LIDA exciplex lasers for atmospheric studies

S.E. Kunts, S.V. Mel'chenko, A.N. Panchenko, and V.F. Tarasenko

Institute of High-Current Electronics,
Siberian Branch of the Russian Academy of Sciences, Tomsk

Received March 1, 2001

The experience gained in the development of LIDA exciplex lasers intended for use in lidars is summarized. Peculiarities of application of LIDA-based lidars in atmospheric studies are described, and the efficiency of UV radiation conversion into other spectral regions is considered as a function of different parameters of the exciplex laser radiation. It is shown that LIDA lasers can be efficiently used in atmospheric studies by use of the DIAL technique. The exciplex laser radiation (λ = 308 nm) can serve both for sensing and for pumping dye and/or SRS lasers.

Introduction

The advances, as well as further development of atmospheric optics are connected with the development and improvement of lidar systems for remote sensing of the atmosphere. Lidars based on pulsed lasers are used most widely in the studies of optical characteristics of the atmosphere and atmospheric aerosol. Laser sources of lidar systems should meet the following requirements: (1) lasing at given wavelengths, (2) pulse energy stability, and (3) high efficiency.

Exciplex lasers, being the sources with highest achieved power of pulsed UV radiation, satisfy all these requirements and, in addition, provide for laser emission in wide spectrum. This also can be considered as an advantage when solving problems of atmospheric monitoring by use of DIAL technique. This method is based on measurements of the intensity ratio of two lidar returns at different wavelengths (one falling within the atmospheric transmission window, and the other – within an absorption band of a gas species under study); it is now one of the most widely used methods for measuring the atmospheric concentration of industrial pollutants (SO_2, NO_2) and ozone.

The lidars that allow the maximum number of gases to be detected without significant changes in the laser configuration are obviously most promising. Therefore, the wide spectrum of exciplex lasers is their advantage: on the one hand, using the up-to-date methods of spectral selection it is easy to separate out a narrow line at the wavelength needed, and, on the other hand, the wavelength can be easily changed by spectral tuning. In addition, the output power of exciplex lasers is high enough for these lasers to be considered as an efficient source for radiation conversion into other spectral regions by the methods of nonlinear optics and as a pump source for dye lasers.

In this paper, we summarize the experience gained in the Laboratory of Optical Radiation of the Institute of High-Current Electronics (IHCE) in the development of exciplex lasers for atmospheric sensing.[1–3] The lasers were used both as sources of sounding radiation in lidars and pump sources for nonlinear media of dye lasers.

LIDA laser for lidar monitoring of SO_2 concentration

The LIDA lasers have been developed in the IHCE SB RAS as versatile electric-discharge lasers with the joule per pulse level of the output energy.[4–7] The lasers employ mixtures with the exciplex molecules of XeCl*, XeF*, KrCl*, and KrF*. After minor modifications, these lasers can be successfully used as a source of IR radiation at HF, CO_2, and other molecules.[8]

However, XeCl lasers have found the widest utility in atmospheric studies, therefore hereinafter we will have in mind that LIDA lasers operate at the XeCl laser frequency of 308 nm. The general layout of the LIDA laser is shown in Fig. 1*a*.

In the free-running lasing mode, the spectrum of a XeCl laser consists of four lines corresponding to the *B-X* electronic transitions. The lines of the 0–1 and 0–2 transitions are usually considered as strong, and the lines of the 0–0 and 0–3 transitions are considered as weak, because the intensity of the former exceeds that of the later by an order of magnitude (see Fig. 1*b*, curve *1*).

This spectral structure proved to be very convenient for measuring the atmospheric concentration of SO_2 molecules, because λ = 307.7 nm (weak line) falls within the atmospheric window, and λ = 308.45 nm (strong line) lies in the absorption band of the SO_2 molecule.

However, development of the differential absorption lidar faces a number of additional problems connected, first of all, with obtaining a narrow-band signal at the wavelength of weak line. This signal must have contrast against the broadband noise due to spontaneous emission at the wavelengths of strong spectral lines. Obviously, the sufficiently high-power

0235-6880/01/05 416-05 $02.00

signal can be obtained in this case only with the use of a "master oscillator–amplifier" system, however even in this case the optical arrangement is not trivial. We have undertaken special studies of the effect of pump parameters on the spectral structure of XeCl laser radiation.[9] These studies, enabled us to formulate the following requirements:

– the pump power density should exceed 1 MW/cm^3; this must provide for the gain increment $gL > 6$ (g is the gain for a weak signal; L is the active length);

– the duration of the pump pulse must be longer than the time needed for saturation of strong transitions (~ 20 ns for typical laser parameters);

– the xenon concentration in the mixture must exceed by 1.5–2 times that optimal from the point of view of the maximum output power.

