laser system with an energy of the order of 200 μJ in the 70–100-fs pulse. It is also important that such systems operate with repetition rates of the order of 1 kHz, which makes possible efficient signal accumulation during ten seconds, i.e., during a time in which no substantial displacement or deformation of a gaseous or aerosol cloud being analysed takes place.

3. Estimate of the possible range of probing by a femtosecond IR lidar

One can estimate the possible range of probing by a femtosecond IR lidar by substituting into the well-known lidar equation [1] the characteristics of a receiving telescope and a photodetector, the minimum energy detected for obtaining the prescribed signal-to-noise ratio, parameters of an atmospheric aerosol containing molecules being detected, etc. This algorithm is described in sufficient detail in [1]. The threshold laser energy for an IR DAS lidar is determined from the relation

$$E_L^{\min} \approx \frac{2R^2(S/N)_{\min}}{\beta(\lambda_0, R)\xi(R)U^*(\lambda_0)} \exp \left[2 \int_0^R k(\lambda_0, r) dr \right],$$

where U^* is a modified parameter of the system whose value for a solid-state photodetector is given by the formula $U^*(\lambda_0) = A_0 \xi(\lambda_0) c D^* / (A_d B)^{1/2}$; $k(\lambda_0, R)$ is the attenuation coefficient for molecules being probed; $\beta(\lambda_0, R)$ is the coefficient of volume backscattering of the atmosphere; $\xi(R)$ is the probability, based on the geometrical analysis, that emission from a surface element reaches a detector; A_0 is the area of an objective mirror; $\xi(\lambda_0)$ is the spectral transmission coefficient, which, like $\xi(R)$, is approximately equal to unity [1]; D^* is the detection ability of a photodetector; A_d is the area of the sensitive photodetector

element; $B = 1/(2\tau_d)$ is the photodetection bandwidth; and R is the probing range.

The above relation was used in [1] to estimate the minimum energy of a probe pulse required to obtain the 3-km probing range, with the minimum signal-to-noise ratio in a single measurement as large as $S/N = 1.5$, in a DAS lidar based on a DF laser. The minimum energy required for the lidar was 50 mJ, and it was obtained for the following parameters of the receiving system (the parameters are taken from Ref. [14]): $A_0 = 792 \text{ cm}^2$; $D^* = 10^{10} \text{ cm Hz}^{1/2} \text{ W}^{-1}$, $A_d = 0.01 \text{ cm}^2$; $\tau_d = 75 \text{ ns}$; $\beta(\lambda_0 = 3.64 \mu\text{m}) = 8 \times 10^{-9} \text{ cm}^{-1} \times \text{sr}^{-1}$; and $k(\lambda_0, R) = 5 \times 10^{-7} \text{ cm}^{-1}$. The spatial resolution $\Delta R = 75 \text{ m}$ was determined by the 1- μs pulse duration.

In our estimates made for the 3- μm range, we took the detection ability of modern cooled multielement detectors $D^* \approx 10^{11} \text{ cm Hz}^{1/2} \text{ W}^{-1}$ [15], the probing range $R_{\max} = 1 \text{ km}$, and the minimum signal-to-noise ratio $S/N = 1.5$. For the spatial resolution $\Delta R = 100 \text{ m}$ (comparable to the value presented above) to which corresponds the photodetector response time $\tau_d = 750 \text{ ns}$, the minimum pulse energy required for a single measurement is $E_{\min} \approx 55 \mu\text{J}$. However, it should be noted that the value obtained for E_{\min} is the energy required for one spectral channel of the receiving system, whereas the actual laser pulse energy required to retain the prescribed signal-to-noise ratio should be increased in proportion to the number of spectral channels. Starting from the estimate of the typical spectral width of the absorption line of polyatomic molecular pollutants (10–20 cm^{-1}), it is reasonable to choose in the case of a small signal-to-noise ratio the same spectral width of a separate channel. As shown in recent papers [16, 17], in the case of remote sensing of such multicomponent mixtures by an IR FGA, 50- cm^{-1} spectral resolution is a reasonable compromise for minimising the probability of a false alarm and increasing the probability of detecting a cloud of toxic substances.

For the 200–300- cm^{-1} spectral width of the OPO emission and the 20- cm^{-1} width of a spectral channel, the number of spectral channels should be equal to 10–15. If the OPO pulse energy cannot be increased (note that the measurements in all spectral channels are carried out simultaneously), one should accumulate signals and perform averaging over a sufficiently large number of pulses.

For gaseous and aerosol clouds moving with an average velocity of several meters per second, the spatial resolution $\Delta R = 100 \text{ m}$ assumed for lidar measurements allows averaging during several seconds without a substantial loss of accuracy in the measurement of cloud parameters. For the 1-kHz pulse repetition rate, the averaging period contains $10^3 - 10^4$ pulses. This means that one can increase the signal-to-noise ratio by a factor of 30–100, which more than compensates for the spread of pulse energy over 10–15 channels.

Thus, our estimates show that a femtosecond IR lidar is capable of detection and remote sensing of gaseous and aerosol clouds at distances of the order of 1 km with spatial and spectral resolutions of 100 m and 20 cm^{-1} , respectively. One can also show that, in the case where lidar measurements are performed on a path and a signal is scattered from a topographic reflector located at a distance up to 1 km, the scattered signal increases by a factor of about $10^3 - 10^4$, which allows us to determine the integrated content of pollutants on the probing path in a single radiation pulse from an aircraft using scattering from an underlying surface.

Thus, the scheme proposed for parametric generation of femtosecond IR emission using the properties of group veloc-

ity matching is a new approach to the development of spectroscopic lidars. It enables one to obtain broadband coherent radiation in the IR range and carry out remote sensing of atmospheric pollutants with a higher efficiency compared to conventional schemes [18]. Note that parametric frequency up-conversion, which is used for detecting weak signals in the IR region [19], is rather efficient in the case of femtosecond pulses. It is able to provide a higher detection efficiency in comparison with direct detection and can radically improve the spatial resolution owing to the backward-signal gating obtained in this case, which is principally important in finding local point sources of polluting discharges, such as cracks in pipe lines.

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