# Dynamics of laser intracavity absorption

H. Atmanspacher, H. Scheingraber, and C. R. Vidal
*Institut für Extraterrestrische Physik, Max-Planck-Institut für Physik und Astrophysik, D-8046 Garching bei München, Federal Republic of Germany*
(Received 21 March 1985)

The dynamical behavior of a multimode dye laser with an intracavity absorber has been investigated theoretically as well as experimentally. This behavior is described by a system of rate equations which has to include the influence of saturation in the case of large absorption coefficients. With the rate-equation-system model calculations have been performed for the cases of saturated and unsaturated intracavity absorption. Saturation of the absorption is shown to severely reduce the sensitivity of intracavity absorption. In general, the sensitivity is governed by the average lifetime of the individual modes. This mode lifetime is limited by large intensity fluctuations which are capable of quenching individual modes. The probability for mode quenching is shown to increase with increasing spectral power density. For this reason, intracavity absorption gains sensitivity close to the lasing threshold. The theoretical results are found to be in excellent agreement with our measurements. In addition, discrepancies in previous work have been resolved within the framework of our theory. With the detailed understanding of the intracavity absorption, reliable measurements of very small, absolute absorption coefficients have become possible.

## I. INTRODUCTION

Intracavity absorption is known as a spectroscopic method providing an extraordinarily high sensitivity. The smallest absorption coefficients detected so far[1] are on the order of some $10^{-10}$ cm$^{-1}$. Under normal conditions (760 Torr, 300 K) this sensitivity corresponds to a minimum detectable absorption oscillator strength of approximately $10^{-18}$ in two-level systems.

In 1970 Pakhomycheva *et al.*[2] observed absorption features in the emission spectrum of a neodymium-glass laser which could be explained by weak intracavity etalon effects. With the same technique Pakhomycheva *et al.* succeeded to detect weak molecular absorption lines of $NH_3$ and $CH_4$. Shortly thereafter Peterson *et al.*[3] demonstrated that the intracavity absorption technique is much more sensitive to small concentrations of Na vapor and $I_2$ vapor than any external absorption measurement.

Using pulsed dye laser systems a sensitivity enhancement of a factor 1000 was obtained compared with external absorption.[4,5] Hänsch *et al.*,[6] who used a cw dye laser, achieved an enhancement factor as high as $10^5$.

During the years following this pioneering work, the power of intracavity absorption spectroscopy has been demonstrated in many different applications. We refer especially to the Soviet work in this field reviewed by Burakov.[7]

One of the most obvious applications is the analysis of molecular spectra with very small line strengths. The first experiments were applied to the ubiquitous atmospheric water vapor. Intracavity observations of $H_2O$ were reported in 1975 by Antonov *et al.*[8] and Baev *et al.*,[9] who detected transitions to higher vibrational levels of the electronic ground state. In 1977, Bray *et al.*[10] observed the magnetic dipole transitions of the (2-0) band of the $O_2$ ($b$-$X$) system by intracavity absorption. Hill *et al.*[11] succeeded to detect the weaker (3-0) band. Atkinson *et al.*[12] reported intracavity absorption measurements on $NO_2$ in the blue and red spectral region. Antonov *et al.*[13] presented spectra of $NH_3$ and $CH_4$ around 605 and 645 nm, respectively. In 1980 Smith and Gelfand[14] detected a few lines of the (5-0) band of the electronic ground state of HD by means of a photoacoustic detection technique. Furthermore, vibrational overtones of CH and OH groups in large molecules have been investigated.[15,16]

Intracavity absorption was also successfully used for the detection of extremely small particle densities. Maeda *et al.*[17] detected $10^5$ Na atoms per cm$^3$. Comparable sensitivities have been reported by T. D. Harris and Mitchell[18] (Fe) and Burakov *et al.*[19] (Al, Ca).

Spectroscopy on highly excited states in plasmas by means of intracavity absorption has been demonstrated by Burakov *et al.*[20] using hollow-cathode discharges containing Mo, Ni, and Ca. Brink and Heider[21] reported observations using gas discharges of helium, neon, and hydrogen.

Hänsch *et al.*[6] demonstrated an elegant method to study rare isotopes of the $I_2$ molecule. The proposed method was also applied by Datta *et al.*,[22] and Hohimer and Hargis.[23]

The high sensitivity of intracavity absorption also enables the observation of short-lived intermediate products in chemical reactions. Therefore problems related to reaction kinetics can be investigated. Work in this field was carried out in particular on the HCO radical.[24–27] The application of intracavity absorption to combustion diagnostics was first described by Thrash *et al.*[28] and has been summarized by S. J. Harris in 1984.[29] The corresponding Soviet work on reaction kinetics has been reviewed in 1981 by Sarkisov and Sviridenko.[30]

From an experimental point of view, two basically different detection techniques have been used so far:

 

(i) In the first technique, the absorption feature can be detected *directly* as an absorption dip in the laser emission spectrum. The spectral resolution is achieved by tuning the detection system (spectrometer) or by tuning the laser wavelength. Furthermore, one can tune the absorption line across the laser emission spectrum by means of an external magnetic or electric field. In this manner intracavity "laser magnetic resonance"[31] and intracavity Stark spectroscopy[32] have been carried out.

(ii) In a second technique, intracavity absorption can be observed in an *indirect* manner. In this case intracavity absorption processes can be detected, for instance, in an external probe cell by measuring changes of the fluorescence signal from the external probe.[6] In a similar manner, optogalvanic[33] and photoacoustic[34] detection in an external cell have been demonstrated.

Besides the extreme enhancement of the sensitivity of absorption, also other spectroscopic methods gain greatly in sensitivity, if the probe is placed inside a laser cavity. The following techniques have already been applied:

(i) Photoacoustic spectroscopy (Stella *et al.*[35]).

(ii) Polarization spectroscopy (Radloff and Ritze,[36,37] Mak *et al.*[38]).

(iii) Double- and triple-resonance spectroscopy in the infrared (ir), microwave (mw), and radio frequency (rf) spectral regions (ir-rf: Dangoisse *et al.*;[39,40] ir-mw: Arimondo and Oka,[41] Oka;[42] ir-mw-rf: Ioli *et al.*[43]).

(iv) Linear and nonlinear Raman effect (Werncke *et al.*,[44] Baev *et al.*,[45] Silvera and Tommasini[46] and others).

(v) Two- and three-photon absorption (Chen *et al.*,[47,48] Kowalski *et al.*[49]).

Methods (iv) and (v) take additional advantage of the higher intensity of the intracavity radiation field.

In spite of the large number of different applications, the theory of intracavity absorption and the different mechanisms which determine its extremely high sensitivity, are still not yet understood. The different models which have so far been proposed, can be classified into two groups:

(i) In the first group it is basically assumed that the laser system reaches a steady state. In this steady-state limit all quantities determining the sensitivity of intracavity absorption are regarded as being constant in time. Models of this kind have been developed by Hänsch *et al.*,[6] Brunner and Paul,[50,51] and Stepanov *et al.*[52] They predict a crucial influence of spatial inhomogeneities in the gain medium on the sensitivity of intracavity absorption. However, various experimental results have shown that the steady-state models are inadequate. For a summary discussing the different discrepancies see S. J. Harris[53] and S. J. Harris and Weiner.[54]

(ii) The second group of models starts from the assumption that even a cw laser system generally does not reach a steady state (Baev *et al.*[55]). In this manner pulsed laser systems have already been treated by Keller *et al.*[5] in 1972. The unperturbed temporal development in cw laser systems is regarded as being limited by different perturbing mechanisms such as, for example, mechanical instabilities. For this reason, the lifetime of individual modes has been introduced as a crucial quantity for the sensitivity of intracavity absorption. This concept implies that the sensitivity is time dependent. Such a time dependence has been first observed in 1975 by Antonov *et al.*[8] and Belikova *et al.*[56]

Based on the latter concept, we present an improved theoretical model which allows us to explain the different discrepancies reported in the literature. In particular, the origin of the threshold condition (growing sensitivity at decreasing pump power) is investigated in detail. The described model is demonstrated to agree with the experiments. The results are discussed in Sec. IV. Finally, we present an overview of the different mechanisms which are responsible for the enhanced sensitivity of intracavity absorption.

## II. THEORETICAL MODEL

### A. The rate-equation system

A quantitative analysis of a laser containing an intracavity absorber requires in general a system of rate equations which has to account for the time-dependent variation of the following parameters:

(i) Population densities of the dye molecules inside the gain medium.

(ii) Population densities in the initial and final states of the absorbing transition.

(iii) Radiation field (equal to photon number density) inside the cavity.

Similar rate-equation systems have already been used for passive $Q$ switching and optical bistability in lasers.[57] However, for the purposes of intracavity absorption different approximations are required. Inside the gain medium, effects such as triplet quenching or transitions between inverted states can be neglected. In the absorbing medium one has to consider the different line-broadening mechanisms as well as the possibility of transitions between more than two states. In the following the three different kinds of rate equations are presented which have been used in this paper.

For the gain medium the following approximations are made:

(i) The wavelength dependence of the gain profile is neglected. Throughout this paper the relative depth of an absorption dip is taken as a measure for the sensitivity of the intracavity absorption. For this quantity it makes no difference whether the absorption line is located in the maximum of the emitted line profile or in its wing. It is therefore assumed that the radiation field in the different modes is exposed to the same gain factor.

(ii) Spatial inhomogeneities inside the gain medium such as "spatial hole burning" are neglected. For a linear jet-stream dye laser calculations[58] have shown that spatial inhomogeneities do not influence the sensitivity of intracavity absorption until the system has typically developed for a few milliseconds without any perturbations. Estimates of Baev *et al.*[55] have also demonstrated that spatial effects in a jet stream can be neglected for the intracavity absorption.

(iii) The number of dye molecules in the lower laser level is neglected. The pump rate is assumed to be constant.

With the preceding approximations the rate equation for the gain medium is given by

$$\frac{dn_{\text{inv}}}{dt}=p-A_{\text{inv}}n_{\text{inv}}-B_{\text{inv}}n_{\text{inv}}E_{\text{photon}}\sum_{j=1}^{M}N_{\text{photon}}(j)\ , \quad (1)$$

where $p$ is the pump rate, as obtained from the pump laser power $P$; $A_{\text{inv}}$ is the Einstein $A$ coefficient of the upper level of the gain medium; $n_{\text{inv}}$ is the number of dye molecules in the inverted level; $M$ is the number of axial cavity modes; $N_{\text{photon}}(j)$ is the number of photons in mode $j$; $E_{\text{photon}}$ is the energy of a photon emitted by the gain medium; $B_{\text{inv}}$ is the Einstein $B$ coefficient of the inverted state.