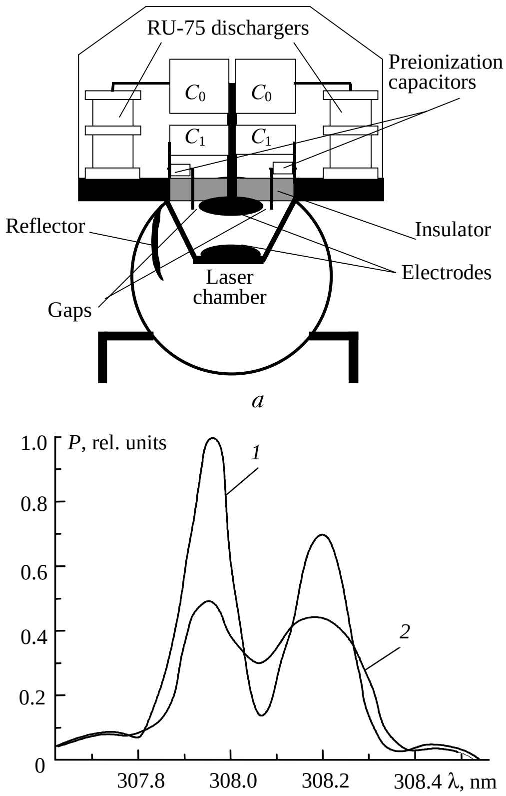

Fig. 1. General layout of a LIDA laser and the spectrum of electric-discharge XeCl laser in free-running lasing mode (*1*) and mode with the increased relative intensity of weak lines (*2*).

If these conditions are fulfilled, the output spectrum is characterized by the increased relative intensity of weak lines (Fig. 1*b*, curve *2*). It is just this case that was realized in the experiment. However, the radiation at strong lines remains still more intense, and efficient separation of a narrow weak line from the spectrum requires an optical cavity having, on the one hand, high selectivity and, on the other hand, minimum losses in the optical elements. Such a cavity is shown schematically in Fig. 2.

The diffraction grating *1* operates in the autocollimation mode, thus providing the reflection coefficient of 0.7 in the first diffraction order; this value is comparable with that of an aluminum mirror. The spatial selection of radiation is provided by the filter consisting of a lens *2* with long focal length ($F = 2$ m) and the diaphragm *3* of 30–100 μm in diameter placed at its focus. Because of the high power density of the radiation formed in the cavity, the output mirror *5* was separated by 10 cm from the focus and placed after the re-collimating lens *4*. Such a design of the cavity has allowed the laser active volume (3.5×1.2×60 cm) to be used most fully and the energy of 3 mJ (line width of 0.25 , λ = 307.7 nm) and 30 mJ (line width of 0.4 , λ = 308.45 nm) to be obtained at the output from the master oscillator. The signal-to-noise ratio (contrast) exceeded 100 at the distance of 2 m from the laser exit window, because the contrast increases proportionally to the squared distance.

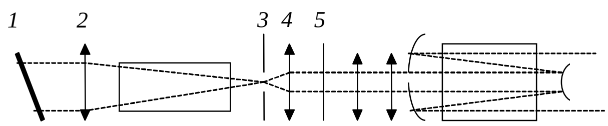

Fig. 2. Optical arrangement of master oscillator and amplifier operating in the mode of injection synchronization for obtaining high-power narrow-band high-contrast radiation at the wavelength of the weak XeCl* spectral line.

The LIDA laser was also used as an amplifier of the selected narrow-band signal. Two amplification modes were studied: single-pass mode and the mode of injection synchronization. In the latter case, the competition between the spontaneous noise at the wavelength of strong lines and the signal at the wavelength of a weak line in the amplifier medium is of critical significance.

To provide for maximum contrast at the output, the optimal delay between turning-on of the master oscillator and the amplifier must be ~ 30 ns. This allows the signal-to-noise ratio to be 5–10 at the distance of 2 m from the amplifier exit window.

Higher contrast of radiation has been achieved in the single-pass amplification mode. In this case, it proved to be possible to increase the contrast by changing the geometry of the beam propagation in the amplifier, and the contrast of 100 has been obtained when amplifying a divergent beam (Table 1). This mode was selected as most suitable for solution of sensing problems, and in the final version it gave the output energy of 120 mJ (line width of 0.25 , λ = 307.7 nm) and 250 mJ (line width of 0.4 , λ = 308.45 nm). The optimal delay between the master oscillator and the amplifier was 5–10 ns.

