

Two-frequency lidar based on an ammonium laser

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Abstract. A two-frequency differential absorption lidar is proposed in which the reference beam is formed from CO₂ laser radiation and the radiation frequency of the working beam is tuned consequently to the lines of the NH₃ laser spectrum. It is shown that this lidar can be used to measure the concentration of freons and some other atmospheric pollutants as low as ~ 1 ppm with an error of about 20%.

1. Introduction

Comprehensive studies of a repetitively pulsed NH₃ laser resonantly pumped by CO₂ laser radiation (see, e.g., Ref. [1]) made it possible to design a high-power source of coherent radiation in the 11–14- μ m spectral range, which is close in properties (power, efficiency) to an extensively used CO₂ laser. Using an ammonium laser, some problems associated with the resonance interaction of radiation with matter were solved, in particular, the separation of carbon and chlorine isotopes by selective dissociation of CCl₄ molecules [2] and the dissociation of the UF₆ molecule by exciting its composite vibrations [3].

Of particular interest is the possibility of using NH₃ laser radiation in differential absorption lidars (hereafter, referred to simply as lidars) [4–6]. A unique spectral range of this laser is suitable for probing substances that cannot be detected using CO₂ lidars, such as various fluorine- and chlorine-containing molecules (freons) able to destroy the ozone layer of the Earth's atmosphere [6], dioxins [8], and some other substances. Note that because NH₃ and CO₂ lidars have close spectral ranges, the main parameter of these devices, namely, the detection range, is approximately the same, i.e., its value for an output laser energy of ~ 1 J may lie in the range from several hundred meters to several kilometres.

The application of an NH₃ laser for lidars was first proposed in Ref. [5]. Later on [8], a lidar using an NH₃ laser with a nonselective cavity, which simultaneously produces coherent radiation at several lines, was studied. In this case, one can determine the absorption spectrum of a substance using a single laser pulse, which is an obvious advantage of this device. However, because of the competition of laser transitions, the radiation energy in each separate line is

considerably lower than the energy in a laser with a selective cavity, which limits the lidar detection range. Moreover, the spectral range of radiation in the case of multifrequency lasing is considerably narrower than the range of an NH₃ laser with a selective cavity.

In this paper, the design of a two-frequency lidar free of these disadvantages is considered. Its specific feature is that an object is illuminated with two-frequency radiation. One of its parts is formed from the pump laser radiation, and the other part represents tunable NH₃ laser radiation. Radiation at the pump frequency represents a reference beam, and tunable radiation is used as a working beam. In the case of simultaneous lasing at reference and working frequencies, strict requirements to the repetition rate of probing radiation are absent. In the case of alternate lasing at these frequencies, the repetition rate should be no less than 100 Hz [9].

In this paper, we analyse the possibility of using the two-frequency lidar based on an ammonium laser for determining the chemical composition of atmospheric pollutants and measuring their concentration.

2. Detection of atmospheric pollutants

The identification of impurities and the measurement of their concentration with a two-frequency lidar are carried out by the well known method of double-beam absorption spectroscopy [10]. Like in a double-beam spectrograph, the absorption of radiation by impurity molecules is determined by comparing the intensities of the working and reference beams. The radiation frequency of the working beam is chosen close to one of the absorption lines of an impurity being detected. The reference beam is coincident in space with the working beam, but it is not resonantly absorbed. In this case, the energy loss through scattering and reflection for the reference beam is approximately the same as the loss for the working beam. By comparing the intensities of the reference and signal beams at the output of a lidar and after passing the probing path in the atmosphere, one can eliminate the effect of nonresonant losses (turbulence and thermal density fluctuations in the atmosphere) on the accuracy of impurity concentration measurements. Note that the difference in coefficients of atmospheric volume backscattering for the reference and working beams is not important because the impurity concentration is determined by the ratio of intensities of working and reference beams near the absorption line centre and at its wings, where resonance absorption is almost absent.

