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Temporal Evolution of the Emission Spectra of Color Center Lasers*

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Abstract. The mode spectrum of a homogeneously broadened laser is mainly influenced by mode competition effects. In this paper the temporal evolution of the emission spectra of color center lasers caused by mode competition is investigated. The results are compared with a theoretical estimation.

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Multimode laser oscillators with homogeneously broadened gain profiles are more complex than single mode oscillators or multimode oscillators with inhomogeneously broadened gain profiles. This is caused by the coupling of individual modes to a common reservoir. Although the correlation between individual modes is weak, the temporal development of the laser spectrum is mainly characterized by this coupling.

From the start of laser oscillation the laser spectrum, initially given by the spontaneous emission profile, is subject to strong mode competition. The increasing enhancement of modes with higher net gain and the ensuing suppression of modes with lower gain gives rise to a reduction in spectral bandwidth. In general, however, spectral narrowing does not result in single mode oscillation. Spontaneous emission noise and spatial holeburning in case of a Fabry Perot type cavity are effects favoring mode coexistence.

The dependence of the spectral bandwidth on the generation time (the time ellapsed since the onset of laser oscillation) is essentially determined by the strength of mode coupling. Pump intensity, cavity losses, and the gain profile are the main parameters of influence.

The characteristic periods of time necessary to observe a steady state in spectral evolution, are of basic interest in laser physics and especially in the field of intracavity laser spectroscopy (ILS) [1].

ILS is widely used to detect absorption from atomic and molecular species in gases, flames, plasmas, and liquids [2–4]. The growing interest in ILS is due to its extraordinarily high sensitivity in measuring weak absorption. Due to cavity losses a well-defined spectral distribution of the emission develops during a characteristic period of time. The sensitivity of ILS increases with the effective absorption path length which is proportional to the generation time. Therefore, the specific time scales of spectral narrowing and the number of free-running modes in steady state are important in order to predict the sensitivity.

Due to the advantage of a high sensitivity over a wide spectral range, broad-band lasers with a homogeneously broadened emission spectrum are preferable for use in ILS. As a consequence, mode coupling and the spectral evolution, particularly that of the dye laser, have been investigated experimentally and theoretically by numerous authors [2, 5–7].

Whereas the dye laser covers the visible spectral range, color center lasers (CCL) are the equivalent laser systems operating in the NIR. They cover the spectral range 0.8–3.8 μm. For the understanding of the physics of the spectral narrowing process, these lasers offer a favorable situation: the F_A(II) and F_B(II) color centers both emit in the same spectral region between 2.5 μm and 2.9 μm, but they differ in the relaxation time of spontaneous emission. Since the strength of mode coupling depends on this relaxation

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time, differences in the time dependence of the emission spectra are to be expected.

Brunner and Paul presented numerical results for the spectral narrowing of the dye laser [8], which can be qualitatively compared with those of the CCL. In particular, the dependence of the spectral bandwith on the generation time was estimated by an analytical expression.

1. Spectral Evolution of a Homogeneously Broadened Laser

The temporal spectral evolution of a laser with homogeneously broadened gain band can be described by means of the rate equations. Individual modes coupled to a common inversion distribution interact via the spatial structure of the photon density. Hence, the spatial distribution of both must be considered. It is thus necessary to establish equations of motion for the spatial Fourier components of the inversion density. This has been demonstrated by Brunner and Paul [8, 9] in order to obtain a numerical calculation method for the spectral evolution. These equations can be used to derive a simple approximate formula which describes the FWHM of the laser emission spectra as a function of time.

In the following the main results of this theory with the adaptation to the individual characteristics of CCL's are given. Since the final equation is obtained with various approximations its validity range will be discussed briefly.

Following the procedure of Brunner and Paul, we will derive an expression for the time-dependent photon occupation number $n_j(t)/n_0(t)$. n_0 is the photon occupation number of the mode oscillating at the center of the fluorescence line. The subscript j denotes the spectral distance to the line center in units of the resonator mode spacing.

In contrast to the aforementioned work, we assumed a gain profile of gaussian shape, which fits better to the luminescence profile of color centers. Therefore the Einstein coefficients B_j with mode index j read

$$B_j = B_0 \exp[-\ln 2(2j\delta w/\Gamma)^2] \quad j = 0, \pm 1, \pm 2, \ldots, \quad (1)$$

where δw is the mode spacing of the resonator and Γ is the FWHM of the homogeneous linewidth. By using these coefficients the ratio of the time dependent photon occupation numbers is

$$\frac{n_j(t)}{n_0(t)} = \frac{n_j(t_0)}{n_0(t_0)}\left[\frac{n_0(t)}{n_0(t_0)}\right]^{-\ln 2(2j\delta w/\Gamma)^2} \times \exp[-\ln 2(2j\delta w/\Gamma)^2 \kappa t]. \quad (2)$$