The simplest rate equation of the absorbing medium is obtained for a two-level system. If the absorption acts only on one mode $j'$, the rate equation for the population densities is given by

$$\frac{dn_2}{dt}=-\frac{dn_1}{dt}=B_{12}(n_1-n_2)E_{\text{abs}}N_{\text{photon}}(j')-A_{21}n_2\ , \quad (2)$$

where $E_{\text{abs}}$ is the energy of the absorbed photons; $n_1(n_2)$ is the population density in the lower (upper) level; $A_{21}$ is the Einstein $A$ coefficient of the upper level; $B_{12}$ is the Einstein $B$ coefficient for the absorbing transition.

The third contribution to the system of rate equations describes the number of photons per mode. It provides the coupling between the gain medium and the absorber medium. Therefore, all those terms of the gain equation and of the absorber equation have to appear which are coupled to the radiation field (i.e., the number of photons inside the cavity). The rate equation describing the photon balance per mode $j$ is given by

$$\frac{dN_{\text{photon}}(j)}{dt}=B_{\text{inv}}n_{\text{inv}}E_{\text{photon}}[N_{\text{photon}}(j)+1]-\frac{N_{\text{photon}}(j)}{T_{\text{photon}}}$$
$$-B_{12}(n_1-n_2)E_{\text{abs}}N_{\text{photon}}(j')\ . \quad (3)$$

(i) The last term is identical with the corresponding term of the absorber rate equation. It represents the frequency-dependent losses which are caused by the absorption in mode $j'$. Any spontaneous emission inside the absorber is neglected.

(ii) The first term describes the stimulated emission in the gain medium giving rise to the radiation field. In order to get the system started, an additional term $B_{\text{inv}}n_{\text{inv}}E_{\text{photon}}$ is incorporated. It simulates the spontaneous emission inside the gain medium which corresponds to the second term of Eq. (1).

(iii) $N_{\text{photon}}(j)/T_{\text{photon}}$ characterizes the broadband losses of the cavity. The photon lifetime $T_{\text{photon}}$ is given by

$$T_{\text{photon}}=\frac{L}{c\sigma}\ , \quad (4)$$

where $L$ is the cavity length; $\sigma$ defines the broadband losses per cavity round trip; $\sigma$ contains losses due to the laser output, diffraction, scattering, etc.

Previously used rate-equation systems have not yet treated the influence of the absorber by a separate rate equation. The absorption is usually regarded as a constant, time-independent loss for a particular mode. Only Tohma[59] has already given a numerical analysis involving a separate absorber equation. However, this analysis was confined to the special case of identical absorption cross sections in the gain medium and in the absorber medium.

If the absorption cross section of a particular transition is large enough or if the absorber particle density is sufficiently small, saturation phenomena can occur. They appear as a time-dependent variation of the absorption coefficient due to the change of the population density in the absorbing levels. (This is crucial for the $Q$-switching effect with saturable absorbers.) The parameters of the absorbing medium determine whether the absorption has to be treated as saturated or unsaturated.

In the limiting case of saturated absorption the rate-equation system can no longer be solved in closed form. A numerical integration of the rate equations is required.

For the limiting case of unsaturated absorption, the absorption term in the photon balance equation can be treated as a time-independent loss which affects one or more modes. In this case, one can drop the absorber equation and one is left with an equation system for the gain medium and the photon balance.

Most of the work on intracavity absorption has been concerned with the case of unsaturated absorption. An important analysis for this case has been carried out by Baev *et al.*[55] Within the adiabatic approximation for the inverted medium ($dn_{\text{inv}}/dt=0$), a system of $j$ differential equations for $j$ modes is obtained which can be solved analytically and allows a simple interpretation. In this paper the influence of spontaneous emission and of spatial inhomogeneities in the gain medium on the sensitivity of intracavity absorption has been considered. Contributions due to spatial inhomogeneities are shown to be about three orders of magnitude smaller than the contribution of spontaneous emission. This is true even in the steady-state limit for the photon balance of a typical jet-stream dye laser.

The sensitivity of intracavity absorption is therefore limited by spontaneous emission and corresponds to a minimum detectable absorption coefficient of $10^{-11}$—$10^{-12}$ cm$^{-1}$. These values were first reported by Belikova *et al.*[4]

As a key result Baev *et al.*[55] derived a modified Beer's law for intracavity absorption. Instead of the classical absorption path length a modified path length is obtained. It depends on the average time duration $t$ describing the unperturbed development of the particular modes, and the length of the absorber cell $l$ normalized with respect to the cavity length $L$. In this manner an effective length for the interaction between the laser mode and the absorber is obtained. With $I$ being the intensity of the absorbed mode, $I_0$ the intensity in the absorption-free case, and $\kappa$ the absorption coefficient in cm$^{-1}$, the modified absorption law is given by

$$I(t)=I_0\exp(-\kappa ctl/L)\ . \quad (5)$$

$I/I_0$ represents an intensity ratio defining the depth of an absorption dip which was introduced above to define the

sensitivity of intracavity absorption. The natural logarithm of $(I/I_0)$ equals the effective optical depth $\kappa ctl/L$ of the absorber system.

For the time-dependent linewidth of the laser emission, Baev *et al.*[55] obtained

$$\gamma(t)=\Gamma\left[\frac{t_{\text{mode}}}{t}\right]^{1/2} \quad (\text{for } 0<t\leq t_{\text{mode}}) . \tag{6}$$

Here $t_{\text{mode}}$ is the average mode lifetime which depends on the particular experimental conditions. $\Gamma$ is the laser linewidth at the time $t_{\text{mode}}$ and $\gamma(t)$ is the laser linewidth at some time $t$. This spectral narrowing in time has often been reported (e.g., Antonov *et al.*,[8] see also a theoretical treatment of Meyer and Flamant[60]).

The crucial assumption for deriving Eqs. (5) and (6) is an unperturbed time development of the mode system which is finally limited by mechanical instabilities (jet, resonator), air contaminations, and other "perturbing mechanisms" (see II B). On the average, a mode lifetime $t_{\text{mode}}$ will be established. During this lifetime the system will develop without any perturbations. After a perturbation, the system can be regarded as starting all over again at $t=0$.

Typical mode lifetimes in cw systems range somewhere from 10 $\mu$sec to milliseconds.[58,55] These finite mode lifetimes were observed by Baev *et al.*[61]

### B. Intensity fluctuations in multimode laser systems

Mironenko and Yudson[62] and Kovalenko[63] have investigated the fluctuations of the photon number in the individual modes as well as the fluctuations of the total number of photons. Following the paper of Kovalenko,[63] the fluctuations can be evaluated by means of a Langevin formalism.

The rate-equation system for the gain medium and for the number of photons per mode is extended by stochastic Langevin forces $F(t)$. These stochastic forces simulate quantum-statistical fluctuations of the gain medium and of the radiation field. Their dominant contribution stems from spontaneous emission. In the following we analyze the intensity fluctuations for a particular system of modes which is independent from any intracavity absorber. Hence the appropriate rate-equation system is given by

$$\frac{dn_{\text{inv}}}{dt}=p-A_{\text{inv}}n_{\text{inv}} -B_{\text{inv}}n_{\text{inv}}E_{\text{photon}}\sum_{j=1}^{M}N_{\text{photon}}(j)+F_n(t) , \tag{7}$$

$$\frac{dN_{\text{photon}}(j)}{dt}=B_{\text{inv}}n_{\text{inv}}E_{\text{photon}}[N_{\text{photon}}(j)+1] -\frac{N_{\text{photon}}(j)}{T_{\text{photon}}}+F_j(t) . \tag{8}$$

This classical rate-equation system is equivalent to the quantum-mechanical equations of motion.[64,65]

Within the adiabatic approximation $dn_{\text{inv}}/dt$ is assumed to vanish after the buildup stage of the system. In this manner, an equation for $dN_{\text{photon}}(j)/dt$ is obtained and subsequently linearized. For this purpose the variable of interest $N_{\text{photon}}(j)=N_j$ is expanded around some operating point $\langle N_j\rangle$. As a result, the temporal variation of the fluctuations $\Delta N_j=N_j-\langle N_j\rangle$ is given by

$$\frac{d(\Delta N_j)}{dt}=-\alpha_j(\Delta N_j)-\beta_j\sum_{j=1}^{M}(\Delta N_j)+F_j(t) \tag{9}$$

where $\alpha_j$ and $\beta_j$ are coefficients which originate from the transformation from $dN_j/dt$ to $d(\Delta N_j)/dt$. The $\alpha_j$ will be of crucial importance for the explanation of the experiments. They are given by

$$\alpha_j=\frac{B_{\text{inv}}p}{A_{\text{inv}}\langle N_j\rangle(1+\langle I\rangle B_{\text{inv}}/A_{\text{inv}})} , \tag{10}$$

where $\langle I\rangle=\sum_j\langle N_j\rangle$. $B_{\text{inv}}$ is assumed to be identical for all modes.

Equation (9) represents a system of Langevin equations. The quantum-statistical fluctuations $F_n(t)$ in the gain medium have been neglected with respect to the quantum-statistical fluctuations $F_j(t)$ of the radiation field.

Summing Eq. (9) over all modes, one obtains the time variation of the fluctuations of the total photon number

$$\frac{d(\Delta I)}{dt}=-\Lambda(\Delta I)+F(t) , \tag{11}$$

where $\Delta I=\sum_j(\Delta N_j)$, $F(t)=\sum_j F_j(t)$, and $\Lambda=\sum_j\beta_j$. In Eq. (11) the $\alpha_j$ have been neglected, since they are small compared with $\Lambda$ as shown by Kovalenko.[63] Indeed, the fluctuations of the total photon number turn out to be independent of $\alpha_j$.