Table 1. Output energy and contrast of single-pass amplifier depending on the geometry of propagation of the amplified beam

Beam type	$E_0 = 0.3$ mJ		$E_0 = 3$ mJ		$E_0 = 15$ mJ		$E_0 = 30$ mJ	
	E_L, mJ	K	E_L, mJ	K	E_L, mJ	K	E_L, mJ	K
1	4	4	30	23	115	51	183	66
2	3	18	20	51	60	79	90	94
3	3.5	13	27	43	82	75	120	98
4	3	8	18	40	55	71	84	92

Note. E_0 is the amplifier input energy; E_L is the amplifier output energy; K is signal-to-noise ratio. Beam types: parallel beam *1*, beam focused at the beginning of the active medium *2*, beam focused 30 cm far from the beginning of the active medium *3*, beam focused at the center of the amplifier active medium *4*.

MDL laser system for lidar diagnostics of NO_2

The advent of high-power exciplex lasers has stimulated strongly the development of high-power tunable dye lasers. Exciplex-laser-pumped dye laser systems are capable of emitting radiation in the spectral region from 430 to 550 nm with the mean power thrice as high as that of similar systems pumped with the emission of Nd–YAG lasers.[10] Such systems find wide application in various fields, including atmospheric sensing. In particular, they can be used for solution of a very urgent, especially for big cities, problem on measuring the NO_2 concentration in the atmosphere. For this purpose, the radiation at two wavelengths $\lambda = 446.9$ nm (in the absorption band) and $\lambda = 448.2$ nm (in the atmospheric transmission window) is used. Rather high-power and narrow-band radiation (150 mJ, 0.1) has been obtained at these wavelengths in the MDL dye (coumarin 2) laser system (Fig. 3).

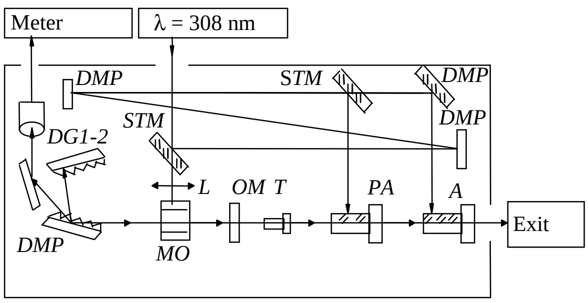

Fig. 3. Optical arrangement of the MDL dye laser system[3]: master oscillator *MO*, preamplifier *PA*, amplifier *A*, semitransparent mirror *STM*, mirrors *DMP*, diffraction gratings *DG1-2*, output mirror *OM* of the master oscillator, telescope *T*, and a cylindrical lens *L*.

This laser system has been especially developed at the Laboratory of Laser Physics of V.D. Kuznetsov Siberian Physical-Technical Institute at Tomsk State University to be used with the LIDA laser as a pump source. With this system, the conversion efficiency has been studied as a function of the pump pulse duration (Fig. 4).

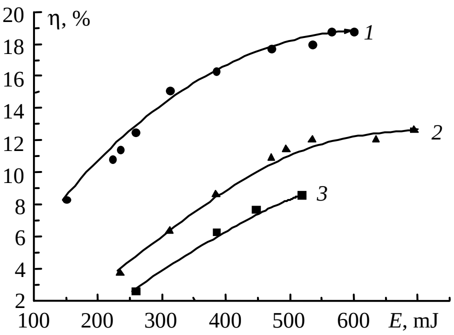

Fig. 4. Efficiency of the MDL-03 laser as a function of the pump energy from a XeCl laser with the pulse duration of 40 (*1*), 55 (*2*), and 75 ns (*3*) (Ref. 3).

It turned out that the conversion efficiency decreases as the pulse becomes longer. This can be explained by mismatching of the optical delay inside the MDL laser (the time between the appearance of lasing in the master oscillator and the arrival of pump pulse to the amplifiers). The point is that the master oscillator of the MDL laser has been developed for the pump pulse of 40 ns duration with the leading edge ~ 10–15 ns as steep. So, the change of these parameters to 75 and 30 ns, respectively, calls for the increase in the optical delay and, consequently, the corresponding change in the laser optical arrangement.

The studies of the effect of pump beam divergence on the efficiency of MDL laser showed that if the exciplex laser is used for pumping a dye lasers, such parameters of the former, as divergence and aperture are also important.