Laser oscillation starts at time t_0. The above relation is valid for $t \gg t_0$ and $(2j\delta w/\Gamma)^2 \ll 1$. The loss rate κ is not subscribed by an index. Hence, the validity is restricted to nondispersive intracavity losses. The dominant term of (2) is the exponential one, so that the FWHM of the emission profile Δw is finally given by the following estimate

$$\Delta w = \Gamma/(2\sqrt{\kappa t}). \quad (3)$$

In fact, this formula represents an upper limit for the bandwidth, because the omitted term $[n_0(t)/n_0(t_0)]^{-\ln 2(2j\delta w/\Gamma)^2}$ gives rise to a further reduction of the linewidth. However, this term will only contribute appreciably in the early stage of the spectral evolution, when the value of j corresponding to the FWHM is large enough.

It has to be mentioned that the spontaneous emission was neglected when deriving (2) from the rate equations. Therefore in this theory the onset of laser oscillation coincides with the onset of pumping. In reality the starting point at time t_0 of the considered laser process is delayed with respect to the pump pulse.

For steady-state conditions the resulting equation is also inadequate to describe the experiment. The calculated bandwidth is too small because spontaneous emission noise and the interference term due to spatial holeburning were omitted.

2. Experimental Set-Up

The experiments were performed in a free running laser system. Frequency selection due to atmospheric absorption or interference effects within the cavity were carefully avoided to ensure that only mode coupling is responsible for the observed spectral properties and to establish a well-defined situation for the time resolved measurements of the dynamic mode behavior. Accordingly, the laser cavity was entirely evacuated. Figure 1 gives a schematic overview of the experimental setup.

It has to be pointed out that one must choose a cavity design which prevents undesired Fabry-Perot interference from parallel optical surfaces. Such interference would influence the spectral distribution of

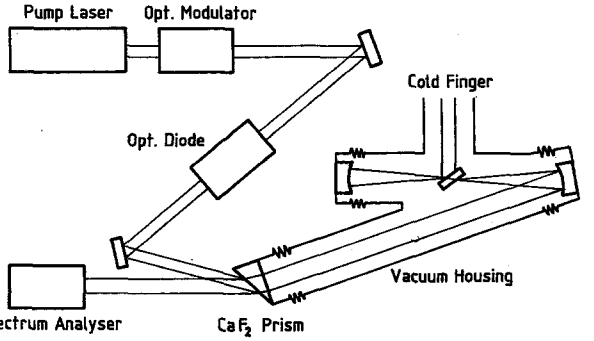

Fig. 1. Schematic overview of the experimental set-up

resonator modes. For this reason we used gold-coated spherical mirrors. With dielectric coatings reflections from the rear surfaces would appear, which give rise to interference effects. Furthermore the spectral reflectivity of gold in the NIR is much more uniform over wide ranges than that of dielectric coatings. Instead of pumping through the spherical end mirror [10] the use of gold-coated mirrors requires an unconventional pump arrangement. In our experiment the pump beam ($\lambda_p = 647$ nm) and IR-laser beam were separated by a CaF_2 prism which simultaneously served as an output coupler with 2% transmission. The dielectric coating was specially designed for a constant transmission over a sufficiently large spectral range. The folding mirror and the spherical end mirror had radii of curvature of 100 mm and 50 mm respectively.

The laser crystal was mounted on a copper cold finger at the Brewster angle and operated at liquid nitrogen temperature to ensure a high quantum efficiency. Formation of a standing pump wave was prevented by means of a Faraday isolator.

Initial laser oscillation was expected after time periods $(t-t_0) > 80$ ns (200 ns), since the color centers investigated had relaxation times of spontaneous emission in this range.

At the very beginning of laser oscillation a rather broad bandwidth must be analyzed with high temporal resolution because the spectral bandwidth is reduced rapidly at this stage. Later the speed of spectral narrowing decreases more and more; at this stage a high spectral resolution is necessary for the measurement.

We therefore used different experimental techniques for generation times shorter or longer than 500 ns. At time scales less than 500 ns we determined the FWHM of the emission with a Spex IR-grating monochromator of 300 GHz spectral resolution. The pump beam was modulated by a Pockels cell to generate pump pulses of rectangular shape with 2 ns rise and fall time. The length of the excitation pulses could be varied from a few nanoseconds to a maximum of 500 ns at 1 Hz repetition rate.

An InSb detector (with a response time of 0.8 μs and 12×10^3 V/W sensitivity) was used to measure the spectral distribution as a function of generation time. The detector was operated at LNT for a good signal to noise ratio.