With $\Delta N_j=a_j\exp(\lambda t)$ and $\Delta I=b\exp(\lambda t)$ the eigenvalues $\lambda$ of the Langevin equation system (9) and (11) can be determined from

$$(\lambda+\alpha_j)a_j+\beta_j b=0 , \tag{12}$$

$$(\lambda+\Lambda)b=0 , \tag{13}$$

to be $\lambda=-\Lambda$ and $\lambda=-\alpha_j$, respectively. Using these eigenvalues, Eqs. (9) and (11) can be integrated. In this manner the fluctuations of the total photon number and of the photon number in the individual modes can be expressed analytically. In order to interpret the temporal behavior and the mutual dependences of these fluctuations, we now investigate their correlation functions. The following three correlation functions are required:

(i) The autocorrelation of the fluctuations $\Delta I$ for the total photon number

$$\langle\Delta I(t)\Delta I(t+\tau)\rangle=\frac{\langle I\rangle}{2(\eta-1)}\exp(-\Lambda\tau) . \tag{14}$$

(ii) The cross correlation of the fluctuations $\Delta N_j$ in mode $j$ with the sum of the fluctuations in all the other modes $i\neq j$

$$\left\langle\Delta N_j(t)\sum_{i\neq j}\Delta N_i(t+\tau)\right\rangle=-\frac{\langle N_j\rangle^2}{2}\exp(-\alpha_j\tau) +\frac{\langle N_j\rangle}{2(\eta-1)}\exp(-\Lambda\tau) . \tag{15}$$

(iii) The autocorrelation of the fluctuations $\Delta N_j$ in mode $j$

$$\langle \Delta N_j(t)\Delta N_j(t+\tau)\rangle = +\frac{\langle N_j\rangle^2}{2}\exp(-\alpha_j\tau) + \frac{\langle N_j\rangle^2}{2\langle I\rangle(\eta-1)}\exp(-\Lambda\tau)\,, \quad (16)$$

where $\eta$ is the pump rate $p$ normalized with respect to the pump rate at threshold $p_{\text{thr}}$.

Estimates of $\Lambda$ and $\alpha_j$ show that $\Lambda$ is the largest root of Eqs. (9) and (11), and that the $\alpha_j$ are very small.[63] As observed experimentally, the total number of photons fluctuates only very weakly with the small time constant $\Lambda^{-1}$ in agreement with Eq. (14). This is of course also true for the number of photons in the single-mode case.

The eigenvalues $\lambda=-\alpha_j$ vanish in the single-mode case. They only contribute to the correlation functions of those fluctuations which differ from the single-mode case. Hence, the autocorrelation function (14) for the total number of photons contains no $\alpha_j$ dependence. In Eqs. (15) and (16) the $\alpha_j$ describe a slow exponential decrease (weak damping) of the fluctuations. It follows from stability considerations[64] that they give rise to weakly stable solutions of the rate-equation system (9) and (11).

The "$\alpha$ fluctuations" are anticorrelated between individual modes. For a positive $\alpha$ fluctuation of mode $j$ in Eq. (16), the cross correlation (15) with the sum of the fluctuations in all the other modes provides a negative contribution of the same amount.

There are of course also "$\Lambda$ fluctuations" in the individual modes as in case of a single-mode operation. They are correlated in phase and add up to the fluctuation of the total number of photons.

The autocorrelation function (16) for the photon number in one mode is of particular relevance to the sensitivity of intracavity absorption. It will therefore be discussed in more detail.

The coefficient $\langle N_j\rangle^2/[2\langle I\rangle(\eta-1)]$ describes the amplitude of the $\Lambda$ fluctuations. Two cases have to be considered:

(i) Close to threshold $[(\eta-1)\ll 1]$ the $\Lambda$ fluctuations become rather large. This agrees with the result that photon statistics undergo a second-order thermodynamical phase transition from Bose-Einstein statistics to Poisson statistics.[64]

(ii) Well above threshold one has $\langle N_j\rangle \gg \langle N_{\text{thr}}\rangle$ and $(\eta-1)\sim\langle N_j\rangle$. For

$$(\langle \Delta N_j\rangle^2)_\Lambda = \langle \Delta N_j(t)\Delta N_j(t+0)\rangle$$

one obtains with

$$(\langle \Delta N_j\rangle^2)_\Lambda \sim \langle N_j\rangle\,, \quad (17)$$

a pure Poisson distribution.

The coefficient $\langle N_j\rangle^2/2$ describes the amplitude of the $\alpha$ fluctuations. Well above threshold it greatly exceeds the coefficient of the $\Lambda$ fluctuations. The amplitude of the $\alpha$ fluctuations is of the same order of magnitude as the photon number itself. The $\alpha$ fluctuations are therefore large enough to completely quench existing modes. Contrary to (17) $\alpha$ fluctuations are proportional to the photon number and their statistics are non-Poissonian

$$(\langle \Delta N_j\rangle^2)_\alpha \sim \langle N_j\rangle^2\,. \quad (18)$$

From a thermodynamical point of view, this result represents a simple example of a dissipation-fluctuation theorem.[66] In Eq. (18), $\langle N_j\rangle$ can be regarded as a measure for the dissipation of the laser system. The existence of the $\alpha$ fluctuations was observed experimentally by Baev *et al.*[67] using a color-center laser. The stability of the mode system with respect to fluctuations is directly connected to the quantity $\alpha_j$. For smaller damping constants $\alpha_j$, the $\alpha$ fluctuations decrease more slowly in time. If they decrease slowly, the amplitude of the $\alpha$ fluctuations at a certain time is large. Hence, with decreasing values of $\alpha_j$ one has an increasing probability for mode quenching.

According to Eq. (10) we find to a good approximation, that $\alpha_j\sim\langle N_j\rangle^{-1}$ for a multimode system. Therefore, $\alpha_j$ decreases with a growing number of photons per mode which is equivalent to an increasing spectral power density. According to the preceding arguments an increasing spectral power density therefore raises the probability that a mode is completely quenched. For this reason the mode lifetime $t_{\text{mode}}$ is determined by the $\alpha$ fluctuations as long as other effects do not prevail. In other words, the mode lifetime increases with decreasing spectral power density. This result is used for the interpretation of the experiments in Sec. IV.

It is worth noting the analogy to the case of a two-mode operation where the intensities in the individual modes also undergo strong fluctuations. These fluctuations appear as regular oscillations with a fixed frequency which grows with increasing spectral power density.[68] If the period of the oscillation is considered as a two-mode analog of the mode lifetime, the same dependence on the spectral power density is obtained as in a multimode system.

### C. Model calculations

The special case of a single-mode operation has already been treated by Holt.[69] Because of the missing interaction between different modes, the sensitivity enhancement of the intracavity absorption is reduced with respect to the multimode case. In the general case of a multimode operation the problem can be modeled by considering the absorption of only one mode. This applies if the absorption linewidth is smaller than the mode spacing of the laser cavity.

From an experimental point of view, Harris[70] has distinguished narrowband and broadband absorbers. Broadband absorbers can be approximated by the single-mode case, because the interaction between the modes is a higher-order effect with respect to the absorption which affects all modes simultaneously. However, in absorbing systems with lines which are considerably narrower than the laser linewidth, only a small fraction of the total number of modes is influenced by the absorption. Under these conditions a redistribution of the energy in the different modes takes place inside the gain medium. For this particular situation the sensitivity of intracavity absorption is

enhanced most strongly compared to a normal absorption outside the laser cavity.

In the following model calculations the limiting cases of saturated and unsaturated absorption are distinguished in treating the rate equations described above. Under the important assumption of a finite-mode lifetime, a steady state in a cw laser system, as it was assumed in Refs. 6, 50–52, can for technical reasons never be reached. A numerical integration of the rate equations has been carried out to test the validity of the different theoretical models. Most of the subsequent numerical solutions are obtained for the special case of a two-mode operation where only one mode is exposed to an absorption by a two-level system.

### 1. *Unsaturated absorption*

The effects limiting the mode lifetime are not included directly in the rate-equation system. Instead, the mode lifetime determines the overall integration time of the system. In this manner it is an external boundary condition for the numerical integration of the rate equations.

The temporal development of the rate-equation system is shown in Fig. 1. The pump parameter $P^*$ is defined as the actual pump power $P$ normalized with respect to the pump power at threshold $P_{\text{thr}}$. $M$ is the number of oscillating modes. The total particle density inside the absorber is given by $n = n_1 + n_2$. Figure 1 initially reveals a linear dependence in time for the sensitivity $S = -\ln(I/I_0)$ of the intracavity absorption. This is the

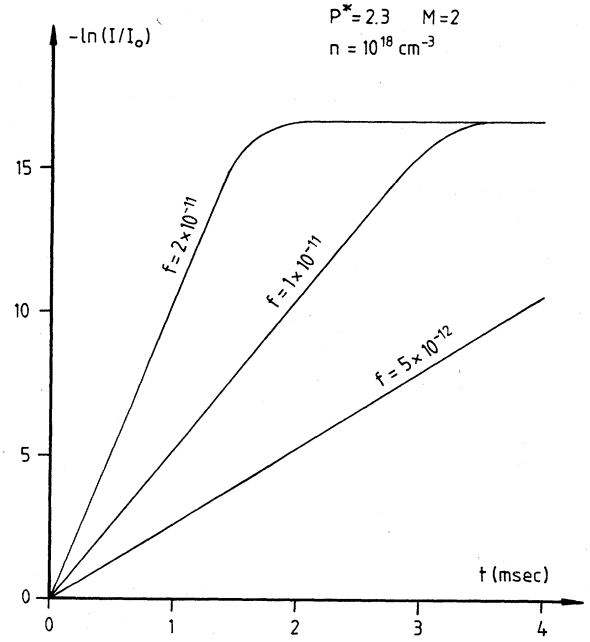


FIG. 1. Model calculations of unsaturated intracavity absorption for three different absorber oscillator strengths $f$, and for $n = 10^{18}$ cm$^{-3}$. The sensitivity $S = -\ln(I/I_0)$ is shown as a function of the integration time $t$ of the rate-equation system. The linear relation between $S$ and $t$ represents the validity region of Beer's law. Note that the limiting sensitivity does not depend on $f$.

prediction of the modified Beer's law in Eq. (5). The slope of the line is $\kappa cl/L$. The relative absorption depth reaches its maximum value when the temporal development has advanced far enough that in the absorbed mode the spontaneous emission of the gain medium takes over. The limiting value which corresponds to the maximum sensitivity is therefore independent of the absorption coefficient. This numerical result reveals very clearly the importance of the temporal dynamics of the laser system. It also demonstrates that for almost any practical situation a steady-state analysis is inadequate.

For the unsaturated case the absorption coefficient does not change in time. $(n_2/n_1)$ can be neglected because the depletion rate out of the upper excited state exceeds the absorber pump rate at any time. This situation can be treated using a constant, frequency-dependent absorption loss in the photon balance. The linear dependence of $S$ on the absorption coefficient $\kappa$ according to Beer's law (5) can be exploited for a quantitative determination of $\kappa$. This will be explained in more detail below.

