

LASER SPECTROSCOPY

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Optimisation of external cavity parameters of a weak absorption laser spectrometer

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Abstract. **We consider some peculiar features of the optimisation procedure of external optical cavity parameters of a laser spectrometer, caused by the use of a three-beam measurement scheme and the presence of losses in the mirrors. It is found that the maximum sensitivity to the absorption of an intracavity medium can be achieved only at a certain choice of the value and the ratio of the reflection coefficients of the mirrors. For example, registration of the spectra of the methane impurity in the atmosphere shows that in accordance with the calculation model, for the same resonator *Q*-factor the use of an input mirror with a smaller reflection coefficient allows the measurement sensitivity to be increased by approximately two times.**

Keywords: *diode lasers, external optical resonator, absorption spectroscopy, low concentrations.*

1. Introduction

To measure low concentrations of gas impurities, use is widely made of absorption spectroscopy based on the registration of radiation absorption in an external optical cavity containing a gas in question [1]. By measuring the photon lifetime in the resonator (cavity ring-down spectroscopy – CRDS [2]), a methane concentration sensitivity of 29 ppt is reached, which is equivalent to an absorption coefficient 4.4×10^{-12} cm^{-1} at an optimum averaging time of 32.4 min [3]. There exists a so-called integral modification of this method (integrated cavity output spectroscopy – ICOS [4]), which allows one to replace the temporary measurements by the amplitude ones. The ICOS technique, unlike the CRDS method, where the spectrum is recorded by the points at fixed frequencies, allows one to measure spectra with continuous tuning of the laser radiation frequency. In phase shift CRDS (PS-CRDS), there are ways of recording the spectrum with continuous frequency tuning, but they require rather complex mathematical signal processing [5].

The accuracy, sensitivity and reliability of measurements largely depend on the right choice of the reflection coefficients of external cavity mirrors. There are currently developed technologies of fabrication of mirrors with a reflectance $r = 0.9999$ or higher in different spectral ranges, which provides a high sensitivity of the above-discussed measurements. However, during the exploitation such reflective coatings have a tendency to degrade, the degradation being significant.

It is usually assumed that the sensitivity of the absorption measurements is preserved while maintaining the empty-cavity *Q*-factor, which depends on the product of the reflection coefficients of the mirrors ($r_1 r_2$ = const) and determines the effective number of roundtrips for radiation in the cavity. This is true in the case of a laser resonator, where dissipative losses on the mirrors are compensated for by the active medium gain. For an externally excited passive resonator the situation is different and analysed in this paper. This analysis aims at the development and experimental verification of the method of optimisation of the resonator parameters, which allows one to ensure an acceptable value of the measured signals and the necessary absorption measurement accuracy.

2. Transmission and reflection of radiation by a cavity

The optimisation procedure of the reflection coefficients of resonator mirrors can be based on well-known formulas describing the reflection and transmission of radiation by a cavity [6]:

$$r_c = \frac{\sqrt{r_1}[1 - (\sqrt{r_1 r_2} + \sqrt{r_1/r_2}\, t_1)\, t_m \exp(i2\delta)]}{1 - \sqrt{r_1 r_2}\, t_m \exp(i2\delta)}, \quad (1)$$

$$t_c = \frac{\sqrt{t_1 t_2}\sqrt{t_m}\exp(i\delta)}{1 - \sqrt{r_1 r_2}\, t_m \exp(i2\delta)}. \quad (2)$$

Here r_c and t_c are, respectively, the amplitude reflection and transmission coefficients of the cavity (power coefficients $R = |r_c|^2$ and $T = |t_c|^2$ corresponding to them); t_1, t_2 are the transmission coefficients of the mirrors with respect to power; r_1, r_2 are the reflection coefficients of the mirrors (with respect to power); t_m is the transmission coefficient of an intracavity medium (with respect to power); and δ is the phase shift per single pass of a wave in the resonator. The transmission and reflection coefficients of the mirrors are related as $1 - r_{1,2} - t_{1,2} = a_{1,2}$, where $a_{1,2}$ are the mirror losses, which will be assumed the same below: $a_1 = a_2 = a$. The coefficients r_c and t_c, as

