Spectrally narrow pulsed dye laser without beam expander

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We have developed a simplified version of the side-pumped pulsed dye laser which has a spectral halfwidth of 1.25 GHz and a peak power of 10 kW at 600 nm. The basic laser consists of only four components (output mirror, dye cell, diffraction grating, and tuning mirror) and is exceptionally easy to align. Since the beam expander has been eliminated, the laser cavity can be made quite compact. Under the condition of reduced gain, the laser has been operated in a single mode.

The pulsed tunable dye laser has had a profound impact upon optical spectroscopy. Even relatively weak optical transitions in atoms and molecules are easily and selectively excited by such lasers because of their large spectral power density which can be readily tuned over a region extending from the near ir to the near uv. These dye lasers are typically pumped by high peak power lasers with several nsec pulse duration such as N_2 lasers and frequency multiplied ruby or Nd:YAG lasers.

Pulsed dye lasers are often modeled after the design of Hansch[1] which is characterized by a spectral width (exclusive of intracavity etalons) that is determined by diffraction and the telescope–grating combination. From Eq. (5) of Ref. 1, if w_2 is taken to be the radius of the expanded beam, we find

$$(\Delta\lambda)/\lambda = \lambda/(\pi l \sin\theta), \qquad (1)$$

where $\Delta\lambda$ is the halfwidth of the spectral distribution of the output light at wavelength λ, l is the width of the illuminated part of the grating, and θ is the angle between the grating normal and incident beam. For typical parameters, $l \sim 3$ cm, $\sin\theta \sim 0.5$, and $\lambda \sim 5000$ Å, we find that $\Delta\lambda \sim 0.05$ Å or $\Delta(1/\lambda) \sim 0.2$ cm^{-1}. Lawler et al.[2] have analyzed this laser configuration using the spectrometer model and obtain an expression similar to Eq. (1) for the bandwidth.

To reduce alignment problems and cost, several authors[3-6] have experimented with prism beam expanders, which expand the beam in one dimension only. Hanna

et al.[5] find that the resolution of a dye laser using a prism expander in place of the telescope is also given by Eq. (1). Note that for all these lasers the resolution depends on the illuminated width of the grating (perpendicular to the grooves), and not on the height, diffraction order, or groove spacing.

We have developed an improved pulsed dye laser oscillator which does not need a device to expand the beam, yet it still provides a spectrally narrow high-power output. In our design the diffraction grating is filled with the incident unexpanded dye laser beam by using it at a grazing angle as shown in Fig. 1. The diffracted beam at angle ϕ is returned to the grating by a mirror. The grating diffracts this reflected beam, reduced to its original size, back toward the optically pumped region of the dye for further amplification. As in the case of other dye lasers, the diffraction grating disperses the beam so that only a narrow spectral component is amplified.

Observe that the grating is not used at the Littrow angle, and the laser wavelength is changed by rotation of the tuning mirror.[7] Note also that the beam is not expanded if ϕ is close to 90°. Design advantages of this laser include:

(1) The expensive high-quality achromatic telescope or prism expander is eliminated.

(2) Alignment is simple. Careful adjustment and focusing of the beam expander are not necessary.

(3) Elimination of the beam expander reduces the number of surfaces in the optical cavity. The result is less loss and fewer reflections.

(4) The large grating can be replaced with a less expensive thin strip that is only a few mm high.

(5) The laser can be made extremely compact so that short duration pump light can be used more effectively.

(6) A pressure scanned[8] system can be constructed with the tuning mirror mounted external to the pressure cell thereby providing greater flexibility.
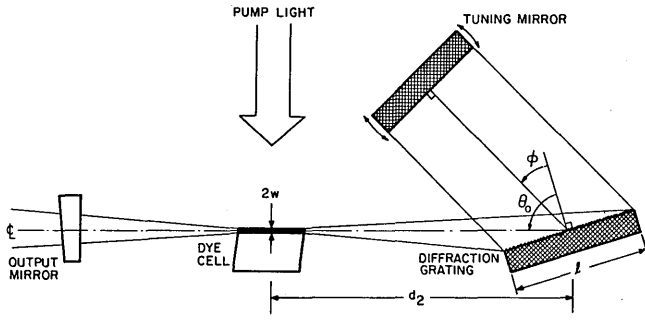
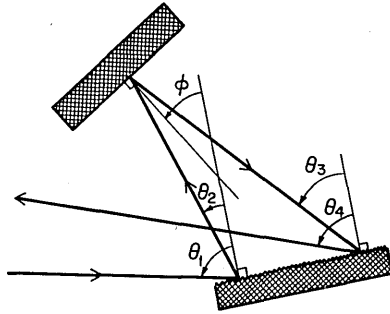Fig. 1. Schematic diagram of basic grazing incidence dye laser.

