# INCREASING THE SENSITIVITY OF THE METHOD OF NARROW-BAND INTRACAVITY LASER SPECTROSCOPY BY INTRACAVITY SECOND-HARMONIC GENERATION

**S.D. Burakov, A.P. Godlevskiĭ, E.P. Gordov, N.E. Gubaidullin, A.I. Zhiliba, and S.A. Ostanin**

*Special Design Office "Optika" and Institute of Atmospheric Optics, Siberian Branch of the USSR Academy of Sciences, Tomsk*
*Received March 20, 1989*

*A scheme for narrow-band spectroscopy, in which probing of weak absorption is performed at the frequency of the laser radiation, while detection is performed at the doubled laser frequency, was studied theoretically and experimentally. It is shown that this scheme has a higher sensitivity than the traditional method.*

In the method of intracavity laser spectroscopy (ICLS)[1] weak absorption is most often detected using the following general approach: the decrease in power is determined at the frequency of the laser radiation. In this paper the efficiency of a different approach is studied: is not the absorption of laser radiation at the fundamental frequency that is recorded, but rather the effect of the absorption on the relative changes in the power of the second harmonic (SH). Coupling of the laser radiation with the radiation at the second harmonic occurs in the process of intracavity generation of the second harmonic (IGSH). The possibility of combining the ICLS method with IGSH was discussed in Refs. 2 and 3.

We shall describe the IGSH process by the following system of equations:

$$\dot{n}_1 = -(C_1+kn_1)n_1 + WNn_1, \qquad \dot{N} = v - \frac{N}{T_1} - Wn_1N, \tag{1}$$

where $n_1$ is the photon number density at the frequency of the laser radiation ω; $N$ is the population inversion; $C_1$ is the width of the cavity band with no losses to second-harmonic generation; $k = g^2/C_2$, where $g$ characterizes the nonlinear coupling of the laser radiation and the second-harmonic radiation and $C_2$ is the width of the cavity band at 2ω; the quantity $W$ is proportional to the gain cross section of the active medium; $v$ is the pumping rate; and, $T_1$ is the relaxation time of the population inversion.

In deriving the system (1) we employed equations describing the dynamics of a laser such as $YAG:Nd^{3+}$, $CO_2$ (Ref. 4) under the assumption that the atoms of the active medium do not interact with the second-harmonic field. The IGSH process was described using the equations derived in Ref. 5. The equation for the second harmonic is excluded adiabatically ($C_2 \gg C_1, T_1^{-1}$). In the case when the population inversion "follows" the field ($T_1^{-1} \gg C_1$) the dynamics of the laser in the system of the intracavity laser spectrometer combined with IGSH is described by Lamb's model.[3] It follows from the system (1) that the average power $P_1$ of the radiation generated by the active medium at the frequency ω and converted into the second harmonic with power $P_2$ has the form:

$$P_1 = (C_1+kn_1)n_1; \tag{2}$$

$$P_2 = kn_1^2. \tag{3}$$

We shall evaluate the sensitivity with which $P_{1,2}$ are recorded by studying the relation $\delta P_{1,2}/P_{1,2}$, where $\delta P_{1,2}(C_1 + \delta C_1) - P_{1,2}(C_1)$:

$$\delta P_1/P_1 = \delta n_1/n_1 \tag{4}$$

$$\frac{\delta P_2}{P_2} = 2\,\frac{1-\alpha}{1+2\alpha}\cdot\frac{\delta P_1}{P_1}; \tag{5}$$

where $\alpha \equiv kn_1/C_1$ characterizes the relative fraction of the nonlinear load of the cavity with respect to the linear load. In the case $\alpha < 1$

$$\delta P_2/P_2 = 2\,(1-\alpha)(\delta P_1/P_1). \tag{6}$$

We note that the sensitivity with which the absorption at в is determined does not drop below the sensitivity of the variant of ICLS without IGSH.[3] Thus under these conditions of IGSH the sensitivity with which small changes are detected in the signal at the frequency ω by recording $\delta P_2/P_2$ is almost two times higher than that of the traditional method. The case of IGSH when $\alpha > 1$ is unfavorable from the viewpoint of increasing the sensitivity.

In the laboratory experiment we studied the dependence of the change in the power at the wavelength 0.53 μm accompanying a change in the losses in the cavity of a laser lasing at the wavelength 1.0641 μm. Figure 1 shows a block diagram of the experimental arrangement. The laser cavity is formed by three mirrors with multilayer dielectric coatings based on zirconium oxides: a nontransmitting flat mirror *7* with transmission coefficient not greater than 0.024%, a parametric mirror *3* with a transmission coefficient of 92.4% at the wavelength 0.532 μm and 0.2% at the wavelength 1.0641 μm (the radius of curvature is equal to 251 mm), and a flat mirror *5* with a reflection coefficient of 99.2% at the wavelength 0.532 μm and 99.84% at the wavelength 1.0641 μm. The radiation at the fundamental wavelength (1.0641 μm) was generated by the active medium *6*, which consisted of a YAG:$Nd^{3+}$ crystal 5 mm in diameter and 100 mm long. The laser lased in the continuous-wave regime. Conversion into the second harmonic was performed with a frequency converter *4* (MCh-109), consisting of a lithium iodate nonlinear crystal.