### 2. *Saturated absorption*

Already for an oscillator strength $f$ as low as $10^{-8}$ and for realistic particle densities, relative population densities $(n_2/n_1)$ in the range of 0.01–0.001 can occur in a two-level system. Even for such a situation the saturation of the absorption has to be taken into account. A solution of the rate-equation system provides a rather weak threshold behavior of the sensitivity on the spectral power density. This effect increases with a growing oscillator strength. Figure 2 shows the relative population density $(n_2/n_1)$ in the absorber together with the sensitivity $S = -\ln(I/I_0)$ as a function of the spectral power density $(P^*-1)/M$. The curves have been calculated for $f = 10^{-8}$, a total particle density of $10^{15}$ cm$^{-3}$, and an integration time of 0.1 msec. If the absorption oscillator strength is large, saturation phenomena in intracavity absorption can already occur at rather small radiation fields in the milliwatt range. This is true for absorber particle densities which are not too large. Saturation effects in intracavity absorption were first mentioned by Maeda *et al.*[17] and observed by Mironenko and Pack.[71]

Apart from the total radiation power, it is the ratio of the absorption linewidth with respect to the bandwidth of the radiation field which is important for the degree of saturation. The relevant parameter is the spectral power density as demonstrated by Antonov *et al.*[72] Measured sensitivities from Ref. 54 taken at different pump parameters $P^* = P/P_{\text{thr}}$ and at different laser bandwidths $\Delta\lambda$, were normalized with respect to the bandwidths. As a result, one common curve was obtained for the sensitivity as a function of the spectral power density $(P^*-1)/\Delta\lambda$ which was independent of the particular bandwidths.

Absorption lines with large transition moments are, for example, atomic resonance lines and molecular lines such as those of $I_2$ (with oscillator strengths $f \simeq 10^{-4}$). In such systems the reduced sensitivity at high spectral power densities appears to be primarily due to saturation effects in the absorber. Calculations with different particle densities and oscillator strengths ranging from $10^{-4}$ to 1 were

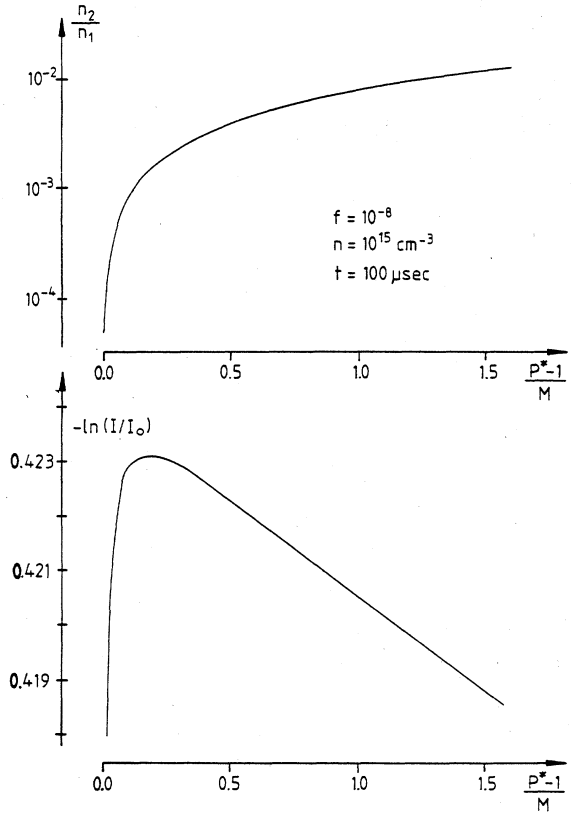


FIG. 2. Model calculation of intracavity absorption for a weakly saturated case ($f=10^{-8}$, $n=10^{15}$ cm$^{-3}$, $t=100$ μsec). The relative population density ($n_2/n_1$) (upper part) and the sensitivity $S=-\ln(I/I_0)$ (lower part) are shown as a function of the spectral power density.

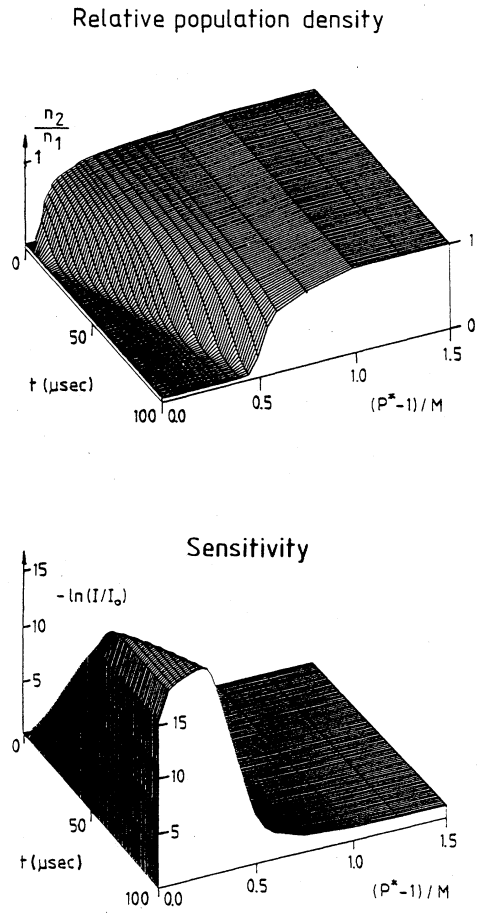


FIG. 3. Model calculations of saturated intracavity absorption ($f=0.65$, $n=10^9$ cm$^{-3}$). The relative population density ($n_2/n_1$) and the sensitivity $S=-\ln(I/I_0)$ are shown as a function of the spectral power density, and of the integration time of the rate-equation system. The sensitivity enhancement at laser threshold is clearly due to the absence of saturation in this region.

carried out to prove this point.

As a typical example of these calculations Fig. 3 shows the sensitivity $S$ as well as the relative population density ($n_2/n_1$) as a function of the spectral power density $(P^*-1)/M$ and of the time $t$. The other parameters were $f=0.65$ (Na $D_2$—resonance line with an assumed linewidth of 0.01 cm$^{-1}$) and a total particle density of $10^9$ cm$^{-3}$.

The sensitivity clearly increases with decreasing spectral power density and with progressing time. A comparison of the upper and lower part of Fig. 3 shows that the largest sensitivity occurs in the region where the relative population density ($n_2/n_1$) vanishes, i.e., for the unsaturated case. In this example the threshold effect due to the saturation in the absorption is much more pronounced than in Fig. 2.

As the main results of the numerical integrations, two points should be emphasized for the case of saturated absorption:

(i) The absorption coefficient is not constant, but changes in time according to the population difference in the absorbing levels.

(ii) Only a time-dependent absorption coefficient can be obtained from the relative absorption depth. Similar to the unsaturated case with a constant absorption coefficient $\kappa$, a modified Beer's law holds again with $\kappa=\kappa(t)$.

## III. EXPERIMENTAL

For an experimental test of the model calculations suitable absorbers had to be selected which satisfy the conditions of saturated and unsaturated absorption.

As a transition with a very small absorption coefficient (unsaturated absorption) the $^PP(11)$ line and the $^PQ(11)$ line of the (2-0) band of the $b\,^1\Sigma_g^+$-$X\,^3\Sigma_g^-$ system of the $O_2$ molecule were chosen. Their line positions are 630.276 and 630.200 nm, respectively, where no overlap with other $O_2$ lines or with atmospheric water vapor lines[73] has to be considered. The relative absorption depths of the two lines can be compared with theoretical values.

The case of saturated absorption was studied by means of two transitions which were selected from the (11-3) band and the (9-2) band of the $B\,^3\Pi_u^+$-$X\,^1\Sigma_g^+$ system of the $I_2$ molecule. Since the experiment was carried out under atmospheric pressure, the lines showed a pronounced pressure broadening. They had a full width at half maximum (FWHM) of approximately 0.3 cm$^{-1}$. For this reason individual absorption lines could not be resolved. In the fol-

lowing table the line positions and the quantum numbers of the corresponding transitions[74] are given together with the line numbers according to the atlas of Gerstenkorn and Luc[75]:

| Line No. | Position ($cm^{-1}$) | $v''$ | $v'$ | $J''$ | $J'$ |
|---|---|---|---|---|---|
| 2154 | 16280.569 | 2 | 9 | 93 | 94 |
| 2155 | 16280.607 | 3 | 11 | 90 | 89 |

These lines are again not blended by any atmospheric $H_2O$ lines.[73]

A cw jet-stream dye laser with a folded cavity (length $L=60$ cm) has been operated with an optimized mixture of Rhodamine-6G and Rhodamine-101. The wavelength was tuned by means of an interference filter coated for 450 nm. The commercial birefringent filter had to be removed since it produced a rather small laser linewidth which is unfavorable for sensitive intracavity absorption.

In case of the $O_2$ absorption the normal laboratory atmosphere was sufficient to detect the absorption lines mentioned above. In this case, $l/L=1$. The $I_2$ measurements were carried out in a 10-cm-long absorption cell placed inside the cavity. The cell has been sealed off with Brewster windows. To ensure a rather small $I_2$ concentration, the absorption cell containing the $I_2$ was alternately heated and evacuated for several days before taking the spectra. The absorption features of both molecules were observed under normal environmental conditions (760 Torr, 300 K). Apart from the tuning filter and the absorption cell, the standard mirrors for Rhodamine-6G were the only optical elements inside the cavity.

The dye laser was operated on a vibration-free table to minimize mechanical instabilities. A laminar flow box was covering the whole experimental setup to avoid any perturbing dust particles inside the laser cavity.

The dye laser was pumped by a 5-W argon-ion laser. Between the pump laser and the dye laser a chopper element was inserted. By means of this chopper element the duration of the dye laser emission is accurately defined as the duration of the transmitted pump pulses. Depending on the particular experimental requirements a mechanical chopper or an electro-optic modulator (Pockels cell) was used.

The radiation emitted by the dye laser was observed by means of a 2-m grating spectrometer. The spectrum was scanned photoelectrically and detected by means of a lock-in amplifier. In addition, the modulation frequency of the pump laser served as the reference signal for the lock-in amplifier. With the described setup, dye laser pulses as short as 700 nsec were analyzed.

An experimental arrangement described by Stoeckel *et al.*[76] and Chenevier *et al.*[77] provides a similar kind of information. In addition to an electro-optic modulator placed between pump laser and dye laser, they inserted a fast acousto-optic modulator between the dye laser and the detection system. This second modulator was triggered by the first one with a certain time delay. The signal transmitted by the delayed modulator then provides the actual spectrum of the dye laser as it has evolved until the instant specified by the time delay. In contrast to these experiments, our setup provides an integral measurement. The time interval over which our measurements extend, is very large compared with the pulse duration of the laser system. This fact ensures that the measurements yield a statistical average of the analyzed signals.

## IV. RESULTS AND DISCUSSION

### A. Relations between spectral power density, mode lifetime, and sensitivity

Convincing evidence against the hypothesis of a steady state in a cw multimode laser system was presented by Baev *et al.*[61] in 1980. They observed that the individual axial modes of a cw dye laser have a finite lifetime. The interactions in the coupled gain-absorber system therefore show a dynamical behavior in time. For the limiting case of unsaturated absorption the temporal dynamics have already been described.[55] The influence of the spectral-power density, however, has not yet been understood.