Beyond 0.5 μs the mode spectrum was recorded with a piezoelectrically scanned Fabry-Perot Interferometer (FPI). The free spectral range of this optical spectrum anylzer was varied stepwise from 1200 GHz to 25 GHz so that the spectral resolution is increased with decreasing laser bandwidth. For these measurements the modulation of the pump beam was performed by a mechanical chopper. As the length of the excitation pulses could not be varied, time resolution was achieved by use of a Boxcar integrator. The excitation pulses had a length of 0.7 ms at a repetition rate of 1 kHz with rise and fall times of less than 400 ns.

While slowly scanning the FPI across the free spectral range a 50 ns observation gate was set on the detector signal at particular times $(t-t_0)$. By integration over 20 events a smooth emission spectrum was obtained.

In order to prevent intense absorption by water vapor the detection equipment was kept in an air-tight cover flooded with dry nitrogen.

3. Experimental Results

Laser experiments were performed with F_A(II) color centers in KCl : Li and F_B(II) centers in RbCl : Na. The essential laser parameters are listed in Table 1 together with the optical and spectroscopic properties of each material. The two color centers show nearly identical emission spectra, however, they differ in their fluorescence lifetimes τ_F and consequently in their emission cross sections. Hence, it is possible to investigate spectral laser behavior as a function of time for two values of τ_F.

The width's of the emission spectra of the F_A(II)-CCL and the F_B(II)-CCl are shown in Figs. 2 and 3, respectively. The measurements were taken under essentially the same conditions.

In both cases we observed the onset of laser oscillation after approximately the fluorescence life-

Table 1. Summary of relevant resonator and laser crystal data

Laser parameters		
L	Resonator length [cm]	23.7
l	Crystal length [cm]	0.3
$\delta\nu_R$	Resonator mode spacing [GHz]	0.63
$\delta\nu_H$	Holeburning mode spacing [GHz]	1.50
κ	Loss rate equivalent to 5% intracavity losses and 2% outputcoupling [1/s]	0.77×10^8
R	Pump rate equivalent to 1 W pump power at 647 nm [1/s]	3.27×10^{18}

Crystal properties			
	Host crystal	KCl : Li	RbCl : Na
	Type of color center	F_A(II)	F_B(II)
τ_F	Fluorescence lifetime at LNT [ns]	85	≈200
n	Refractive index	1.49	1.49
λ_0	Central wavelength [μm]	2.68	2.70
Γ	FWHM of spontaneous emission [1/s]	1.45×10^{13}	1.50×10^{13}
η	Quantum efficiency at LNT	0.4	≈0.4

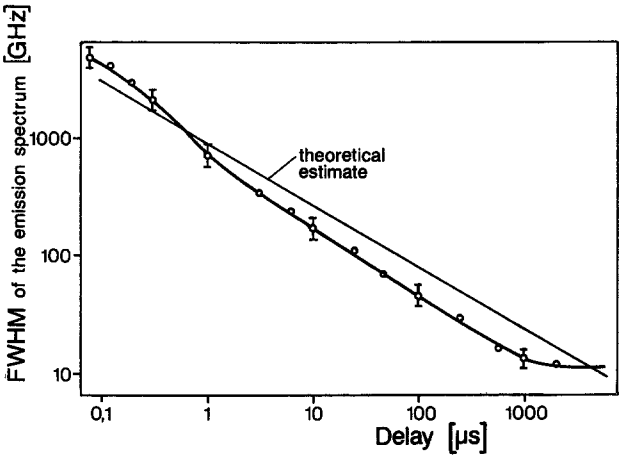

Fig. 2. Spectral band width of the F_A(II)-CCl in KCl:Li during the build-up time of laser oscillation

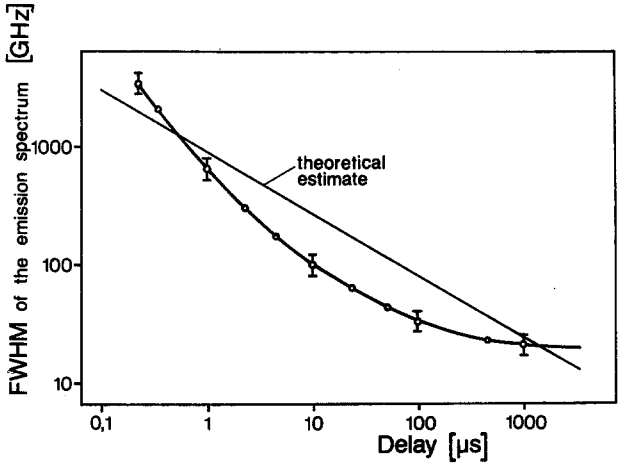

Fig. 3. Spectral band width of the F_B(II)-CCL in RbCl:Na during the build-up time of laser oscillation

time. This behavior corresponds to our expectations because the build-up-time of the laser inversion should be on the order of the fluorescence lifetime, if the pump power is about the threshold pump power.