Fig. 2. Grating and tuning mirror arrangement illustrating angles used in the analysis.

We will now show that a single-pass analysis of this grating–mirror arrangement (Fig. 2) results in an expression similar to Eq. (1). The basic grating equations are

$$m\lambda = x(\sin\theta_1 + \sin\theta_2), \qquad (2a)$$

$$m'\lambda = x(\sin\theta_3 + \sin\theta_4), \qquad (2b)$$

where

$$\theta_2 + \theta_3 = 2\phi, \qquad (3)$$

m and m' are the diffraction orders, and x is the groove spacing. The laser wavelength, λ_{laser}, is defined by the above equations for the case where $\theta_1 = \theta_4 = \theta_0$. Choosing the most efficient diffraction order for both grating reflections requires $m = m'$, and therefore $\theta_2 = \theta_3 = \phi$, so that

$$\lambda_{laser} = (x/m)(\sin\theta_0 + \sin\phi). \qquad (4)$$

The spectral bandwidth of the laser $\Delta\lambda$ is determined by the grating acceptance angle (input f-number) and the ratio of the effective spectrometer length to the width of the active region (exit slit width). Thus the bandwidth of this system is given by

$$\Delta\lambda = [(\Delta\lambda_{input\ angle})^2 + (\Delta\lambda_{exit\ angle})^2]^{1/2}$$

$$= \left[\left(\frac{\partial\lambda}{\partial\theta_1}\Delta\theta_1\right)^2 + \left(\frac{\partial\lambda}{\partial\theta_4}\Delta\theta_4\right)^2\right]^{1/2}\bigg|_{\theta_1=\theta_4=\theta_0}, \qquad (5)$$

assuming a Gaussian beam profile. In the case when the beam spread between the grating and tuning mirror is negligible we obtain $\Delta\theta_1 = l\cos\theta_0/2d_2$ and $\Delta\theta_4 = w/d_2$, where w is the beam waist of the active dye region and d_2 is the separation between the dye cell and grating. To calculate $(\partial\lambda)/(\partial\theta_1)$ for fixed θ_4, we differentiate Eqs. (2) with respect to λ and rearrange terms to obtain

$$\frac{\partial\lambda}{\partial\theta_1} = \left(\frac{\lambda\cos\theta_1}{\sin\theta_3 + \sin\theta_4}\right)\left(\frac{\cos\theta_3}{\cos\theta_2 + \cos\theta_3}\right). \qquad (6)$$

In a similar manner we find

$$\frac{\partial\lambda}{\partial\theta_4} = \left(\frac{\lambda\cos\theta_4}{\sin\theta_1 + \sin\theta_2}\right)\left(\frac{\cos\theta_2}{\cos\theta_2 + \cos\theta_3}\right). \qquad (7)$$

Substitution of Eqs. (6) and (7) in the expression for the laser linewidth, Eq. (5), gives

$$\Delta\lambda = \frac{\lambda\cos\theta_0}{2(\sin\theta_0 + \sin\phi)}\left[\left(\frac{l\cos\theta_0}{2d_2}\right)^2 + \left(\frac{w}{d_2}\right)^2\right]^{1/2}. \qquad (8)$$

First we consider the situation where d_2 is greater than the Rayleigh length,[9] $L_R \equiv \pi w^2/\lambda$, in which case the beam diverges due to diffraction with half angle $\Delta\theta_{diff}$ (Ref. 9) so that

$$(l\cos\theta_0)/(2d_2) = \Delta\theta_{diff} \equiv \lambda/(\pi w). \qquad (9)$$

We combine Eqs. (8) and (9) to find

$$\Delta\lambda_{d_2>L_R} = \frac{\lambda^2}{\pi l(\sin\theta_0 + \sin\phi)}\left[\left(\frac{d_2}{L_R}\right)^2 + 1\right]^{1/2}. \qquad (10)$$

Next we consider the case where $d_2 < L_R$ (i.e., $(l/2)\cos\theta_0 = w$) and find that Eq. (8) becomes

$$\Delta\lambda_{d_2<L_R} = \frac{\lambda^2}{\pi l(\sin\theta_0 + \sin\phi)}\left(\frac{\sqrt{2}L_R}{d_2}\right). \qquad (11)$$