Obviously, the aperture of the pump beam coming to the amplifiers should coincide with their input aperture, and, therefore, the radiation divergence should be sufficiently small for the delay time to be not enough for the aperture to exceed the given value (in our case, 48×16 mm; this corresponds to the divergence of 3×1.5 mrad). The results of these studies are summarized in Table 2.

Table 2. Total pump energy E_p^{XeCl} of MDL-3 laser, input energies of the laser parts E_p^{MO}, E_p^{PA}, and E_p^{A}, and the efficiency of the dye laser; XeCl laser pulse duration $\tau_p = 40$ ns

E_p^{XeCl}, mJ	E_p^{MO}, mJ	E_p^{PA}, mJ	E_p^{A}, mJ	η, %
390	59	117	215	13
195	28.5	58	108	7.8
283	42.5	85	156	11.8
283	59	117	107	7

The LIDA lasers (master oscillator and amplifier) and the MDL laser system have been used in the United Space Device Corporation (Moscow) for the development of a lidar system to measure SO_2 and NO_2 concentrations. Measurements of the SO_2 concentration involved only the exciplex lasers, whereas in measurements of the NO_2 concentration the XeCl laser radiation was used for pumping an MDL system. Since 1997 this lidar system has been successfully used in monitoring of NO_2 in the air basin of Moscow.[3]

SRS conversion of exciplex laser radiation

The radiation of exciplex lasers can be converted into other spectral regions by using various nonlinear-optics methods. The most widely used and efficient method of such conversion is stimulated Raman scattering (SRS) in compressed gases and metal vapors. SRS yields a set of spectral components shifted relative to each other by the vibrational quantum characteristic of a scattering medium molecules, while acquiring all spatial characteristics of the pump radiation. Obviously, such a spectrum is ideal for remote sensing, if one succeeds in selecting the needed pair of wavelengths.

The XeCl (XeF) lasers with the SRS conversion have found the widest application in monitoring of stratospheric ozone.[11,12] Actually, in this case, the wavelength $\lambda = 307.9$ nm falls within the absorption band, and the wavelength $\lambda = 353.2$ nm lies in the atmospheric transmission window. This corresponds to the first Stokes component at scattering in hydrogen. If necessary, the SRS cell in such a lidar can be replaced with a XeF laser operating at $\lambda = 351$ nm.

Although these works (development of lidars based on SRS conversion) were not performed at the Laboratory of Optical Radiation, we have gained a wide experience in the use of the LIDA lasers as a pump source for obtaining the SRS[13] shifted radiation and this experience allows us to consider the LIDA lasers as promising in this field.

The exciplex laser radiation has some peculiarities: first, a rather wide spectrum and, second, high divergence of the output beam due to small number of passes inside the cavity. As was revealed in Ref. 14, the width of the pump spectrum is not important from the viewpoint of the SRS efficiency, if the power density in the focal spot of a lens (conversion region) exceeds the critical value (~ 40 MW/cm^2). It is the divergence, which determines the homogeneity of the intensity distribution in the focal spot and strongly affects the conversion efficiency (Fig. 5). Thus, the radiation with the divergence close to the diffraction-limited one is preferable at the laser output. This, in its turn, leads to the drop of the output energy and requires development of "master oscillator–amplifier" laser systems. If the high-quality pump beam is used, the maximum total quantum efficiency of conversion to the Stokes components achieves 90% already at the pulse energy ~ 20 mJ. The energy distribution among the components is determined by the length of the interaction zone (focusing geometry) and the parameters of the active medium. Consequently, varying the experimental conditions, we can obtain domination of one or other component in the spectrum (as a rule, three first main Stokes components are considered).

When using the seed signal at the needed wavelength, the efficiency of conversion into the given component can increase up to 95–97%. All the studies described above (including SRS in hydrogen, methane, their mixture, and lead vapor) have been systematically conducted at the Laboratory of Optical Radiation in 1980–2000. In these studies, the corresponding equipment has been developed, and the record conversion efficiency has been achieved.[15–17]

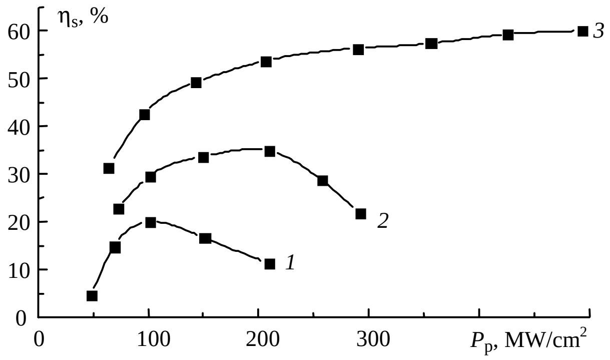

Fig. 5. Efficiency of SRS conversion into the Stokes region as a function of the pump power density at the beam ($\lambda = 308$ nm) divergence of 1 (*1*), 0.6 (*2*), and 0.4 mrad (*3*); hydrogen pressure of 20 atm (Ref. 13).

