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C. N. Man, P. Cerez, A. Brillet, F. Hartmann

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A FREQUENCY STABILIZED CW DYE LASER FOR SPECTROSCOPIC AND METROLOGICAL APPLICATIONS (*)

C. N. MAN, P. CEREZ, A. BRILLET

Laboratoire de l'Horloge Atomique (**), Université Paris-Sud, 91405 Orsay Cedex, France

and

F. HARTMANN

Laboratoire de Spectrométrie Physique (***)
 Université Scientifique et Médicale de Grenoble, 38041 Grenoble Cedex, France

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Résumé. — Nous avons construit un laser à colorant continu en vue d'applications spectroscopiques et métrologiques. L'emploi de plusieurs boucles d'asservissement permet d'obtenir une stabilité élevée. La largeur de raie observée est de 5×10^{-10} (250 kHz), tandis que la stabilité de fréquence à long terme atteint $1,5 \times 10^{-12}$ pour des temps d'observation supérieurs à 100 s. La stabilité d'amplitude est meilleure que 10^{-4} . A titre d'application, nous avons étudié par absorption saturée la structure hyperfine de deux raies de l'iode et déterminé l'élargissement des composantes hyperfines avec la pression d'iode.

Abstract. — A cw dye laser for spectroscopic and metrological applications has been constructed. A high stability is obtained through the use of several servo loops. The observed relative linewidth is 5 parts in 10^{10} (250 kHz FWHM) whereas the long term frequency stability reaches 1.5 parts in 10^{12} for observation times longer than 100 s. Amplitude stability is better than one part in 10^4 . As an application, the hyperfine structure of two iodine lines has been studied by saturated absorption spectroscopy and the variation of the linewidth of the hyperfine components with iodine pressure has been determined.

Since the first successful operation of a cw dye laser in 1970 [1], this device has been considered to be a very promising tunable and monochromatic source for spectroscopic and even metrological purposes. However, the subsequent huge amount of work performed in this direction in many laboratories has revealed the numerous difficulties to be solved [2]. Sources suitable for sub-Doppler atomic spectroscopy, where one is dealing with linewidths of some 10 MHz, are now readily obtained but the much narrower sources needed for molecular spectroscopy and metrological applications are only beginning to become available [3, 4]. In this paper we describe a dye laser to which several servo loops confer a linewidth of 5 parts in 10^{10} (250 kHz FWHM), a frequency stability of 1.5 part in 10^{12} over 100 s and an amplitude stability better than one part in 10^4 . The device has been tested with iodine saturated absorption lines.

The basic structure chosen for our laser is the three mirror, jet stream configuration of a commercial unit (Spectra Physics 580 A). The 38 cm cavity length corresponds to a frequency separation of 390 MHz between adjacent modes. Two mechanically adjustable thin Fabry-Perot filters define the oscillation domain within the gain curve of the dye (Rhodamine 6 G). Single frequency operation is provided by a third, piezoelectrically tunable etalon whose centre frequency is brought into coincidence with a cavity mode. This coincidence is maintained during frequency scans by a servo loop which also takes care of etalon thermal drifts, so that no mode hops occur during time intervals as long as several hours. The total scan width reaches 5 GHz (0.16 cm^{-1}) and is limited by the sensitivity of the pzt which drives the cavity output mirror.

The spectrum of the frequency fluctuations of such a laser turns out to be essentially conditioned by the perturbations occurring in the jet stream region. The low frequency fluctuations (0-2 kHz) can be ascribed to thermal drifts, drafts, air transmitted noise and

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(**) Equipe propre du C.N.R.S.

(***) Associé au C.N.R.S.

mainly jet stream thickness fluctuations due to vibrations in the dye circulator. They result in a frequency jitter of a few megahertz and a drift of 1 MHz/min., provided elementary precautions are taken for vibration and acoustical isolation. In an intermediate frequency range (tens of kilohertz) the mechanical resonances of the nozzle can induce a line broadening of about 150 kHz. Finally, we could observe a high frequency noise (400 kHz-1 MHz) which we think to be due to surface waves in the dye stream.