The initial FWHM is approximately equal to the tuning range of the respective laser system. This is to be expected as well, because the initial photon density in the laser modes corresponds to the spontaneous emission spectrum.

During the first few microseconds the linewidth decreases drastically. This is due to two facts:

1. There is a large difference in gain between modes close to the line center and those closer to the margin of the laser line. This difference results in a shorter build-up-time for the central modes than for the marginal modes.

2. When the central modes have sufficient intensity, the marginal modes are completely suppressed by the central modes due to mode coupling.

After a few 100 μs the FWHM reaches a steady state with some 10 GHz. This final bandwidth is due to spontaneous emission and due to spatial holeburning which favors multimode oscillation.

However, the two color center lasers revealed differences in the rate of line narrowing and in the FWHM at the steady state. The more rapid decrease in linewidth of the F_B(II)-CCl can be explained by its larger fluorescence lifetime which results in a smaller spontaneous emission rate. Spontaneous emission, however, supports the existence of marginal modes as long as the laser intensity is considerably below the saturation intensity.

The F_B(II)-CCl has a larger steady state FWHM because its net gain is smaller (due to higher crystal losses caused by absorption and thermally induced inhomogenieties) than that of the F_A(II)-CCl. Lower gain results in lower mode intensity and a corresponding reduction of mode competition. In addition, it is possible that different inhomogeneities at the sites of color centers in F_A(II) and F_B(II) color center crystals cause different line broadening mechanismus.

During 700 μs excitation a red-shift of 300 GHz was observed. This frequency shift is supposed to originate from a temperature-induced shift of the gain profile.

The experimental data are described well by the theoretical estimate of (3) (straight line on Figs. 2 and 3), which is surprising because of the complexity of this dynamic process. Anyhow, one must bear in mind that this formula is not valid in the early stages and in the final stages of the spectral evolution as previously discussed. In between it actually gives an upper limit for the spectral bandwidth.

If the expression $[n_0(t)/n_0(t_0)]^{-\ln 2(2j\delta w/\Gamma)^2}$ in (2) is considered as well, whereby $n_0(t)$ is taken from the experimental results in Fig. 4, an additional reduction of <10 GHz in bandwidth will result. Therefore sub-

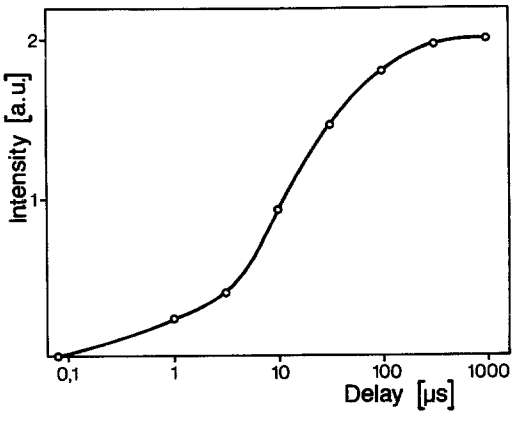

Fig. 4. Time behavior of the central mode intensity during the build-up time of laser oscillation

stantial corrections come from the neglected coupling terms of the modified rate equations.

The numerical results of these equations show a definite correlation between pump power and the rapidity of spectral narrowing. For an experimental verification we varied the pump power up to an accessible value of 4 W. However, the additional reduction was only 10–20 GHz, observed within the first 50 μs generation time. This negligible change is due to the relatively slight variation in pump power. Significant changes are expected if the pump power is varied over several decades.

4. Conclusions

We have investigated the temporal spectral evolution of mode spectra in the build-up stage of laser oscillation.

Experiments were performed on F_A(II) and F_B(II) color center lasers operating in the spectral regime around 2.7 μm. Efforts were made to exclude disturbances from atmospheric absorption and frequency selection due to Fabry-Perot interference effects.

In case of the F_A(II) color center laser the initial value of 4×10^{13} GHz spectral halfwidth decreases within 10 μs to 180 GHz, whereas a mean constant distribution of about 25 GHz is observed after times >0.5 ms. This is the bandwidth of the F_A(II) color center laser without any frequency selection.

The F_B(II) color center laser exhibits a somewhat different behavior. Due to a lower emission cross section and higher internal crystal losses the bandwidth diminishes from 3×10^{12} Hz to 180 GHz in just 5 μs. The whole dynamic process takes only 0.2 ms time and leads to a spectral bandwidth of about 45 GHz.

These experimental results agree with the predictions of an analytic approach.

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