The reference and working beams should be sent along the probing path either simultaneously or with a time delay during which parameters of the atmosphere on the path remain unchanged. In one of the versions of a lidar based

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on a CO₂ laser, alternate lasing at two wavelengths is used [9]. In this case, the time interval between radiation pulses should be less than 10 ms, which imposes rigid engineering requirements both on the laser radiation source and on the time response of a device used for laser frequency tuning. In a multifrequency lidar based on an ammonium laser, lasing on several frequencies takes place simultaneously [7], but the competition of laser transitions causes a decrease in the spectral density of laser radiation. Moreover, the necessity of spectral analysis of multifrequency signals requires that complex spectral instruments and high-power computers be used. Because of this, it seems attractive to use a two-frequency lidar based on an ammonium laser [5, 6].

In this lidar, the reference beam is produced by a CO₂ laser, which is also used to pump an NH₃ laser, and the radiation frequency of the working beam is tuned to the lines of the NH₃ laser spectrum.

Comparison of intensities of the reference and working beams is carried out by digital processing of electric signals from photodetectors measuring radiation intensity in each of the beams at the output of the lidar. The signals entered the receiving lidar telescope after passing the probing path in the atmosphere. The signal detection system took into account the difference in the radiation pulse form of the reference and working beams and separated out the desired signal proportional to the radiation energy resonantly absorbed by impurity molecules in an atmospheric layer being probed.

Probing radiation can be directed into the receiving telescope of the lidar in two ways. In the first case, the radiation source and the receiving unit are positioned at opposite ends of the measuring path (bistatic scheme). In the second case, the telescope receives scattered radiation or radiation returned back by a corner reflector or a topographic target. In this case, the laser and the receiving unit are located in the same place (monostatic scheme), and the probing-path length is equal to the double distance between the lidar and the reflector. The maximum lidar sensitivity can be obtained in the bistatic scheme because the energy losses in the scheme using a corner reflector or a topographic target decrease the sensitivity. The greatest problems are associated with detecting scattered radiation, and further analysis will be related to lidars operating in the bistatic scheme or using reflectors. The concentration of small amounts of impurities in the atmosphere detected by a lidar is calculated by the formula

$$n = A(\lambda_1, \lambda_0) \frac{1}{L} \ln \frac{I(\lambda_0)}{I(\lambda_1)}, \quad (1)$$

where L is the probing-path length; λ_1 and λ_0 are the wavelengths at the absorption line centre and in the wing, where absorption is almost absent, respectively; $A(\lambda_1, \lambda_0) = n_0 L_0 / \ln[I_0(\lambda_0)/I_0(\lambda_1)]$ is an element of the 'spectral portrait' of a molecule obtained for the known thickness L_0 and concentration n_0 of an absorbing layer; $I(\lambda_0, \lambda_1) = I_w / (I_{\text{ref}} k)$; I_w and I_{ref} are the average intensities of the working and reference beams in the plane of the receiving-telescope aperture; and k is the ratio of these intensities at the lidar output.

To obtain efficient monitoring of atmospheric pollutants and reveal their sources, one should provide reliable detection of impurities with a concentration of $\sim 10^{-2}$ of the maximum permissible concentrations specified by sanitary standards [11], which corresponds to $\sim 10^{13} \text{ cm}^{-3}$ ($\sim 1 \text{ ppm}$) for the majority of pollutants. It is assumed in formula (1) that the ratio of intensities of the working and reference beams passed along

the probing path is changed only due to resonance absorption. Changes caused by any other reason give an error in the calculation of impurity concentration.

In any lidar using double-beam absorption spectroscopy, errors of this kind are caused by the spatial mismatch of reference and working beams because of their different divergences or different optical axes of these beams. For the lidar based on an ammonium laser, which is proposed here, it is of principal importance to reveal and eliminate such errors because the reference and working beams are generated by different active media found in different optical cavities, and therefore they may have different radiation patterns.

Consider the effect of mismatch of radiation patterns of reference and working beams on the accuracy of impurity concentration measurement. One of the reasons of this mismatch is that the beams may have different divergences. The resulting change in the ratio of intensities of the reference and working beams in the plane of the receiving-telescope aperture causes errors. If their divergences differ by $\Delta\alpha$, the error Δn_1 in the measurement of impurity concentration can be estimated by the formula

$$\Delta n_1 \approx \frac{2}{\sigma x} \frac{\Delta\alpha}{\alpha}, \quad (2)$$

where α is the working-beam divergence; σ is the absorption cross section of a pollutant; and x is the corresponding path length.