In order to obtain a high short and long term frequency stability together with some tunability of the laser, it is necessary to use three successive servo loops [3, 4, 5] whose principle is depicted in figure 1. The first, acting on the cavity output mirror pzt, locks the dye laser frequency to the side of the transmission curve of an external reference cavity. One mirror of this cavity is also provided with a pzt, allowing it to be locked in turn to the emission line of a He-Ne transfer oscillator. The joint action of the two above mentioned servo loops results in transferring to the dye laser the very high short term frequency stability of the transfer oscillator. Finally, the remaining long term drifts are eliminated by a third servo loop which locks, with a variable frequency interval, the frequency of the transfer oscillator to the output of an I₂ — stabilized He-Ne standard via a frequency offset locking technique [3-6].

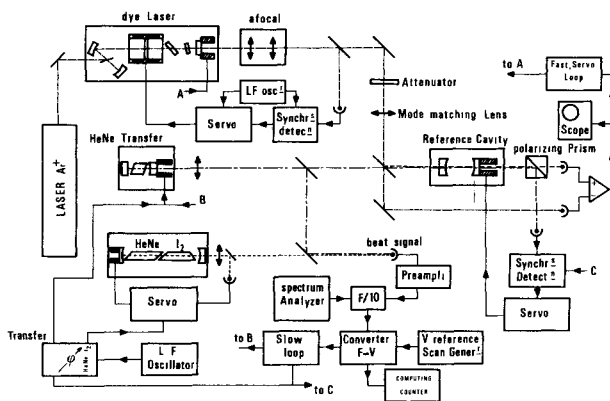


FIG. 1. — Block diagram of the dye laser frequency stabilization system.

The best way to evaluate the frequency stability of such a source would be to study the beat note between it and a similar unit or, even better, an optical frequency standard. A more accessible and nevertheless reasonably correct method is to make use of a high finesse cavity as a frequency discriminator. The resulting amplitude variations can be translated back into the frequency domain by use of a voltage to frequency converter and subjected to statistical analysis. Figure 2 shows the result of such a study for the free-running and servoed laser. As mentioned before, frequency stability in this last case is essentially equal to that of the transfer oscillator for intermediate observation times and to that of the iodine-stabilized standard

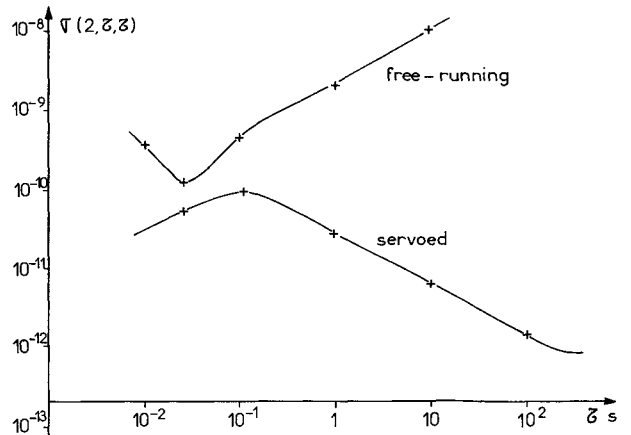


FIG. 2. — Fractional frequency stability σ (Allan variance) of the free-running and servoed laser as a function of observation time τ .

for longer times ($\tau > 100$ s), reaching then a value of 1.5 part in 10^{12} .

The short term frequency stability is governed by the bandwidth of the first servo loop. Up to now we have used a relatively slow loop (5 kHz BW) which is very reliable but does not allow the linewidth to be reduced below 5 parts in 10^{10} (250 kHz FWHM). We have also tested a much faster loop (300 kHz BW) which narrows the linewidth down to 70 kHz but unlocks in times ranging from a few seconds to some tens of minutes depending on the number of bubbles present in the dye stream.

For spectroscopic studies of weak and narrow transitions, a good amplitude stability is also required. An electrooptic amplitude corrector [7, 3] is used for this purpose, giving an amplitude stability exceeding one part in 10^4 .

In order to test the spectroscopic potentialities of the device, we have set up a saturated absorption experiment on iodine vapour in a ring configuration. Enhancement of the signal to noise ratio was obtained through a modulation of the saturating beam together with observation of the difference between two probe beams [8]. Optical feedback was nearly suppressed by using crossed polarizations for the probe and saturating beams. It was thus possible to align them perfectly and minimize the residual Doppler broadening, while increasing the interaction length and therefore the signal to noise ratio. The interference fringes appearing on the saturated absorption signal due to the remaining feedback could be eliminated by modulating the phase of the retroreflected light [5, 9].