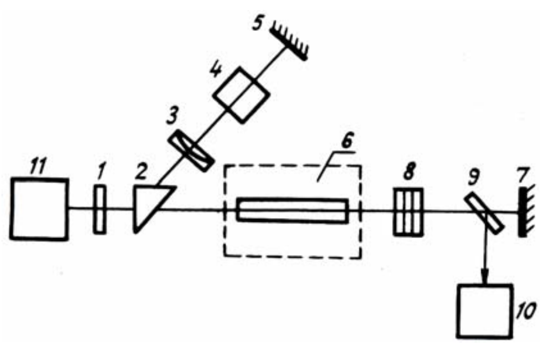


*FIG. 1. Diagram of experimental arrangement: 1 – output window; 2 – prism; 3, 5, 7 – cavity mirrors; 4 – MCh-109 frequency converter; 6 – K-301B active medium; 8 – attenuators; 9 – glass plate for extracting radiation; 10, 11 – LM2 power meters.*

The 1.0641 μm radiation was extracted from the cavity with a half-transmitting plate *9* and recorded with an M2 power meter (manufactured by the Karl Zeiss Jena Company). The 0.53 μm radiation was deflected by the prism *2* through the output window *1* into an analogous power meter *11*.

To change the losses at 1.0641 μm radiation attenuators *8*, consisting of thin glass plates, were successively inserted into the cavity.

Figure 2 shows the experimental data on the dependence of the laser power at the second harmonic ($\lambda$ = 0.53 μm) on the change, produced by inserting attenuators into the cavity, in the lasing power, at the wavelength 1.0641 μm. The relative change in power $\delta P(\delta C_1)/P$, where $P$ is the power at the corresponding wavelength with no attenuators in the cavity and $\delta C_1$ is the optical thickness of the attenuator, was plotted along the axes.

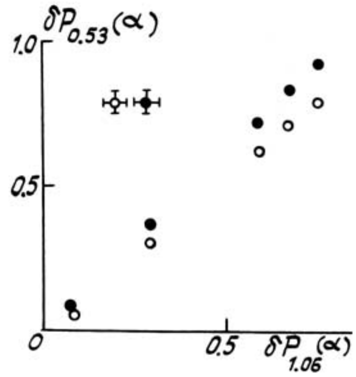


*FIG. 2. The relative change in the lasing power of a YAG:$Nd^{3+}$ laser at $\lambda$ = 0.53 μm versus the relative change in the lasing power at $\lambda$ = 1.0641 μm: • – t = 10°C; ○ – t = 30°C.*

One can see from the graph that any change in lasing power at $\lambda$ = 1.0641 μm corresponds to a greater change in the lasing power at $\lambda$ = 0.53 μm; this can be interpreted as an increase in sensitivity. The increase in sensitivity can be expressed quantitatively by the relation

$$\Delta \equiv (\sigma P_{0.53}/P_{0.53})/(\sigma P_{1.06}/P_{1.06}).$$

In the experiments the value $\Delta$ = 1.2 was recorded with the temperature of the YAG:$Nd^{3+}$ crystal equal to $t_{cr}$ = 10°C and $\Delta$ = 1.1 with $t_{cr}$ = 30°C.

Thus it has been shown that the signal at the second harmonic can in principle be recorded efficiently. In the case of relative measurements of variations of the laser radiation, for example, owing to selective absorption, detection of the signal at the doubled frequency gives an additional (relative to the traditional method of detection) gain in sensitivity. When absolute measurements of the attenuation are performed at the frequency of the laser radiation it is important that the absolute sensitivity of photodetection devices in the shorter-wavelength region of the spectrum (to 1 μm) is at least an order of magnitude higher than in the far- and middle-IR region.

## REFERENCES

1. L.A. Pakhomicheva, E.A. Sviridenkov, A.F. Suchkov, L.V. Titova, and S.S. Churilov, Pis'ma Zh. Eksp. Teor. Fiz. **12**, 60 (1970).

2. S.F. Luk'yanenko and M.M. Makagon in: *The Sixth All-Union Symposium on High- and Ultrahigh-Resolution Spectroscopy*, Tomsk Affiliate of the Siberian Branch of the USSR Academy of Sciences, 208, Tomsk (1982).

3. E.P. Gordov, A.I. Zhiliba, and M.M. Makagon in: *Proceedings of the Eighth All-Union Symposium on High-Resolution Spectroscopy*, Tomsk Affiliate of the Siberian Branch of the USSR Academy of Sciences, Part 1, 50, Tomsk (1988).

4. H. Haken, Laser Light Dynamics, North-Holland Elsevier, No. 4 (1985), [Russian translation] (Mir, Moscow, (1988).

5. L. Mandel, Xiago-Guang, J. Opt. Soc. Amer., **3B**, 940 (1986).