In 1982 S. J. Harris[58] analyzed the Fourier spectrum of a cw multimode dye laser. At high spectral power densities the individual modes turned on and off with frequencies as high as 100 kHz. This value corresponds to a mean mode lifetime of 10 $\mu$sec. He also reported that the fluctuation frequencies decreased at low spectral power densities.

In order to investigate the relation between the mode lifetime and the spectral power density, measurements have been carried out using the $O_2$ molecule as an absorber which is clearly unsaturated. The absorption coefficient at the center of the $O_2$ lines can be calculated from the oscillator strengths of the corresponding transitions, the particle density in the initial state, and the FWHM of the lines $\Delta\bar{\nu}$. The oscillator strengths were determined by the Einstein $A$ coefficient of the (0-0) band,[78] the Franck-Condon factors of the ($b$-$X$) system,[79] and the Hönl-London factors as given by Watson.[80] The particle density in the initial state was calculated for a standard atmosphere ($O_2$ fraction of 21 %), and for normal pressure (760 Torr) and temperature (300 K). The thermal population density in the rotational levels is of course given by a Boltzmann distribution. With these quantities one obtains

$$\kappa({}^{P}P(11))=(3.54\times10^{-8}\ \mathrm{cm}^{-2})\frac{1}{\Delta\bar{\nu}} \tag{19}$$

and

$$\frac{\kappa({}^{P}P(11))}{\kappa({}^{P}Q(11))}=0.899\ . \tag{20}$$

$\Delta\bar{\nu}$ ($cm^{-1}$) can be taken from measurements. As indicated above, intracavity absorption follows a modified Beer's law [Eq. (5)]. Since the absorption coefficient $\kappa$ for the lines of interest can be obtained from Eq. (19), the measurement of $(I/I_0)$ provides a measure of $t_{\mathrm{mode}}=-\ln(I/I_0)/\kappa c$. Hence the dependence of $t_{\mathrm{mode}}$ on the spectral power density can be determined from $S=-\ln(I/I_0)$.

Figure 4 shows the observed absorption dips of the ${}^{P}Q(11)$ and ${}^{P}P(11)$ lines for a pump pulse duration of

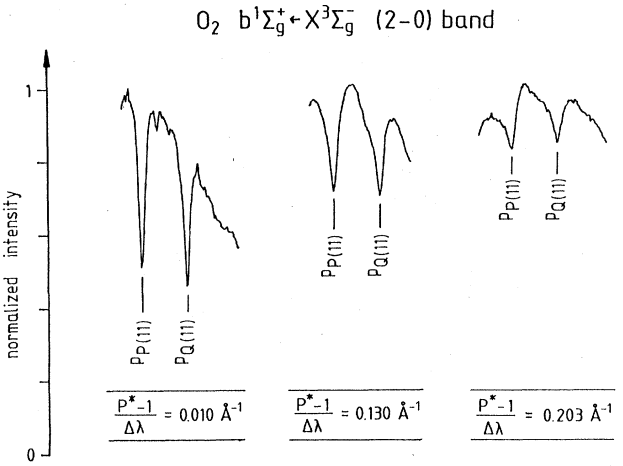


FIG. 4. Unsaturated absorption lines of atmospheric molecular oxygen for three different spectral power densities. The intensities are normalized with respect to the maximum intensity of the laser output. The sensitivity decreases with increasing spectral power density $(P^*-1)/\Delta\lambda$.

$t_{chop}=175$ $\mu$sec, and for three different spectral power densities. The absorption depths clearly decrease with growing spectral power density.

Quantitative results for the sensitivity as a function of the spectral power density are presented in Fig. 5. The

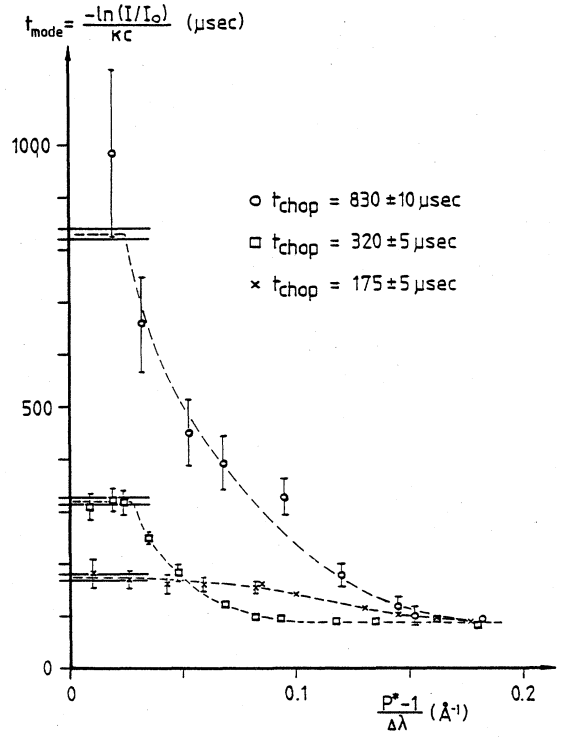


FIG. 5. Mode lifetime $t_{mode}=-\ln(I/I_0)/\kappa c$ as obtained from Beer's law [Eq. (5)], determined as a function of the spectral power density. The measurements have been performed for the case of unsaturated absorption and for three different pump-pulse durations $t_{chop}$. At small spectral power densities $t_{mode}$ approaches $t_{chop}$ as indicated by the double bars. The dashed lines are drawn to guide the eye.

measurements were carried out for three different pump-pulse durations $t_{chop}$. The value of $t_{chop}$ defines an upper limit for the mode lifetime $t_{mode}$. With decreasing spectral power density $(P^*-1)/\Delta\lambda$ the mode lifetime $t_{mode}$ increases until the value of $t_{chop}$ is reached. A further decrease of the spectral power density $(P^*-1)/\Delta\lambda$ does not alter $t_{mode}$ any more. This result strongly supports the validity of Beer's law for intracavity absorption. The slightly different behavior of the curve corresponding to $t_{chop}=320$ $\mu$sec results from a readjustment of the whole laser system.

In order to exclude systematic errors due to weak etalon effects,[81] the ratio of $\kappa(^PQ(11))$ with respect to $\kappa(^PP(11))$ was tested for different spectral power densities and $t_{chop}=175$ $\mu$sec. The calculated ratio of the absorption coefficients is directly related to the measured ratios of $S$. For an average over 70 measurements we obtained a ratio of $0.892\pm0.013$ in excellent agreement with the theoretical value 0.899 given by Watson.[80]

The increased mode lifetime close to laser threshold gives rise to an enhanced sensitivity of intracavity absorption. This threshold behavior has been mentioned in various papers. Nevertheless, its origin has so far not been understood. Spatial inhomogeneities (spatial hole burning) can be excluded as a possible reason, because the threshold condition described above has also been observed in a ring dye laser.[1] The running-wave character of the radiation field avoids any spatial hole burning inside the gain medium.

We attribute the threshold behavior to intensity fluctuations in each particular mode of the laser system. The amplitudes of these intensity fluctuations are large enough to completely quench individual modes. In Sec. II B the probability for such a process was shown to increase for increasing spectral power densities. On a temporal average this manifests itself in a decrease of the mean mode lifetime as observed in the experiments.

It is important to realize that the threshold condition due to the intensity fluctuations is of fundamental nature and cannot be avoided by any technical efforts. Other effects limiting the mode lifetime, such as mechanical instabilities, inhomogeneities of the dye jet (bubbles), or air contaminations, can be minimized by a proper experimental arrangement.

In order to define the mode lifetime by a quantity which is experimentally accessible, the pump radiation has been periodically modulated with a time constant $t_{chop}$. The sensitivity $S=-\ln(I/I_0)$ has been measured as a function of the pump pulse duration $t_{chop}$ for three different pump parameters $P^*=P/P_{thr}$.

Figure 6 presents the measured sensitivities together with their statistical errors. At a pump-pulse duration of some 10 $\mu$sec the curves for different $P^*$ start to separate and become nonlinear. The largest pump parameter $(P^*=3.5)$ provides the lowest sensitivity. The sensitivity corresponding to $P^*=1.13$ increases still up to $t_{chop}\simeq1.5$ msec. Furthermore, the spectral width $\Delta\lambda$ of the laser emission narrows for an increasing pump-pulse duration in agreement with Eq. (6).

According to Beer's law the mode lifetime $t_{mode}$ can be calculated, if the absorption coefficient $\kappa$ and the sensi-

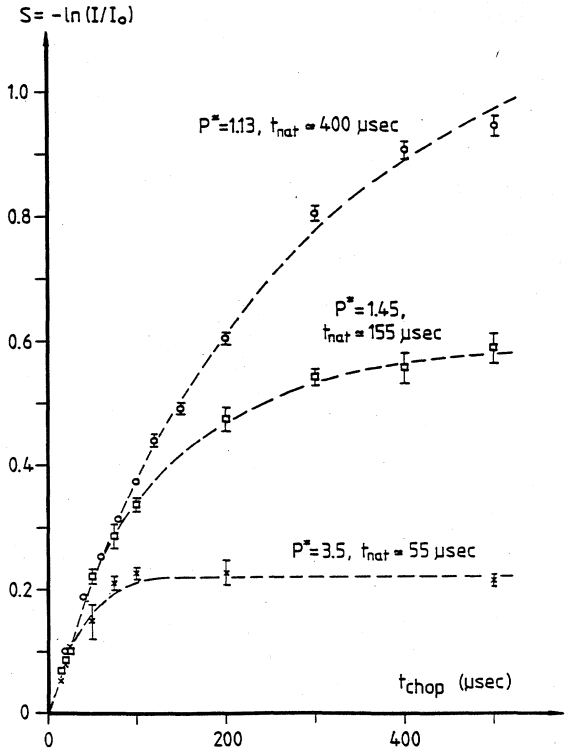


FIG. 6. Sensitivities $S=-\ln(I/I_0)$ as a function of the pump-pulse duration $t_{chop}$. The measurements have been performed for the case of unsaturated absorption and for different pump parameters $P^*$ which give rise to different natural mode lifetimes $t_{nat}$. The dashed curves are calculated by means of a model in which the average mode lifetime $t_{mode}$ is obtained from the pump-pulse duration $t_{chop}$.

tivity $S$ are known. If the pump radiation is not modulated, a natural mode lifetime $t_{nat}$ of the laser system can be defined. In Fig. 6, $t_{nat}$ is indicated together with the corresponding pump parameter. As already explained, $t_{nat}$ decreases with increasing spectral power density.