In conclusion, we would like to note that in addition to atmospheric sensing applications by use of DIAL method, UV lasers are widely used in atmospheric studies by the method of elastic scattering, in measurements of the vertical temperature distribution, and others (see, for example, Ref. 18). The parameters of the LIDA lasers make them promising for these measurements as well, and both the ground-based and airborne (spaceborne) measurements are possible.

References

1. K.S. Gochelashvili, M.V. Kabanov, V.B. Kaul, T.N. Kopylova, G.V. Maier, S.V. Mel'chenko, A.N. Panchenko, A.M. Prokhorov, V.F. Tarasenko, and E.N. Tel'minov, Laser Phys. **2**, No. 5, 802–804 (1992).
2. V.B. Kaul, S.E. Kunts, S.V. Mel'chenko, and V.F. Tarasenko, in: *Proc. Conf. Lasers'97* (STS Press, 1998), pp. 761–764.
3. V.F. Tarasenko, E.H. Baksht, T.N. Kopylova, S.E. Kunts, R.T. Kuznetsova, G.V. Maier, S.V. Mel'chenko, A.L. Onitschenko, A.N. Panchenko, L.G. Samsonova, V.A. Svetlichnyi, and E.N. Tel'minov, in: *Proc. Conf. Lasers'98* (STS Press, 1999), pp. 525–532.
4. S.V. Mel'chenko, A.N. Panchenko, and V.F Tarasenko, Opt. Commun. **56**, No. 1, 51–52 (1985).
5. S.V. Mel'chenko, A.N. Panchenko, and V.F. Tarasenko, Pis'ma Zh. Tekh. Fiz. **12**, No. 3, 171–175 (1986).
6. V.S. Verkhovskii, M.I. Lomaev, S.V. Mel'chenko, A.N. Panchenko, and V.F. Tarasenko, Kvant. Elektron. **18**, No. 11, 1279–1285 (1991).

7. A.N. Panchenko and V.F. Tarasenko, Kvant. Elektron. **20**, No. 7, 663–664 (1993).
8. E.H. Baksht, A.N. Panchenko, and V.F. Tarasenko, IEEE J. Quantum Electron. **35**, No. 3, 261–266 (1999).
9. V.B. Kaul, S.E. Kunts, and S.V. Mel'chenko, Kvant. Elektron. **22**, No. 6, 555–558 (1995).
10. V.Ya. Artykhov, I.V. Sokolova, T.N. Kopylova, G.V. Maier, and V.F. Tarasenko, in: *Proc. Conf. Lasers'94* (STS Press, 1995), pp. 208–215.
11. J.S. McDermid, T.D. Walsh, A. Deslis, and M.L. White, Appl. Opt. **34**, 6201–6211 (1995).
12. V. Kempfer, W. Karnuth, R. Lotz, and T. Trickl, Rev. Sci. Instrum. **65**, 3145–3164 (1994).
13. S.V. Mel'chenko and V.F. Tarasenko, Izv. Vyssh. Uchebn. Zaved., Ser. Fiz., No. 3, 90–116 (1998).
14. S.A. Akhmanov, Yu.E. D'yakov, and L.I. Pavlov, Zh. Eksp. Teor. Fiz. **66**, No. 2, 520–527 (1974).
15. S.V. Mel'chenko, A.N. Panchenko, and V.F. Tarasenko, Kvant. Elektron. **13**, No. 7, 1496–1500 (1986).
16. G.S. Evtushenko, S.V. Mel'chenko, A.N. Panchenko, and V.F. Tarasenko, J. Russian Laser Res. **15**, No. 1, 49–54 (1994).
17. V.B. Kaul, S.E. Kunts, and S.V. Mel'chenko, Kvant. Elektron. **25**, No. 1, 65–68 (1998).
18. V. Raimondi, G. Cecchi, L. Pantani, and R. Chiari, Appl. Opt. **37**, 1089–1099 (1998).