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follows from (1) and (2), are the functions of the phase shifts δ (in the experiment the phase shifts change by varying the frequency of the incident light). From (1) and (2) we obtain the expression for R and T:

$$R = \left| \frac{-\sqrt{r_1/r_2}\{1-[\sqrt{r_1 r_2} + (\sqrt{r_2/r_1} - \sqrt{r_1 r_2} - a\sqrt{r_2/r_1})]\,t_m \exp(i2\delta)\}^2}{[1-\sqrt{r_1 r_2}\,t_m \exp(i2\delta)]^2/\sqrt{r_1 r_2}} \right|, \quad (3)$$

$$T = |[(1/\sqrt{r_1 r_2} - \sqrt{r_1/r_2} - a/\sqrt{r_1 r_2})(1/\sqrt{r_1 r_2} - \sqrt{r_2/r_1} - a/\sqrt{r_1 r_2})$$

$$\times r_1 r_2 t_{\rm m} \exp({\rm i}2\delta)]^{1/2}/[1-\sqrt{r_1 r_2}\,t_{\rm m}\exp({\rm i}2\delta)]|^2. \quad (4)$$

They show that these coefficients at specified losses a depend on both the product $r_1 r_2$, which directly determines the cavity Q-factor, and on the ratio r_1/r_2, which can be varied for a fixed Q-factor of the resonator. We use these relations to analyse the scheme of a specific spectrometer.

3. Analysis of a three-beam R-ICOS scheme

Consider the problem of the resonator optimisation for the case when use is made of one of the latest versions of the ICOS method, i.e. so-called R-ICOS [7]. The schematic of the method is shown in Fig. 1 (a more detailed experimental scheme and its operation are described in [7]). In this method, apart from the intensity I_1 of light passed through the cavity (as in the ICOS method), we also record the intensity I_3, which is proportional to the intensity of light reflected from the resonator, and the intensity I_2, which is proportional to the laser light intensity I_0 (baseline). This allows one to determine the fraction of the power absorbed in the cavity, U, as a differential signal:

$$U = 1 - R - T. \quad (5)$$

By changing the value of U during the laser frequency scanning we can determine the absorption spectrum of the medium in question. In this case, it is possible to reduce the fluctuations of the signal detected at the resonator output (standard ICOS method), which are caused by phase mismatches of radiation and resonator eigenmodes due to the instabilities in the reflected signal. This makes it possible to reduce the time of registration and extend the dynamic range of the intensities of the measured spectra [8]. At reflection coefficients of the cavity mirrors, 0.9–0.99, the R-ICOS method makes it possible to measure the absorption coefficients with a sensitivity of 2×10^{-8} cm^{-1} at a spectrum recording speed of $\sim10^6$ cm^{-1} s^{-1}.

In paper [7], where the R-ICOS method was described, we used a symmetrical external resonator with the same reflection

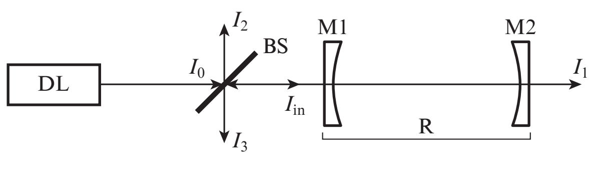

Figure 1. Schematic of the R-ICOS method:
(DL) diode laser; (BS) beam splitter; (R) resonator with mirrors M1 and M2; $I_{\rm in}$ and I_1 are the intensities of light incident on the resonator and transmitted through it; I_2 and I_3 are the intensities of light reflected by the beam splitter, proportional to the intensities of the laser light I_0 and light reflected by the resonator.

coefficients of the mirrors. At the same time, the possibility of improving the spectrometer performance by using an asymmetric resonator with different reflection coefficients of the mirrors has not been investigated. However, such a configuration is expedient from the standpoint of the fulfilment of two requirements. The first one is to ensure comparable intensities of light passed through the resonator and reflected from it at a sufficient level of their powers (in this case, the mutual compensation of phase fluctuations of transmission and reflection coefficients of the resonator is best implemented). The second requirement is due to the necessity of obtaining a maximum value of the derivative of the difference signal for intracavity losses (this makes it possible to record changes in the resonator losses with the highest accuracy). The simultaneous implementation of these requirements consists in the search for a compromise in the choice of the resonator parameters to achieve acceptable values of recorded signals and measurement sensitivity. In this respect, the manipulation of different reflection coefficients of the mirrors ensures a greater search flexibility.