The dashed curves in Fig. 6 are calculated using a model which takes into account the difference between the actual mode lifetime $t_{mode}$ and the pump-pulse duration $t_{chop}$. This model is derived from the statistical nature of the measurements as pointed out at the end of Sec. III, where the average mode lifetime $t_{mode}$ represents a mean value obtained from an observation over a long period in time. Since the mode lifetime $t_{mode}$ follows an exponential distribution, its probability density decreases exponentially in time

$$P(t)=\lambda\exp(-\lambda t)\ . \tag{21}$$

The temporal mean value of $P(t)$ is $\lambda^{-1}$. Hence with $\lambda^{-1}=\langle t_{nat}\rangle$ a probability density is generated which describes the sum over all contributions to an observed "average mode" with an intrinsic lifetime $t_{nat}$. As described above, $t_{nat}$ is accessible to measurement.

By means of a pump-pulse duration $t_{chop}$ any mode lifetime exceeding $t_{chop}$ is cut off. On the other hand, mode lifetimes shorter than $t_{chop}$ give a mean value $\langle t_s\rangle$. As a result, the total average of the mode lifetime in the modulated situation is given by a weighted mean value of $\langle t_s\rangle$ and of $t_{chop}$. The weighting factors are $\int_0^{t_{chop}} P(t)dt$ and $\int_{t_{chop}}^{\infty} P(t)dt$, respectively. Figure 7 illustrates the described model for $t_{nat}=400$ µsec and $t_{chop}=175$ µsec. An average mode lifetime $t_{mode}=142$ µsec is obtained.

As already shown, the mode lifetime $t_{mode}$ depends on the spectral power density $(P^*-1)/\Delta\lambda$. In addition, the bandwidth $\Delta\lambda$ is proportional to $t^{-1/2}$. Hence the resulting mode lifetime $t_{mode}$ has to be corrected for each particular pump-pulse duration $t_{chop}$. According to Beer's law one then can calculate a sensitivity $S$ from each resulting value of $t_{mode}$. The dashed curves in Fig. 6 show $S$ as a function of $t_{chop}$ as determined by the described method. The measurements are very well reproduced by the calculations.

With the preceding method one obtains the quantity $t_{mode}$ which is required in Beer's law, from a quantity which can be directly measured, namely $t_{chop}$. Beer's law therefore provides a quantitative determination of the absorption coefficient $\kappa$ which depends only on $t_{mode}$ and $S$ in the case of unsaturated absorption.

This method simplifies greatly, if $t_{chop}$ is small with respect to $t_{nat}$. In this case $t_{mode}$ is sufficiently well approximated by $t_{chop}$ which can directly be used as the mode lifetime in Beer's law. The dependence of $S$ on $t_{chop}$

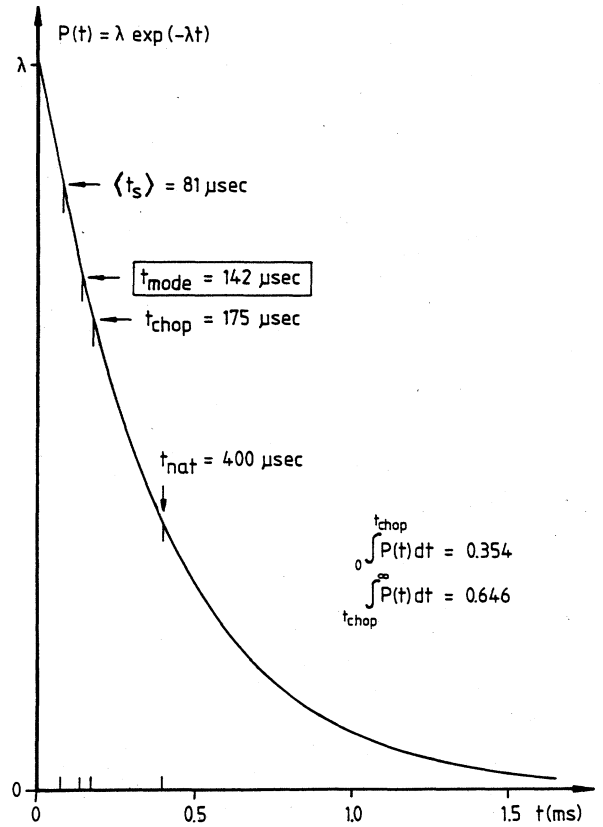


FIG. 7. Exponential probability density characterizing the mode lifetime. In order to illustrate the model described in the text, numerical values are drawn for $t_{nat}=400$ µsec and $t_{chop}=175$ µsec. They give rise to a mode lifetime $t_{mode}=142$ µsec.

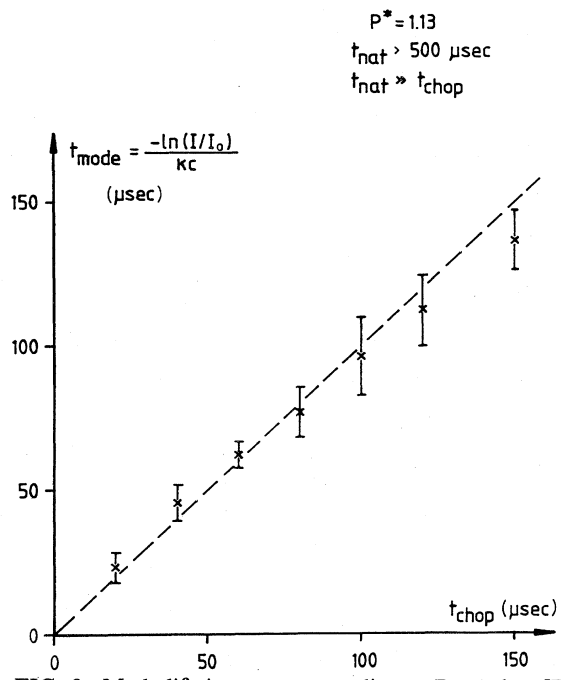


FIG. 8. Mode lifetime $t_{mode}$ according to Beer's law [Eq. (5)], as a function of the pump-pulse duration $t_{chop} \ll t_{nat}$. For the unsaturated case a linear dependence is obtained where $t_{mode} \simeq t_{chop}$. In this manner, the absolute value of small absorption coefficients can be easily determined from $t_{chop}$ and $(I/I_0)$ by means of Beer's law.

is therefore linear for $t_{chop} \ll t_{nat}$.

In order to illustrate this linearity, Fig. 8 presents the actual mode lifetime $t_{mode} = -\ln(I/I_0)/\kappa c$ as a function of $t_{chop}$. $\kappa$ is the calculated absorption coefficient for the $^{P}P(11)$ line as obtained by Eq. (19). The corresponding pump parameter is $P^* = 1.13$ which gives a natural mode lifetime of about 400 $\mu$sec.

Figure 8 clearly shows that $t_{mode}$ and $t_{chop}$ agree within the statistical errors, as long as $t_{chop}$ is smaller than 120 $\mu$sec. From the measured depths $(I/I_0)$ and for $t_{mode} \simeq t_{chop} \leq 120$ $\mu$sec one obtains an absorption coefficient $\kappa$ in the center of the line

$$\kappa_{exp} = (1.36 \pm 0.14) \times 10^{-7} \text{ cm}^{-1} .$$

The absorption coefficient as calculated according to Eq. (19) is

$$\kappa_{calc} = (1.24 \pm 0.13) \times 10^{-7} \text{ cm}^{-1} ,$$

where the FWHM $\Delta\bar{\nu}$ has been taken from measurements. The given error originates from the statistical error of the measured absorption linewidth $\Delta\bar{\nu} = (0.285 \pm 0.029)$ cm$^{-1}$. The experimental and the calculated values of $\kappa$ are in good agreement.

It should be noted that the condition $t_{chop} \ll t_{nat}$ and therefore the linearity of $S$ as a function of $t_{chop}$ is not generally required for the determination of $t_{mode}$ from $t_{chop}$. In order to measure small absorption coefficients, the described method can be applied to any pump-pulse duration which is smaller than the natural mode lifetime.

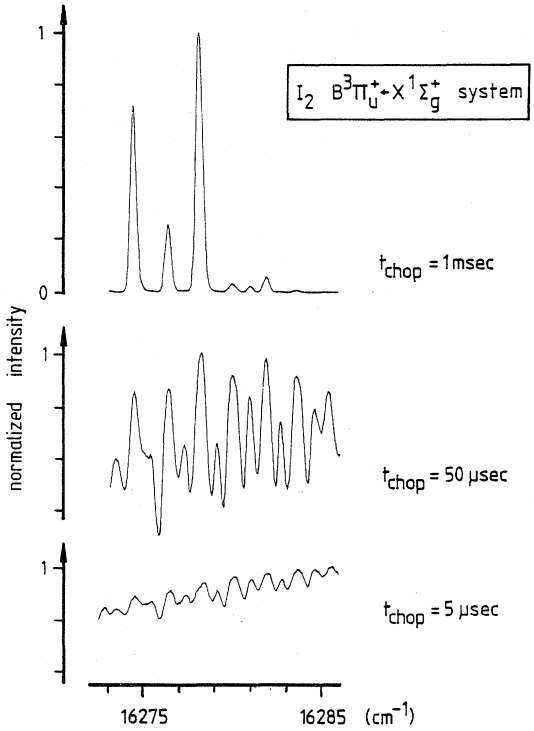


FIG. 9. Saturated absorption features of the iodine molecule around 614 nm for three different pump-pulse durations $t_{chop}$. The intensities are normalized with respect to the maximum intensity of the laser output. For large values of $t_{chop}$ the laser emission spectrum shows a pronounced spectral condensation around those wavelengths where no absorption occurs.

### B. Saturation of the absorption and its influence on the sensitivity

Saturation phenomena in intracavity absorption have been first observed by Mironenko and Pack[71] in 1978. For the Na $D$ lines they demonstrated a dramatic decrease of the absorption depth for increasing spectral power density. On the other hand, the absorption depths of the neighboring $H_2O$ lines were only reduced by a significantly smaller amount. Hence, the behavior of the $H_2O$ lines agrees well with the preceding results for the case of unsaturated absorption.

As a result of the numerical analysis it turns out that significant saturation phenomena may already occur for an oscillator strength as small as $f = 10^{-4}$, if the absorber particle density is not too large. For this reason we studied the saturation behavior of the absorption by means of the $I_2$ ($B^3\Pi_u^+$-$X^1\Sigma_g^+$) system. The selected transitions are given above.

Figure 9 shows $I_2$ spectra obtained for three different pump-pulse durations $t_{chop}$ in the relevant wavelength region. The intensity is normalized with respect to the maximum intensity of the laser output. For short pump-pulse durations, the absorption features are well developed and can easily be identified according to the atlas of Gerstenkorn and Luc.[75] For long pump-pulse durations $t_{chop}$, the total laser power emitted concentrates on a few "lines" at those wavelengths where the $I_2$ spectrum shows no ab-

sorption. In this situation, a measurement of an absorption depth becomes, of course, impossible. However, for shorter pump-pulse durations of some 10 $\mu$sec such an absorption depth can still be measured.