To measure the impurity concentration of $\sim 1 \text{ ppm}$ with a relative error lower than 20%, one should provide the identity of divergences of the reference and signal beams with an error below 10^{-2} , and the probing-path length for impurities with absorption cross sections of $\sim 10^{-18}$ and $\sim 10^{-19} \text{ cm}^2$ should be not smaller than 0.2 and 2 km, respectively.

The angular offset of the reference beam axis with respect to the working-beam axis is the second source of errors in the measurement of impurity concentration because it also causes a change in the ratio of intensities of the reference and working beams in the plane of the receiving-telescope aperture, which has no relation to resonance absorption.

In the case of a Gaussian intensity distribution of radiation in the laser beam aperture, one can estimate the error Δn_2 caused by the nonzero angle θ between the optical axes of beams with the same divergence in the following way:

$$\Delta n_2 \approx \frac{1}{\sigma x} \left(\frac{\theta}{\alpha} \right)^2. \quad (3)$$

For the measurement of impurity concentration of $\sim 1 \text{ ppm}$ with a relative error below 20% under the conditions specified above, the ratio θ/λ should not exceed ~ 0.1 .

In the measurement of the impurity concentration, the errors Δn_1 and Δn_2 are added. If the difference $\Delta\alpha$ of radiation divergences for the reference and working beams remains unchanged from pulse-to-pulse, the error Δn_1 is systematic and can be taken into account by signal processing. An uncontrollable deviation of the directivity diagram axis of the reference beam from the working-beam axis gives a random error Δn_2 , which should be taken into account in lidar measurements of the concentration of impurities in the atmosphere. Note that the path length can be determined with a low error (smaller than 5%) using a laser range finder, so that the effect of the error of path length measurement on the total error can be reduced to a minimum.

Thus, the optical scheme of a lidar should provide the coincidence of axes of the reference and working laser beams

with the error determined by formulas (2) and (3). In the lidar based on an ammonium laser, two-frequency laser radiation satisfying the given requirements is made by a special optical scheme.

3. Optical scheme of a two-frequency lidar

We studied several versions of optical schemes of a two-frequency lidar [6]. Because the construction of a mobile lidar is intended, the criteria governing the choice of an optical scheme are its simplicity and the reliability of a device under conditions of increased vibrations and changes of ambient temperature.

Fig. 1 presents the optical schematic of a monostatic mobile two-frequency lidar, which was developed taking into account the conclusions presented in the previous section. All optical elements of the laser system are mounted on an optical plate with vibration-proof insulation. The optical cavity of the CO₂ laser is formed by a diffraction grating 1, which works in the autocollimation geometry, and a plane mirror 4. The diffraction grating 1 separates out the line 9R(30) in CO₂ laser spectrum. This line is used for optical pumping the NH₃ laser. The NH₃ laser cavity is formed by a mirror 4, a diffraction grating 2, and a mirror 6. The NH₃ laser tuning over individual vibrational-rotational transitions of the NH₃ molecule is performed by rotating the mirror 6 about the vertical axis.

The mirror 4 represents a multilayer dielectric coating deposited onto a plane-parallel BaF₂ plate. The coating has an optimum reflectivity for CO₂ laser radiation and totally reflects radiation in the wavelength range of the NH₃ laser. This is a common mirror used in both laser cavities, which provides a high-accuracy coincidence of axes of the reference and working beams. Apertures 8 and 9 are used for matching the beam divergences. Two-frequency radiation is outcoupled from the NH₃ laser cavity by a plane-parallel plate 10 made of NaCl and is directed along the probing path by mirrors 5 and 7. Mirror 7 is rigidly coupled with the lidar receiving telescope, which is formed by mirrors 11 and 12. Photodetectors 13 and 14 detect the radiation intensities of reference