The choice of reflection coefficients requires a multi-parametric analysis of the behaviour of the difference signal magnitude on the basis of relations (3) and (4). Such an analysis can be reduced to the construction of the functional F, the maximum values of which for a given level of dissipative losses a will determine the optimum range of variation in the coefficients r_1 and r_2. Fulfilling the requirements mentioned above can be achieved by representing the functional in the form of a product:

$$F(r_1, r_2) = R(r_1, r_2)T(r_1, r_2)U_0(r_1, r_2)B_1(r_1, r_2)B_2(r_1, r_2), \quad (6)$$

where U_0 is the same as U in (5), but in the absence of a medium in the cavity, and the auxiliary functions $B_1(r_1, r_2)$ and $B_2(r_1, r_2)$ are determined by the expressions

$$B_1(r_1, r_2) = \exp\{-[R(r_1, r_2) - T(r_1, r_2)]^2/[R(r_1, r_2)T(r_1, r_2)]\}, \quad (7)$$

$$B_2(r_1, r_2) = {\rm d}U/{\rm d}t_{\rm m}. \quad (8)$$

The first function describes the balance of power of transmitted and reflected light and assumes a maximum value of unity at $R(r_1, r_2) = T(r_1, r_2)$. The second function determines the sensitivity of the optical scheme to small changes (in the spectrum) in the transmission of an intracavity medium. The functions included in expression (6) are calculated under resonance conditions at $\delta = \pi n$ (n is an integer). The extrema of the functional generally differ in magnitude and position on the $r_1 r_2$ plane. Therefore, the search for a maximum value of the functional F allows us to give an overall assessment of the impact of different (often conflicting) factors on the resonator characteristics. Figure 2 shows the dependence of the functional values on the reflection coefficients of the mirrors for a = 0.005. The maximum of the functional $F_{\rm max}$ corresponds to r_1 = 0.945, r_2 = 0.988 and is equal to 2.126. In this case, the difference signal U_0 = 0.4, and the derivative dU/d$t_{\rm m}$ = 58.53. In the measurements the difference signal, which represents a system of equidistant peaks, will be modulated by the contours of the detected absorption lines.

A visual representation of the behaviour of both the functional and the functions forming this functional is given in Fig. 3, which shows their dependence on r_2 at r_1 = 0.935. One can see that the maximum values lie in a region where

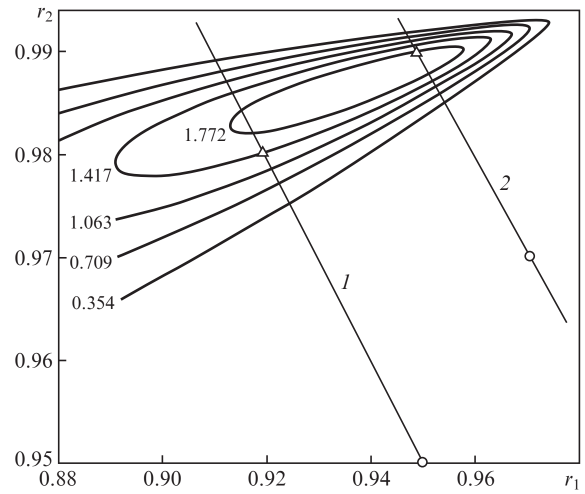

Figure 2. Dependence of the values of the functional F on the reflection coefficients r_1 and r_2 at $a = 0.005$. Inclined lines correspond to expressions $r_1 r_2$ = (*1*) 0.903 and (*2*) 0.941. Symbols ○ and △ denote, respectively, symmetric and asymmetric resonators used in the experiments.