In order to demonstrate saturation effects inside the absorber, we used the results of Sec. IV A in the following manner. With the condition $t_{chop} \ll t_{nat}$, we have already shown that the sensitivity $S$ depends linearly on $t_{chop}$ in the case of unsaturated absorption. This linearity holds until $t_{mode}$ is sufficiently well approximated by $t_{chop}$. At very long pump-pulse durations the mode lifetime eventually becomes independent of $t_{chop}$ and is simply given by $t_{nat}$.

If saturation processes become important for the absorption, the relative population density $(n_2/n_1)$ changes in time, and the absorption coefficient can no longer be regarded as constant. In contrast to the unsaturated case, we therefore expect a nonlinear dependence of $S$ on $t_{chop}$ at rather short pump-pulse durations. Furthermore, an increased spectral power density should result in a reduced sensitivity.

Figure 10 shows the measured sensitivity $S$ as a function of the pump-pulse duration $t_{chop}$ for two different pump parameters $P^*$ and for a constant particle density. The pump parameters are small enough to guarantee a natural mode lifetime $t_{nat} \geq 300$ $\mu$sec. The nonlinear relation between the sensitivity $S$ and $t_{chop} \simeq t_{mode}$ can only be

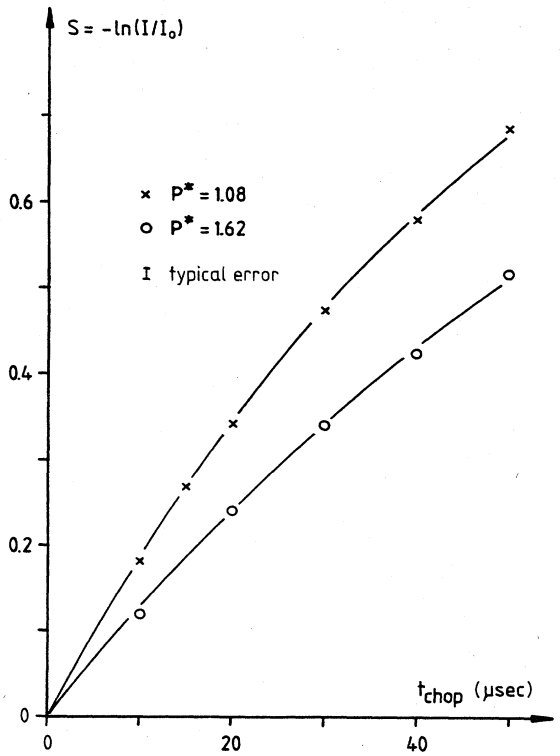


FIG. 10. Sensitivity $S = -\ln(I/I_0)$ as a function of the pump-pulse duration $t_{chop} \ll t_{nat}$. The measurements have been performed for the case of saturated absorption and for two different pump parameters $P^*$. Instead of the linearity noticed in Fig. 8 (unsaturated absorption), $S$ clearly shows a nonlinear dependence on $t_{chop}$. Note that $S$ increases with decreasing $P^*$.

explained in the following manner. Since in Beer's law all quantities are accurately defined with the only exception of $\kappa$, the nonlinearity has to result from a time-dependent absorption coefficient $\kappa$.

This situation generally occurs, if saturation effects give rise to time-dependent population densities in the absorbing levels. Since the population $n_2$ in the upper level can no longer be neglected, the population difference between the absorbing levels has to be considered in the rate-equation system given in Sec. II A. Hence, the dynamical behavior of the coupled rate equations gives rise to a time-dependent absorption coefficient (compare the numerical calculations of II C 2). A modified Beer's law with $\kappa = \kappa(t)$ describes this situation

$$I = I_0 \exp[-\kappa(t) ctl/L] \,. \tag{22}$$

So far the experimental results are clearly understood by the saturation of the absorption. For such a condition, an increased pump parameter causes, of course, a reduced sensitivity. This is also shown in Fig. 10. In this context, observations of Morgan *et al.*[82] can readily be explained by saturation effects. Morgan *et al.* have also investigated intracavity absorption of $I_2$, but their interpretation has been erroneously attributed to spatial inhomogeneities.

The experimental results demonstrate that even in molecular absorber systems (many-level systems) a pronounced saturation of intracavity absorption can occur. This is also intended as a warning, because any quantitative determination of small oscillator strengths or, in particular, of small particle densities may suffer from saturation inside the absorber.

Because of the saturation of the absorption, a second type of threshold condition exists which determines the sensitivity of intracavity absorption as a function of the spectral power density. It depends on the oscillator strength and the particle density of the absorber. This threshold condition is determined by the temporal dependence of the relative population density $(n_2/n_1)$ in the absorbing levels.

## V. SUMMARY

The preceding investigations present for the first time a complete and consistent model explaining intracavity absorption. The theoretical predictions have been verified by experiments. An overview summarizing the different important quantities, is schematically illustrated in Fig. 11.

The theoretical considerations are based on a time-dependent model. Besides mechanical and other externally imposed instabilities, the unperturbed development of a cw multimode laser system is shown to be limited by intensity fluctuations which are driven by stochastic quantum-statistical forces. cw multimode laser systems can therefore be regarded as systems generating a train of individual pulsed modes. For this reason the temporal dynamics of such systems are of paramount importance where the lifetime of individual modes plays the key role for the sensitivity of intracavity absorption. The assumption of a steady state in cw multimode lasers with nar-

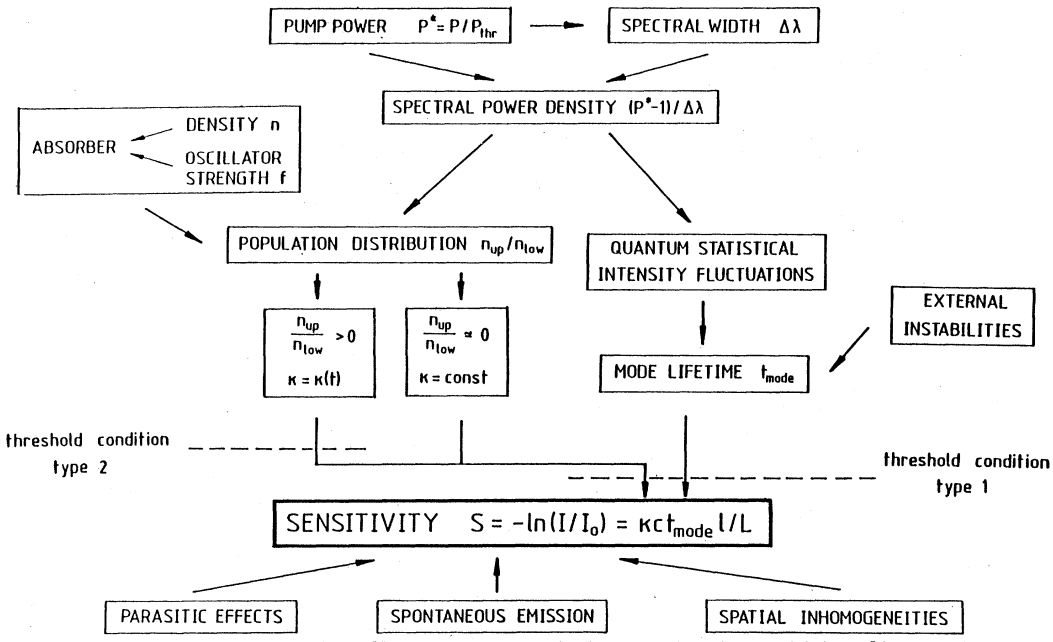


FIG. 11. Overview summarizing the different processes which determine the sensitivity of intracavity absorption.

rowband absorbers can in general not be maintained.

As illustrated in Fig. 11, intracavity absorption follows a modified Beer's law. This implies that the sensitivity $S = -\ln(I/I_0)$ is proportional to the mode lifetime $t_{\text{mode}}$ and the absorption coefficient $\kappa$. The mode lifetime is limited by strong intensity fluctuations, and increases with decreasing spectral power density inside the cavity. For this reason a fundamental threshold condition is imposed which does not depend on any absorber parameters.

The sensitivity of intracavity absorption is eventually limited by the spontaneous emission.[4] The limiting sensitivity does therefore not depend on the value of the absorption coefficient. It might be reduced by spatial inhomogeneities in the gain medium, if the active gain volume is no longer negligible with respect to the cavity volume. Furthermore, the sensitivity can be affected by frequency-selective losses caused by parasitic effects[81] such as weak etalon effects.

Saturation processes inside the absorber have been described by means of a rate-equation system containing a separate absorber equation. In this manner a possible redistribution of the population is taken into account. The time-dependent population densities in the absorber levels depend on the intracavity spectral power density and on the corresponding transition moment. This behavior is described by a modified Beer's law with a time-dependent absorption coefficient. Since saturation decreases with a reduced spectral power density, the sensitivity of intracavity absorption increases close to the lasing threshold. This gives rise to a second type of threshold condition which is superimposed on the fundamental threshold condition due to the power-dependent intensity fluctuations.