Figure 3 shows the r hyperfine component [10] of the P(73)14-1 and the k, l, m components of the R(69)16-2 lines at $\lambda = 586.82$ nm for various iodine pressures and saturation parameters. The total frequency scan of 50 MHz for each curve was performed in 100 kHz steps of 0.3 s duration. A whole set of such curves was recorded, allowing the determination of saturation parameters and pressure broadening. Figure 4 shows the pressure variation of the hyperfine

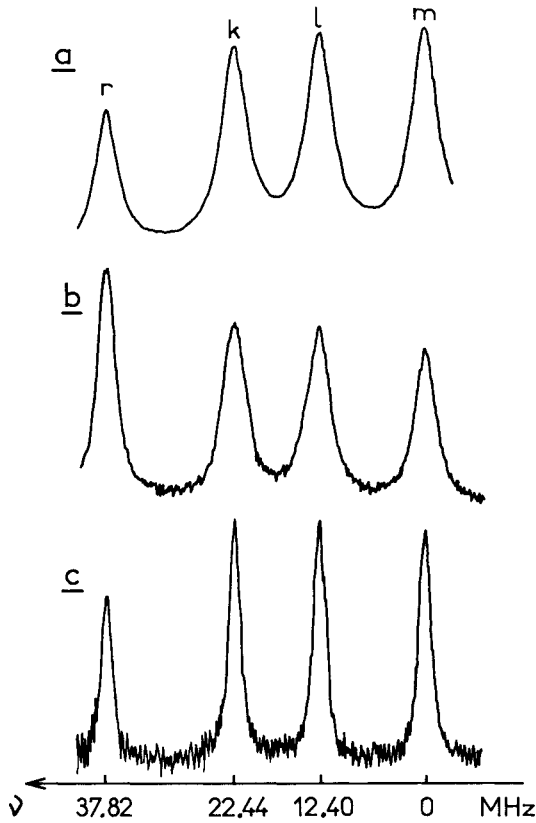


FIG. 3. — High resolution spectrum of the r component of the P(73)14-1 and the k , l , m components of the R(69)16-2 lines of $^{127}\text{I}_2$. a) iodine pressure $p = 140$ millitorr, saturating power $P_s = 7.6$ mW. b) $p = 8.6$ millitorr, $P_s = 7.6$ mW. c) $p = 8.6$ millitorr, $P_s = 0.1$ mW.

components linewidth extrapolated to zero saturating power. The change in slope occurring near 50 millitorr, which can also be observed on the variation of the shift of other lines with pressure [11], is related to

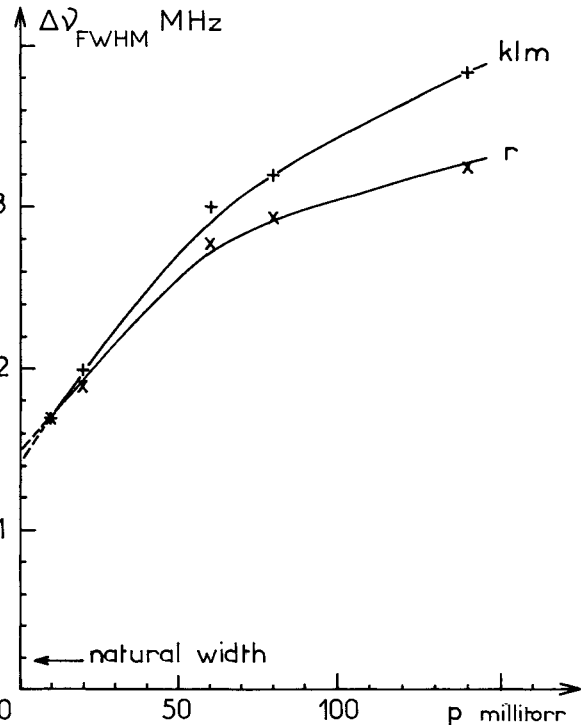


FIG. 4. — Linewidth of the hyperfine components shown in figure 3 as a function of iodine pressure at zero saturating power.

the influence of velocity changing collisions [12, 13]. The extrapolation to zero pressure shows an instrumental broadening of 1.2 MHz due to residual Doppler width, transit time broadening and laser frequency jitter.

With continuing improvements, the device thus appears as a very promising tool for high resolution spectroscopy and metrology : investigations in these directions are now in progress.

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