INTRACAVITY LASER SPECTROSCOPY WITH NONLINEAR FREQUENCY CONVERTER AND AN EXTERNAL SIGNAL

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Received June 19, 1989

It is shown that combining the method of intracavity laser spectroscopy (ICLS) with second harmonic generation (SHG) in the laser cavity and an external signal is more advantageous than using the spectrophotometric method and extracavity SHG.

The spectral range of the method of intracavity laser spectroscopy (ICLS), which is one of the most sensitive methods for analyzing absorption spectra, is fundamentally limited by the range of available laser frequencies. This makes it impossible to use this method to detect gaseous pollutants in the atmosphere which have strong electronic absorption bands in the UV region, such as H_2S, SO_2, benzene, phenol, aromatic hydrocarbons, etc. Nonlinear frequency converters and the spectrophotometric method of measurement are widely employed for studying absorption spectra in the ultraviolet; in this case, the medium under study is located outside the laser cavity. In Ref. 1 it was proposed that a frequency converter and the medium under study be placed in the cavity of the same wide-band laser. Then the power of the radiation at the frequency at the fundamental radiation is dependent on the losses at the frequency of the converted radiation. In this paper, which is a continuation of Ref. 2, we analyze the sensitivity of such a spectrometer for the case of second-harmonic generation and single-mode generation regime of the laser (narrow-band ICLS).

We shall model the system (active medium, nonlinear crystal, and the gaseous medium under study which absorbs the radiation in the common cavity) by the following system of equations;

$$\begin{aligned}
\dot{a}_1 &= \gamma_1 a_1 - ikP_1 + g a_1^* a_2;\\
\dot{a}_2 &= -\gamma_2 a_2 - g a_1^2/2 + E_2;\\
\dot{P}_1 &= -\gamma_\perp P_1 + D a_1;\\
\dot{D} &= \gamma_\parallel (D_0 - D) - k(P_1 a_1^* + P_1^* a_1).
\end{aligned} \tag{1}$$

where D is the total population inversion with relaxation rate $\gamma_\parallel$, a_1 and a_2 are the dimensionless amplitudes of the fields at the fundamental and doubled frequencies, P_1 is the polarization of the laser active medium with relaxation rate $\gamma_\perp$; k takes into account the coupling between active centers with the field a_1; g is the coupling constant between the fields a_1 and a_2, and is proportional to the susceptibility of the nonlinear crystal; γ_1 and γ_2 are the inverse relaxation times of the fields a_1 and a_2, and include the losses in the elements of the spectrometer and possible absorption of the gas under study in the cell; and, E_2 is the field of the external laser source at the doubled frequency. For simplicity and to discuss the fundamental possibilities of the method we shall assume that all fields (a_1, a_2, and E_2) are monochromatic and we shall study the laser equations in the adiabatic, stationary case (this corresponds to the "narrow-band" variant of ICLS with a continuously operating laser).

We shall evaluate the sensitivity of the method and compare it with the photometric method. Solving Eq. (1) in the indicated approximations, we obtain

$$\frac{\Delta I_1}{I_1} = \frac{G_0 - \dfrac{gE_2}{2\gamma_2}}{G_0 - \gamma_1 - \dfrac{gE_2}{2\gamma_2}} \frac{\Delta\gamma_1}{\gamma_1}; \tag{2}$$

$$\frac{\Delta I_2}{I_2} = \left(\frac{\dfrac{gE_2}{2\gamma_2}}{G_0 - \gamma_1 - \dfrac{gE_2}{2\gamma_2}} + \frac{2}{1 + \dfrac{2\rho\gamma_2}{g^2}} \right) \frac{\Delta\gamma_2}{\gamma_2}, \tag{3}$$

where G_0 and β appear in the expression for $P = \dfrac{G_0 a_1}{1 + \beta |a_1|^2}$ and describe the linear and cubic part of the polarization; I is the intensity of the radiation; ΔI is the change in the intensity owing to the gaseous sample under study; $\kappa_{1,2}$ is the absorption coefficient of the sample of length z; $\Delta\gamma_{1,2} = \kappa_{1,2} \cdot z$ — are the losses introduced by the sample at the frequencies 1 and 2. The sensitivity of the spectrophotometric method of detection is

0235-6880/90/03 298–037 $02.00

$$\left(\frac{\Delta I}{I}\right)_{ph} = \Delta\gamma. \tag{4}$$

The expression (2) with $E_2 = 0$ or $g = 0$ transforms into the well-known expression[3] for narrow-band ICLS

$$\frac{\Delta I_1}{I_1} = \frac{G_0}{\gamma_1}\frac{1}{G_0 - \gamma_1}\Delta\gamma_1. \tag{5}$$

Compared with Eq. (4) the sensitivity is higher for two reasons: first, owing to the multiple passage of radiation in the cavity and, second (the resonance factor), owing to the fact that the system operates near threshold. We note that the introduction of a nonlinear crystal ($g \neq 0$) and the external signal ($E_2 \neq 0$) do not change Eq. (2) qualitatively; this means that the spectrometer at the fundamental frequency operates normally in these regimes also ($g \neq 0$, $E_2 \neq 0$).

The sensitivity of the spectrometer to losses at the second-harmonic frequency depends qualitatively on g and E_2. For $g = 0$ $\Delta I_2/I_2 = 0$, and this is natural, since there is no radiation at this frequency. For $g \neq 0$ and $E_2 = 0$ only the second term in Eq. (3) "works". The spectrometer "feels" the losses at the harmonic frequency; the sensitivity increases as γ_2 decreases (the multiple pass nature of the radiation in the resonator is manifested here). For $g \neq 0$ and $E_2 \neq 0$ the first (resonance) term in Eq. (3) starts to have an effect and the sensitivity of the spectrometer (for corresponding choice of g and E_2) can increase substantially.

The following feature is common to the situation with an external signal E_2 when determining both $\Delta\gamma_1$ and $\Delta\gamma_2$ it is possible to decrease the value of the resonance denominator in Eqs. (2) and (3) by using a weak well-stabilized external field and thereby to increase the sensitivity of the method of intracavity spectroscopy. An additional factor increasing the sensitivity is the suppression of the amplitude fluctuations of the fundamental radiation and the second harmonic radiation, which happens in the process of SHG.[4]

The spectral resolution at the doubled frequency can decrease somewhat. In the case of a single-mode continuous laser radiation the radiation line is broadened primarily owing to phase diffusion[5] and in this situation, owing to the coupling of the phases of the waves of the fundamental radiation and the second harmonic radiation, the following relation is obtained[6]:

$$\Delta\nu_2 = 4\Delta\nu_1 \tag{6}$$

($\Delta\nu_{1,2}$ is the width of the radiation spectrum of the first and second harmonics). However this is not a serious limitation of the method described here, since the width of the spectrum is an order of magnitude smaller than the width of the Doppler or Lorentz broadened absorption lines of the gases.

Thus in studying absorption at the doubled frequency of laser radiation, combining ICLS with intracavity harmonic generation gives higher sensitivity than does the use of the spectrophotometric method and extracavity harmonic generation.

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