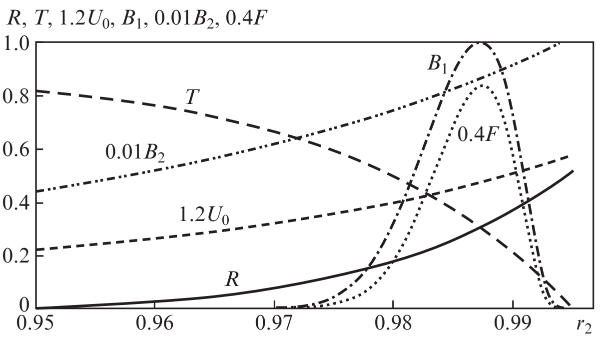

Figure 3. Dependence of the functional F and its functions on r_2 at $r_1 = 0.935$ and $a = 0.005$.

the curves $R(r_2)$ and $T(r_2)$ intersect and the values of U_0 and $\mathrm{d}U/\mathrm{d}t_\mathrm{m}$ are sufficiently large.

The position of the extremum of the functional depends essentially on the losses a. A reduction of a, leading to an increase in the resonator Q-factor, causes a shift of the maximum of the functional to the region of large values of r_1 and r_2. For example, at $a = 0.001$ the maximum of the functional $F_\mathrm{max} = 10.463$ is achieved for $r_1 = 0.985$ and $r_2 = 0.994$ (Fig. 4). In this case, $U_0 = 0.47$, and $\mathrm{d}U/\mathrm{d}t_\mathrm{m} = 316$. It can be seen that

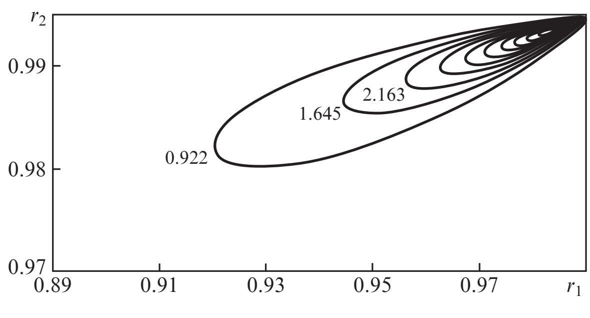

Figure 4. Dependence of the values of the functional F on the reflection coefficients r_1 and r_2 at $a = 0.001$.

the use of a higher Q-factor cavity, only slightly increasing the difference signal, greatly increases the derivative $\mathrm{d}U/\mathrm{d}t_\mathrm{m}$, which means an increase in the measurement sensitivity.

The calculation results show that the highest sensitivity and accuracy are achieved using an asymmetrical cavity (with $r_1 < r_2$). Indeed, if in the case under study, when $a = 0.005$, use is made of a symmetric resonator with reflection coefficients $r_1 = r_2 = 0.961$, without changing its Q-factor (it is determined by the product $r_1 r_2$), the differential signal and its derivative decrease: $U_0 = 0.24$ and $\mathrm{d}U/\mathrm{d}t_\mathrm{m} = 35$. At a higher Q-factor when $a = 0.001$, use of a symmetric resonator with $r_1 = r_2 = 0.989$ yields $U_0 = 0.24$ and $\mathrm{d}U/\mathrm{d}t_\mathrm{m} = 167$, which is approximately two times smaller than the corresponding parameters of an optimised asymmetric resonator.

4. Experiment

To check the results of cavity optimisation based on the above-considered procedure, we used a spectrometer described in detail in [7]. Its external resonator had a length $L = 50$ cm and was formed by two mirrors with a diameter of 25 mm and a radius of curvature of 1 m. The resonator was pumped by radiation of a vertical cavity surface-emitting diode laser [VCSEL, Vertilas, the power of 1.5 mW in the wavelength range near 1650 nm (6060 cm^{-1})]. The spectral width of the laser line was 10 MHz. The tuning range of the laser radiation frequency was equal to 1.15 cm^{-1}. When testing the optical scheme we recorded a portion of the spectrum of methane near the wavelength $\lambda = 1650$ nm at its content in the air 2.5×10^{-5}. We used a set of mirrors with reflection coefficients 0.92–0.99 at this wavelength. In the experiments we used both symmetric and asymmetric resonators. To this end, we compared the properties of the resonators with the same Q-factor. In all cases, as predicted by the above-described optimisation procedure, the best quality of the spectra is achieved when the input mirror had the smallest reflection coefficient.

