LASER SOUNDING OF ATMOSPHERIC GASES USING THE EFFECTS OF STIMULATED RAMAN-SCATTERING AND RESONANCE ABSORPTION

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A possibility of using the SRS on N_2 and O_2 molecules is discussed aiming at an increase of sounding range of DIAL technique for measuring minor atmospheric gases. Some estimates of the sounding range enhancement when the absorption of radiation by a gas exceeds its amplification due to the SRS are obtained for typical parameters of radiation and media.

Differential absorption and scattering technique is widely used for sounding atmospheric gases along extended paths.¹ A sounding range of this method is determined by the absorption coefficient of a gas under study and for strongly absorbing components, like NO₂, CH₄, and organic molecules, this can be insufficient for practical use because of the fast attenuation of the sounding pulse energy along the propagation path. The use of weak absorption lines as the reference ones enables one to increase the sounding range but this way has its own deficiencies. They are caused by the fact that parameters of weak lines (intensity, contour shape, broadening, and shifts) as well as temperature dependences of their intensities are known less accurate.

In this paper we discuss a possibility of using the effect of stimulated Raman scattering (SRS) on molecules of the main atmospheric gas N_2 for the transfer of energy from a beam of auxiliary radiation to the main one during their collinear propagation through the atmosphere that make it possible to increase the sounding range, because the frequency of the auxiliary radiation does not fall in resonance with the absorption line.

Propagation of two collinear laser beams, one at the frequency ω_s , coinciding with the frequency of the absorption line of a gas, and another at the frequency ω_p , satisfying the condition $\omega_p - \omega_s = \omega_0$ (ω_0 is the frequency of rotational or vibrational Raman scattering by nitrogen), i.e., being off an absorption line of a gas under study, can be described by the system of equations²

$$\frac{\partial \varepsilon_s}{\partial \tau} + i \frac{\Delta_\perp}{2\kappa_s \operatorname{Re}\alpha_s} \varepsilon_s = \left(\mu |\varepsilon_p|^2 - \frac{\alpha_s}{2\operatorname{Re}\alpha_s}\right) \varepsilon_s , \qquad (1a)$$

$$\frac{\partial \varepsilon_p}{\partial \tau} + i \frac{\Delta_\perp}{2k_p \operatorname{Re}\alpha_s} \varepsilon_p = -\left(\mu \delta |\varepsilon_s|^2 + \frac{\gamma}{2}\right) \varepsilon_p , \qquad (1b)$$

where $\varepsilon_{\rm s}$, $\varepsilon_{\rm p}$ are the strengths of field of the resonance and pumping wave, respectively; $\gamma = \alpha_{\rm p}/\text{Re} \alpha_{\rm s}$; $\alpha_{\rm p}$ and $\alpha_{\rm s}$ are the complex coefficients of absorption of the resonance and pumping wave, respectively; τ is the optical thickness of a medium at the frequency $\omega_{\rm s}$, $k_{\rm s} = \omega_{\rm s}/c$, $k_{\rm p} = \omega_{\rm p}/c$

$$\delta = \frac{\omega_p}{\omega_s} \frac{|\varepsilon_s(0, 0)|^2}{|e_p(0, 0)|^2}; \quad \mu = \frac{g}{\text{Re}\alpha_s} |\varepsilon_p(0, 0)|^2;$$

and g is the gain of the resonance wave due to the SRS.

For an extended atmospheric path and $g < \text{Re } \alpha_{\rm s}$ the SRS process is weakly linear and for $I_{\rm s} \sim 10^6 - 10^8 \text{ W/cm}^2$ this corresponds to the absorption coefficients Re $\alpha_{\rm s} \sim 10^{-4} - 10^{-6} \text{ cm}^{-1}$ (see Ref. 3). In this case the estimate of the energy characteristics of the beams can be obtained neglecting nonlinear change of beam angular parameters. Then, for the coaxially symmetric beams the solution of system (1) can be represented in the form

$$\varepsilon_{s}(\tau, r_{\perp}) = \varepsilon_{sL}(\tau, r_{\perp}) \left\{ 1 + \frac{\mu}{\text{Re}\gamma} \left(1 - e^{-\text{Re}\gamma\tau} \right) + \frac{\mu^{2}}{2\text{Re}\gamma} \left(1 - e^{-\text{Re}\gamma\tau} \right)^{2} + \ldots \right\},$$
(2a)

$$\varepsilon_p(\tau, r_\perp) = \varepsilon_{pL}(\tau, r_\perp) \{1 + \mu \delta (1 - e^{-\tau}) + ...\},$$
 (2b)

where

$$\begin{split} & \varepsilon_{sL}(\tau, r_{\perp}) = \hat{H}^{-1} \left\{ \overline{\varepsilon}_{s}(0, \rho) \exp \left[-\frac{\tau}{2\text{Re}\alpha_{s}} \left(\alpha_{s} - \frac{i\rho^{2}}{\kappa_{s}} \right) \right] \right\}; \\ & \varepsilon_{pL}(\tau, r_{\perp}) = \hat{H}^{-1} \left\{ \overline{\varepsilon}_{p}(0, \rho) \exp \left[-\frac{\gamma\tau}{2} \left(1 - \frac{i\rho^{2}}{\kappa_{p}2\text{Re}\alpha_{p}} \right) \right] \right\} \end{split}$$

is a solution of the problem on linear propagation of the beams with the frequencies ω_s and ω_p being thus the initial relation for formulating a corresponding inverse problem in the method of differential absorption; \hat{H}^{-1} denotes the inverse Hankel transform.

For the energy transfer from the pumping beam (ω_p) into the sounding (ω_s) it is advisable to use the Raman– active transitions of N₂ and O₂ (the main atmospheric gases). In this case, the frequencies ω_s and ω_p ensure the feasibility of the synchronism condition at a rather long distance⁴ as well as the proper choice of frequency detuning required for the differential absorption method of sounding, $\omega_p - \omega_s = \Delta \omega \sim (5 \dots 10) \gamma_l$, where γ_l is the width of the resonance absorption line of a gas. A criterion for the increase of the sounding range is the ratio of the path length in the resonance-absorbing medium, along which attenuation of radiation reaches a certain level when propagation of two collinear beams is accompanied by energy transfer between them due to the SRS, to the length of a path without the SRS interaction.



FIG. 1. Dependence of the sounding range extension for the case of a resonantly absorbing gas on the medium parameters for $\mu = 0.2$ (a) and $\mu = 0.4$ (b).

The figure presents the results of evaluation of thusly achieved range extension for the case of sounding an absorbing atmospheric component in the mode of weak nonlinearity.

The above–described method of extending the range of sounding radiation penetration into a studied medium can be realized using tunable pulsed narrowband lasers in the visible and near–IR ranges (both solid–body and dye lasers), that are capable of delivering from 1 to 10^3 MW/cm^2 output power per pulse,⁵ and using proper selection of frequencies ω_s and ω_p and synchronization of the pulses of sounding and pumping radiation.

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