[1]A. A. Kachanov and T. V. Plakhotnik, Opt. Commun. **47**, 257 (1983).
[2]L. A. Pakhomycheva, E. A. Sviridenko, A. F. Suchkov, L. A. Titova, and S. S. Churilov, Pis'ma Zh. Eksp. Teor. Fiz. **12**, 60 (1970) [Sov. Phys.—JETP Lett. **12**, 43 (1970)].
[3]N. C. Peterson, M. J. Kurylo, W. Braun, A. M. Bass, and R. A. Keller, J. Opt. Soc. Am. **61**, 746 (1971).
[4]T. P. Belikova, E. A. Sviridenko, A. F. Suchkov, L. V. Titova, and S. S. Churilov, Zh. Eksp. Teor. Fiz. **62**, 2060 (1972) [Sov. Phys.—JETP **35**, 1076 (1972)].
[5]R. A. Keller, E. F. Zalewski, and N. C. Peterson, J. Opt. Soc. Am. **62**, 319 (1972).
[6]T. W. Hänsch, A. L. Schawlow, and P. E. Toschek, IEEE J. Quantum Electron. **QE-8**, 802 (1972).
[7]V. S. Burakov, Zh. Prikl. Spektrosk. **35**, 223 (1981) [J. Appl. Spectrosc. **35**, 843 (1981)].
[8]E. N. Antonov, V. G. Koloshnikov, and V. R. Mironenko, Opt. Commun. **15**, 99 (1975).
[9]V. M. Baev, V. Ya. Gulov, E. A. Sviridenko, and M. P. Frolov, Kvant. Elektron. (Moscow) **2**, 1328 (1975) [Sov. J. Quantum Electron. **5**, 724 (1975)].
[10]R. G. Bray, W. Henke, S. K. Liu, K. V. Reddy, and M. J. Berry, Chem. Phys. Lett. **47**, 213 (1977).
[11]W. T. Hill, R. A. Abreu, T. W. Hänsch, and A. L. Schawlow, Opt. Commun. **32**, 96 (1980).
[12]G. H. Atkinson, A. H. Heimlich, and M. W. Schuyler, J. Chem. Phys. **66**, 5005 (1977).
[13]E. N. Antonov, E. B. Berik, and V. G. Koloshnikov, J. Quant. Spectrosc. Radiat. Transfer **22**, 45 (1979).
[14]W. H. Smith and J. Gelfand, J. Quant. Spectrosc. Radiat. Transfer **24**, 15 (1980).
[15]R. G. Bray and M. J. Berry, J. Chem. Phys. **71**, 4909 (1979).
[16]G. O. Brink, J. Mol. Spectrosc. **103**, 295 (1984).
[17]M. Maeda, F. Ishitsuka, and Y. Miyazoe, Appl. Opt. **16**, 403 (1977).
[18]T. D. Harris and J. W. Mitchell, Anal. Chem. **52**, 1706 (1980).

[19]V. S. Burakov, V. N. Verenik, V. A. Malashonok, P. A. Naumenkov, S. V. Nechaev, and R. A. Puko, Zh. Prikl. Spektrosk. **37**, 197 (1982) [J. Appl. Spectrosc. **37**, 869 (1982)].
[20]V. S. Burakov, P. Ya. Misakov, P. A. Naumenkov, S. V. Nechaev, and S. N. Raikov, Zh. Prikl. Spektrosk. **28**, 413 (1978) [J. Appl. Spectrosc. **28**, 285 (1978)].
[21]G. O. Brink and S. M. Heider, Opt. Lett. **6**, 366 (1981).
[22]S. Datta, R. W. Anderson, and R. N. Zare, J. Chem. Phys. **63**, 5503 (1975).
[23]J. P. Hohimer and P. J. Hargis Jr., Anal. Chem. **51**, 930 (1979).
[24]G. H. Atkinson, A. H. Laufer, and M. J. Kurylo, J. Chem. Phys. **59**, 350 (1973).
[25]J. H. Clark, C. B. Moore, and J. P. Reilly, Int. J. Chem. Kinet. **10**, 427 (1978).
[26]J. P. Reilly, J. H. Clark, C. B. Moore, and G. C. Pimentel, J. Chem. Phys. **69**, 4381 (1978).
[27]R. J. Gill and G. H. Atkinson, Chem. Phys. Lett. **64**, 426 (1979).
[28]R. J. Thrash, H. v. Weyssenhoff, and J. S. Shirk, J. Chem. Phys. **55**, 4659 (1971).
[29]S. J. Harris, Appl. Opt. **23**, 1311 (1984).
[30]O. N. Sarkisov and E. A. Sviridenko, Zh. Prikl. Spektrosk. **35**, 775 (1981) [J. Appl. Spectrosc. **35**, 1184 (1981)].
[31]P. B. Davies and K. M. Evenson, in *Laser Spectroscopy,* proceedings of the Second International Conference on Laser Spectroscopy, Megeve, 1975, edited by S. Haroche, J. C. Pebay-Peyroula, T. W. Hänsch, and S. E. Harris (Springer, Berlin, 1975).
[32]W. H. Weber and E. A. Cohen, Opt. Lett. **8**, 488 (1983).
[33]E. F. Zalewski, R. A. Keller, and C. T. Apel, Appl. Opt. **20**, 1584 (1981).
[34]R. A. Keller, N. S. Nogar, and D. S. Bomse, Appl. Opt. **22**, 3331 (1983).
[35]G. Stella, J. Gelfand, and W. H. Smith, Chem. Phys. Lett. **39**, 146 (1976).
[36]W. Radloff and H.-H. Ritze, Appl. Phys. **20**, 247 (1979).
[37]W. Radloff and H.-H. Ritze, Opt. Commun. **35**, 203 (1980).
[38]A. A. Mak, O. A. Orlov, and V. I. Ustyugov, Kvant. Elektron. (Moscow) **9**, 2412 (1982) [Sov. J. Quantum Electron. **12**, 1574 (1982)].
[39]D. Dangoisse, A. Deldalle, and P. Glorieux, J. Chem. Phys. **69**, 5201 (1978).
[40]D. Dangoisse and P. Glorieux, Opt. Commun. **32**, 246 (1980).
[41]E. Arimondo and T. Oka, Phys. Rev. A **26**, 1494 (1982).
[42]T. Oka, Philos. Trans. R. Soc. London Ser. A **307**, 591 (1982).
[43]N. Ioli, A. Moretti, G. Moruzzi, P. Roselli, and F. Strumia, J. Mol. Spectrosc. **105**, 284 (1984).
[44]W. Werncke, J. Klein, A. Lau, K. Lenz, and G. Hunsalz, Opt. Commun. **11**, 159 (1974).
[45]V. M. Baev, W. Werncke, and E. A. Sviridenko, Kvant. Elektron. (Moscow) **2**, 856 (1975) [Sov. J. Quantum Electron. **5**, 475 (1975)].
[46]I. F. Silvera and F. Tommasini, Phys. Rev. Lett. **37**, 136 (1976).
[47]K.-M. Chen, I. C. Khoo, L. E. Steenhoek, and E. S. Yeung, Opt. Commun. **23**, 90 (1977).
[48]K.-M. Chen, L. E. Steenhoek, and E. S. Yeung, Chem. Phys. Lett. **59**, 222 (1978).
[49]J. Kowalski, R. Neumann, H. Suhr, K. Winkler, and G. zu Putlitz, Z. Phys. **A287**, 247 (1978).
[50]W. Brunner and H. Paul, Opt. Commun. **12**, 252 (1974).
[51]W. Brunner and H. Paul, Opt. Quant. Electron. **10**, 139 (1978).
[52]B. I. Stepanov, A. N. Rubinov, and M. V. Belokon, Zh. Prikl. Spektrosk. **24**, 423 (1976) [J. Appl. Spectrosc. **24**, 293 (1976)].
[53]S. J. Harris, J. Chem. Phys. **71**, 4001 (1979).
[54]S. J. Harris and A. M. Weiner, J. Chem. Phys. **74**, 3673 (1981).
[55]V. M. Baev, T. P. Belikova, E. A. Sviridenko, and A. F. Suchkov, Zh. Eksp. Teor. Fiz. **74**, 43 (1978) [Sov. Phys.—JETP **47**, 21 (1978)].
[56]T. P. Belikova, B. K. Dorofeev, E. A. Sviridenko, and A. F. Suchkov, Kvant. Elektron. (Moscow) **2**, 1325 (1975) [Sov. J. Quantum Electron. **5**, 722 (1975)].
[57]E. Arimondo, F. Casagrande, L. Lugiato, and P. Glorieux, App. Phys. **B30**, 57 (1983).
[58]S. J. Harris, Opt. Lett. **7**, 497 (1982).
[59]K. Tohma, J. Appl. Phys. **47**, 1422 (1976).
[60]Y. H. Meyer and P. Flamant, Opt. Commun. **19**, 20 (1976).
[61]V. M. Baev, T. P. Belikova, S. A. Kovalenko, E. A. Sviridenko, and A. F. Suchkov, Kvant. Elektron. (Moscow) **7**, 903 (1980) [Sov. J. Quantum Electron. **10**, 517 (1980)].
[62]V. R. Mironenko and V. I. Yudson, Opt. Commun. **34**, 397 (1980).
[63]S. A. Kovalenko, Kvant. Elektron, (Moscow) **8**, 1271 (1981) [Sov. J. Quantum Electron. **11**, 759 (1981)].
[64]H. Haken, *Laser Theory* (Springer, Berlin, 1983).
[65]M. Lax, in *Statistical Physics, Phase Transitions and Superfluidity,* proceedings of the Brandeis University Summer Institute in Theoretical Physics, Waltham, 1966, edited by M. Crétien, E. P. Gross, and S. Deser (Gordon and Breach, New York, 1968), Vol. 2.
[66]H. B. Callen and T. A. Welton, Phys. Rev. **83**, 34 (1951).
[67]V. M. Baev, G. Gaida, H. Schröder, and P. E. Toschek, Opt. Commun. **38**, 309 (1981).
[68]N. G. Basov, V. N. Morozov, and A. N. Oraewskij, Zh. Eksp. Teor. Fiz. **49**, 895 (1965) [Sov. Phys.—JETP **22**, 622 (1966)].
[69]H. K. Holt, Phys. Rev. A **11**, 625 (1975).
[70]T. D. Harris, in *Ultrasensitive Laser Spectroscopy,* edited by D. S. Kliger (Academic, New York, 1983).
[71]V. R. Mironenko and I. Pack, Kvant. Elektron. (Moscow) **5**, 2476 (1978) [Sov. J. Quantum Electron. **8**, 1394 (1978)].
[72]E. N. Antonov, A. A. Kachanov, V. R. Mironenko, and T. V. Plakhotnik, Opt. Commun. **46**, 126 (1983).
[73]C. E. Moore, M. G. J. Minnaert, and J. Houtgast, *The Solar Spectrum 2935 Å to 8770 Å,* Natl. Bur. Stand. (U.S.) Monograph No. 61 (U.S. GPO, Washington, D.C., 1966).
[74]S. Gerstenkorn (private communication).
[75]S. Gerstenkorn and P. Luc, *Atlas du Spectre d'Absorption de la Molécule d'Iode* (CNRS, Paris, 1978).
[76]F. Stoeckel, M.-A. Melieres, and M. Chenevier, J. Chem. Phys. **76**, 2191 (1982).
[77]M. Chenevier, M.-A. Melieres, and F. Stoeckel, Opt. Commun. **45**, 385 (1983).
[78]D. E. Burch and D. A. Gryvnak, Appl. Opt. **8**, 1493 (1969).
[79]P. H. Krupenie, J. Phys. Chem. Ref. Data **1**, 423 (1972).
[80]J. K. G. Watson, Can. J. Phys. **46**, 1637 (1968).
[81]E. N. Antonov, P. S. Antsyferov, A. A. Kachanov, and V. G. Koloshnikov, Opt. Commun. **41**, 131 (1982).
[82]F. J. Morgan, C. H. Dugan, and A. G. Lee, Opt. Commun. **27**, 451 (1978).