Experiments were performed using two sets of mirrors. In the first set the mirrors had reflectivities $r_1 = r_2 = 0.95$ and $r_1 = 0.92$, $r_2 = 0.98$ for the symmetric and asymmetric resonators, respectively; and in the second set the mirrors had reflectivities $r_1 = r_2 = 0.97$ (symmetric resonator) and $r_1 = 0.95$, $r_2 = 0.99$ (asymmetric resonator). It was assumed that for this class of mirrors $a = 0.005$. In Fig. 2 the values of the reflection coefficients corresponding to the Q-factors of the resonators lie on additional inclined lines. For ease of comparison of the characteristics of the resonators, points corresponding to the reflection coefficients of the mirrors are shown on these lines by different symbols. Their position confirms the advantage of the asymmetric configuration. Calculations in the region of the symbols indicate that the symmetrical resonator with the first set of mirrors is characterised by the values $U_0 = 0.18$ and $\mathrm{d}U/\mathrm{d}t_\mathrm{m} = 28$, and the asymmetric resonator – by values $U_0 = 0.3$ and $\mathrm{d}U/\mathrm{d}t_\mathrm{m} = 46.7$. In using the second set of mirrors, for the symmetric resonator we have $U_0 = 0.29$ and $\mathrm{d}U/\mathrm{d}t_\mathrm{m} = 36.7$, and for the asymmetric resonator $U_0 = 0.52$ and $\mathrm{d}U/\mathrm{d}t_\mathrm{m} = 67.3$. The magnitude of the experimentally measured signal U for the asymmetric resonator is approximately 1.4 times greater than for the symmetric one with the first set of mirrors; in the case of the second set of mirrors, the corresponding value is about two times larger. These data are close to those calculated numerically.

The advantage of an asymmetric resonator, supported by the above estimates, is confirmed by experimental data, which is illustrated in Fig. 5, showing a portion of the methane absorption spectrum near the line λ = 1650 nm. The spectrum is recorded using an asymmetric (r_1 = 0.95 and r_2 = 0.99) and symmetric ($r_1 = r_2$ = 0.97) resonators. It can be seen that the use of an asymmetric resonator allows one to increase manifold the signal/noise ratio and, consequently, to improve the sensitivity of the spectrometer to weak absorption. A weakly pronounced interference pattern present on the line contour (Fig. 5a) is caused by the above-mentioned spike character of the difference signal, its contrast being dependent on the speed of the recording equipment.

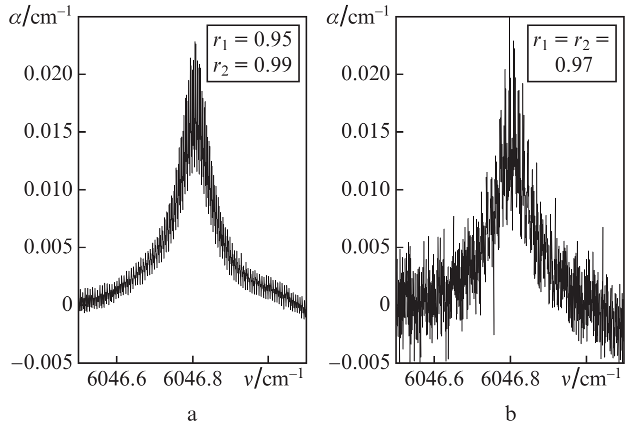

Figure 5. Absorption spectra of methane at λ = 1650 nm, measured using (a) asymmetric and (b) symmetric resonators.

5. Conclusions

The results of the calculation and experiments show that given the same *Q*-factor of external optical resonators, one can increase the sensitivity of the absorption laser measurements by using mirrors with different reflection coefficients. The physical reason behind this fact is a significant reduction in the radiation power absorbed by the output mirror at a higher reflectance. This fact may be useful for optimisation of the external resonator parameters of a weak absorption spectrometer during high-precision spectral measurements using the R-ICOS method of diode laser spectroscopy.

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