Frequency stabilization of a cw dye laser

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A cw dye laser system frequency stabilized to a high-finesse optical reference cavity is described. Laser frequency is servo controlled to the cavity resonance with residual fluctuations less than 50 kHz for short times (20 μ sec) and 100 Hz for long times (10 sec). Drift in absolute laser frequency of about 1.5 MHz/min is observed due to drift of the unstabilized reference cavity. A saturated absorption spectrum of I_2 obtained with this system is shown.

A remarkable improvement in the frequency stability of a single-mode cw dye laser has been obtained by using a wide-band (0-100 kHz) servo system to lock the dye laser frequency to a transmission fringe of a highfinesse optical cavity. The frequency stability for the free-running cw dye laser was typically on the order of \pm 50 MHz for averaging times of 1 sec. By applying corrections to the optical length of the dye laser cavity with two piezoelectrically driven mirrors, frequency variations of the laser were reduced to drifts on the order of 1-2 MHz/min (corresponding to controllable drifts of optical cavity temperature and possibly of optical coupling parameters). The frequency has been made to track these drifts in a single cavity with errors less than 50 kHz rms for short times (20 μ sec) and less than 100 Hz for long times (10 sec). This frequency stability has also been shown to be sufficient to do saturation spectroscopy of optical transitions in molecular gases such as iodine.

The dye laser used in these experiments is similar to the three-mirror astigmatically compensated dye laser of Dienes, Ippen, and Shank.¹ Single-mode operation was obtained by using two intracavity Brewster-angle prisms, two uncoated solid etalons with optical thicknesses of 3 and 19 mm, and a mode-limiting aperture. It was also found that using a very-short-focal-length (R = 1 cm) high-reflectivity mirror next to the Brewster-angle dye cell greatly improved the stability of the single-mode operation,² perhaps owing to spatial saturation effects in the dye cell (1 mm thick). By simultaneously tilting both etalons and displacing the flat output mirror, continuous tuning of the single mode could be accomplished over intervals of 1.5 GHz. Typical single-mode output power was 5 mW (with 1 W argon pump power) with approximately 10% amplitude noise (0 to ~100 kHz). Continuous free-running operation in a given longitudinal ($c/2L \approx 120$ MHz) mode was limited to about 1 min. Single-mode operation was obtainable throughout the high-gain region for the rhodamine 6G dye (~5800-6200 Å).

Due to the relatively poor free-running stability, an optical frequency discriminator with a high signal-tonoise ratio over very short times, such as a highfinesse optical cavity, is needed for servo controlling the frequency. In addition to a high short-term signalto-noise ratio, an optical cavity has the added advantage of being tunable, thus providing the possibility of repeatable controlled-frequency scanning. However, owing to thermal drifts and acoustic disturbances of an optical cavity, the long-term frequency stability of such a frequency discriminator is limited. Thus it will ultimately be interesting to stabilize this reference cavity itself by using frequency offset locking³ from a suitable saturated-absorption-stabilized laser such as the NeHe/CH₄ system at 3.39 μ m.⁴ At present our frequency discriminator is a 17.8-cm Invar-spaced cavity which is enclosed in a sealed tank and optically isolated from the laser with a $\lambda/4$ plate and linear polarizer. The cavity has a finesse of 400 (2.1-MHz FWHM fringes) and one piezoelectrically controlled mirror. The servo lock is made by using the side of a transmission fringe with the zero for the error signal located approximately halfway up the fringe. In order to avoid mapping intensity fluctuations into the frequency domain, a fast differencing technique using a separate reference channel is employed. An alternative approach of servo controlling the laser intensity has also been demonstrated, using a linear polarizer and an electrooptic crystal (KDP).

For servo control, a portion of the dye laser output is mode matched into the Invar frequency discriminator, and an error signal is formed by taking the difference between the reference and transmission fringe channels. This error is then passed through a variable-gain integrator and a 3-dB/octave active filter into a highvoltage amplifier. The resulting correction signal is



FIG. 1. Allan variances, $\sigma[(\Delta \nu)_{av}/\nu]$, of laser frequency; τ is the integration time. ×, variance for laser frequency locked to Invar cavity transmission fringe; \bigcirc , variance for light transmitted by auxiliary cavity with laser frequency locked to Invar cavity. Divergence of the two cases demonstrates relative instabilities of the two discriminator cavities.