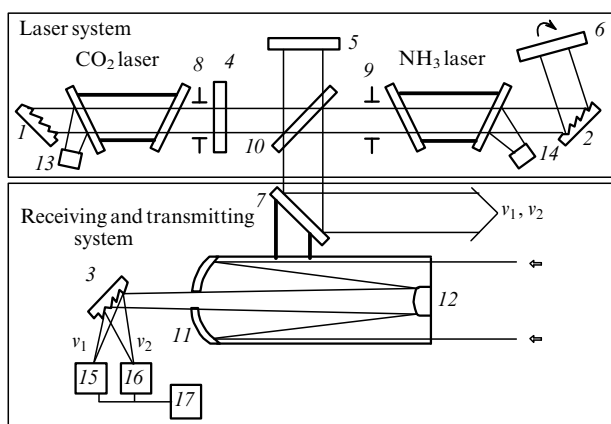


Figure 1. Optical schematic of the two-frequency lidar based on an ammonium laser. (1–3) Diffraction gratings; (4) multilayer dielectric mirror; (5–7) totally reflecting mirrors; (8,9) apertures; (10) plane-parallel plate; (11, 12) receiving-telescope mirrors; (13–16) photodetectors; (17) information processing unit; v_1 and v_2 are the radiation frequencies of the reference and working beams.

and working beams. The radiation that has passed the probing path in the atmosphere is focused by the receiving telescope onto photodetectors 15 and 16. The reference and working beams are spatially separated by a diffraction grating 3.

To target the receiving and transmitting system on an object, to tune the NH₃ laser frequency, and to process output photodetector signals, a computerised unit on the basis of a personal computer is used, which controls the system and processes information. Digital processing of signals from photodetectors 13–16 makes it possible to separate the desired signal, which is uniquely related to the average concentration of an impurity being detected on the probing path. Information on the composition and concentration of atmospheric pollutants is shown in the graphical form on the display and can be transferred by a radio modem to an information acquisition and processing centre.

The optical scheme described above enables one to obtain a divergence of $\sim 5 \times 10^{-3}$ rad (the angle is measured at HWHM) for the reference and working beams, with an error of matching their axes and divergences being not greater than $\sim 10^{-4}$. This means that using a probing path as short as 2 km, one can measure concentrations of the majority of pollutants in the atmosphere, whose lines fall in the spectral range of the NH₃ laser, of the order of 10^{13} cm⁻³ (~ 1 ppm) with a relative error not higher than 20%.

4. Application of the two-frequency lidar for detecting atmospheric pollutants

Fig. 2 presents the relative energies of spectral components of an NH₃ laser. They were obtained using a TEA CO₂–laser operating on the 9R(30) line and producing 2.5 μ s 7.5 J pulses as a pump source. CO₂ laser radiation was directed into a cell filled with a 1 : 75 ammonium-nitrogen mixture. Its total pressure was 60 Torr, and the cell was 1.8 m long. CO₂ laser radiation was resonantly absorbed by ammonium molecules, which made it possible to obtain inverse population for a large number of vibrational-rotational transitions of the first vibrational state of the ammonium molecule. The NH₃ laser cavity was sequentially tuned to each lasing line.

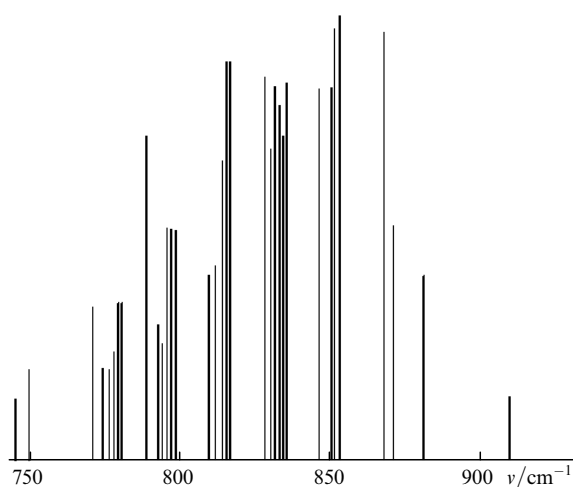


Figure 2. Emission spectrum of the ammonium laser.