FIG. 2. Saturated absorption signal (first derivative) for an iodine triplet near 5975 Å. Dither amplitude, 5 MHz. Intensity stablizer suppressed dither's AM noise by 66 dB.

then divided into low- and high-frequency components by a crossover network. The low-frequency corrections (<1 kHz) are applied to a long-excursion (10 μ m) piezoelectric driver attached to the output coupling mirror (resonant frequency of ~18 kHz), while the high-frequency ones are applied at the opposite end of the dye laser cavity to a very fast piezoelectrically driven mirror (resonant frequency ~200 kHz). Useful servo gain was evident beyond 50 kHz. It should be possible to obtain a servo bandwidth of several MHz by using an electro-optic crystal inside the cavity for phase control.

The frequency stability of the servo controlled laser is best represented by an Allan variance plot (a plot of σ , the normalized averaged first differences $\Delta \nu / \nu$ of the laser's absolute frequency, versus the averaging time τ). Figure 1 shows the results of two separate tests of the frequency stability. The crosses are experimental points obtained by measuring the error signal from the Invar frequency discriminator cavity. The voltage-tofrequency conversion is done by knowing the fringe width (2.1 MHz FWHM) and fringe height (~10 V). These points represent the ability of the servo system to make the laser frequency track the Invar cavity system (including its small instability and drift).

The servo gain increases at 9 dB/octave for frequencies below the unity gain frequency of approximately 50 kHz, which for a white-noise spectrum would result in a σ - τ curve falling at 3 dB/octave ($\tau^{-1/2}$). However, the observed frequency variance increases to a soft maximum near 0.1 msec, before breaking into a regime near τ^{-1} . This effect is consistent with the observed weak spectral noise peak at ~2 kHz, which is thought to be due to dye flow anomalies.

The circles in Fig. 1 are the σ - τ points obtained by measuring the frequency deviation of the locked laser relative to a transmission fringe of an auxiliary opti-

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cally isolated 30-cm Cervit-spaced test cavity. The rapid divergence of the two σ - τ plots for averaging times greater than 1 msec arises from relative thermal and acoustic instabilities of the two optical cavities. The correlation of the two curves for short averaging times shows that the relative stability of the two cavities for short times is comparable to or better than the frequency stability of the dye laser.

A qualitative measure of the absolute stability of the dye laser plus Invar frequency discriminator is illustrated in Fig. 2. Here the saturated absorption signal (first derivative) from an iodine hyperfine triplet is displayed as a function of voltage applied to the discriminator cavity's piezoelectric mirror driver. These peaks are typical of the many iodine multiplets we have observed throughout the rhodamine 6G region. With an ~ 5 -MHz dither, the narrowest peak is approximately 7 MHz FWHM. The scan rate was approximately 1 MHz/sec (100-sec sweep). The spectrum was found to be degraded because of drifts in the discriminator cavity after averaging for approximately 3 or 4 min, which thus indicates an absolute frequency drift of about 1.5 MHz/min. This drift could be greatly reduced by thermal controlling the discriminator cavity and could be essentially eliminated by locking the cavity length to a saturated absorption resonance through variable frequency offset locking techniques.³

A number of problems were encountered while stabilizing the cw dye laser. It was found that the dye laser performance was a very sensitive function of the angular stability of the pump laser, as well as the curvature and mode structure of the argon pump. It was also necessary to gang the mode-suppressing etalons to the output frequency in order to obtain continuous singlemode tuning over ranges larger than the free spectral range of the dye cavity. The servo system also provided a number of problems. The high-frequency servo system needed to stabilize a cw dye laser puts rather large current requirements upon the high-voltage amplifiers. The crossover network between the low- and high-frequency piezoelectric elements requires precise tuning in order to avoid the mechanical resonances of the slow element and to simultaneously compensate for the difference in gain and capacitance of the two elements.

There are many possible applications for a frequencystabilized cw dye laser. In ordinary spectroscopy such stability is necessary for numerous experiments including selective resonant excitation and radiation redistribution studies. Suitable methods of heterodyne spectroscopy should offer the possibility of doing a number of detailed line profile analyses of interesting laboratory — and possibly astrophysical — sources. Among the nonlinear spectroscopic techniques, saturation spectroscopy will probably benefit most from the high stability and wide tunability of this laser, as it will be possible to probe under the Doppler broadening of a large number of molecular and atomic systems. With modest further improvements it should be possible to redetermine with significantly improved accuracy such fundamental constants as the Rydberg.

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