For a CO₂ laser energy density of 0.7 J cm⁻² at the input of the ammonium cell, the conversion efficiency for the strongest lines reached 20%. The laser radiation energy in a single line reached 1.5 J. Emission pulses measured for different spectral components were 0.5–2 μs long. One can see from Fig. 2 that the 745–910 cm⁻¹ spectral range contains many lines emitted by the NH₃ laser. This spectral range contains strong absorption lines of many molecules whose concentration in the atmosphere should be monitored. Some atmospheric pollutants, whose absorption lines coincide with emission lines of the NH₃ laser, are presented in the table.

For the majority of substances mentioned here, data on the absorption cross section at the NH₃ laser lines are not available in the literature. This calls for the measurement of absorption of NH₃ laser radiation by substances that are supposed to be detected by the lidar proposed here. The aim of these measurements is to obtain the 'spectral portrait' of each molecule, which represents a set of absorption cross sections for several (3–5) spectral components of the NH₃ laser under conditions where absorption lines of each molecule are broadened in collisions with molecules of gases contained in the atmosphere. The data base should also contain information on the maximum permissible concentrations of unfavourable substances in the atmosphere, which is required for rapid processing of experimental data.

One can see from the table that some substances have absorption peaks at close frequencies, and therefore to identify molecules of a certain impurity, one should have its 'spectral portrait' obtained with an NH₃ laser under conditions used for the identification.

A substantial advantage of the two-frequency lidar compared to the similar multifrequency lidar proposed in Ref. [7] is that sequential lasing on spectral components of the NH₃ laser in a selective cavity makes it possible to obtain the maximum number of these components. Because of the competition of laser transitions in a nonselective cavity, lasing is observed at 4–5 strongest lines at most, and one is forced to change the gas pressure and the ratio of components in a gas mixture of the NH₃ laser to change the spectral composition of laser radiation. For the two-frequency lidar proposed here, there is no problem of varying parameters of a gas mixture for controlling the emission spectrum because the optimum composition and pressure of a gas mixture are the same for all lines of the spectrum presented in Fig. 2.

5. Conclusions

The optical scheme developed for a mobile monostatic lidar provides high-accuracy spatial matching of the reference and working beams, which is required for decreasing the errors appearing in impurity concentration measurements by double-beam absorption spectroscopy. This lidar can be used to measure the concentration of freons and some other atmospheric pollutants as low as ~ 1 ppm with an error not greater than 20%.

The use of the lidar proposed here for monitoring unfavourable substances in the atmosphere calls for data bases containing 'spectral portraits' of molecules being identified using an NH₃ laser. Moreover, the problem calls for the measurement of reflection coefficients of materials used for possible topographic targets in the wavelength range of the NH₃ laser. At present, we are carrying out experiments in these areas of investigation.

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Table 1. Some atmospheric pollutants whose absorption lines coincide with emission lines of the NH₃ laser

Substance	Molecule	Vibration frequency /cm ⁻¹	Laser transition in the NH ₃ in molecule	Lasing frequency /cm ⁻¹
Difluorochlorobromomethane	CF ₂ ClBr	872	<i>aP</i> (3,1)	872.6
Fluorochloromethane (Freon 11)	CFCl ₃	847	<i>sP</i> (6, <i>k</i>)	847.4
Difluorochloromethane (Freon 22)	CHF ₂ Cl	812	<i>aP</i> (6, 4)	812.0
Dibromomethane	CH ₂ Br ₂	810	<i>aP</i> (6, 5)	809.7
Carbon tetrachloride	CCl ₄	796	<i>aP</i> (7, 3)	796.0
Trifluorochloromethane (Freon 13)	CF ₃ Cl	781	<i>aP</i> (8,1)	780.4
Difluorodichloromethane (Freon 12)	CF ₂ Cl ₂	780	<i>aP</i> (8,1)	780.4
Trichloromethane	CHCl ₃	774	<i>aP</i> (8, 5)	774.0
Chlorotribromomethane	CClBr ₃	745	<i>aP</i> (9, 8)	745.3

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