Observe that Eq. (10) and Eq. (11) simultaneously minimize $\Delta\lambda$ near the boundaries of their valid regions, that is, when $d_2 \sim L_R$. We then find

$$\frac{\Delta\lambda}{\lambda} \approx \frac{\sqrt{2}\lambda}{\pi l(\sin\theta_0 + \sin\phi)}. \qquad (12)$$

This result is essentially the same as that given in Eq. (1). Note that for conventional spectrometers an analogous condition occurs, namely, that the optimum resolution is obtained when the principal diffraction lobe just fills the grating. In our laser the condition $d_2 = L_R$ is easily met, for example, if $w = 0.1$ mm and $\lambda = 600$ nm, $L_R = 50$ mm.

Our laser design is shown schematically in Fig. 1. A high quality dye cell with tilted faces (e.g., Molectron DL051) is illuminated from the side by a high power pump laser, which is focused to a narrow strip by a 10-cm F.L. cylinder lens. We use a frequency doubled Nd:YAG laser beam (peak power = 1.7 mJ/7 nsec = 250 kW, repetition rate = 10 Hz, λ_{pump} = 532 nm) for pumping an unstirred cell of 2×10^{-3} M R6G dye, or a dye mixture of R6G + Cresyl Violet Perchlorate. (We have also pumped a Coumarin 102 dye laser using a N_2 pump laser.)

A wedged 4% reflecting output mirror forms one end of the laser cavity and also provides output coupling. Alignment of this element is important, and so it is mounted in a quality mirror mount with 1-sec of arc
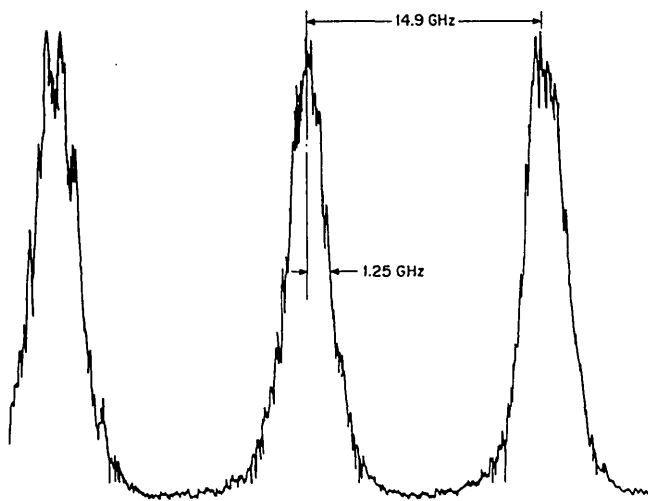
Fig. 3. Spectral analysis of laser output using scanning Fabry-Perot etalon.

was made using a Burleigh Instruments Model RC-40 scanning Fabry-Perot interferometer with a 0.5-cm^{-1} free spectral range and a finesse of 40. The sweep time was 200 sec. The observed linewidth is somewhat smaller than the theoretical one pass estimate for this grating arrangement given by Eq. (12). This is presumably due to multipass effects. Since the efficiency of the holographic grating is much greater for polarization perpendicular to the grooves, it is not surprising that the laser output is highly polarized along the horizontal axis.

When the beam is analyzed by a 0.25-cm^{-1} free-spectral-range Fabry-Perot etalon, a modal structure is evident. (The modes are not resolved in Fig. 3.) A given mode has an observed spectral halfwidth of 150 MHz (limited by the finesse of the analyzing etalon) and is separated from adjacent modes by about 750 MHz. This separation corresponds roughly to $c/2L$, where the centerline length L in this case is 20 cm. We have operated the laser in a single-cavity mode by shortening the cavity and by decreasing the R6G dye concentration to 6×10^{-4} M at the expense of power and amplitude stability. In single-mode operation the laser wavelength is relatively stable from shot to shot and does not jitter more than 300 MHz. This narrow oscillator pulse could be amplified by another dye cell amplifier.[10]

Tuning of our laser is controlled by the rotation of an optical element, and so it can be smoothly scanned over the entire lasing region of the dye. To illustrate tunability we have mechanically scanned the laser over a 60-cm^{-1} interval and plotted the transmission through a 1.15-cm^{-1} free-spectral-range Fabry-Perot etalon as shown in Fig. 4. It is not necessary to make any adjustment to the alignment during scanning. After the
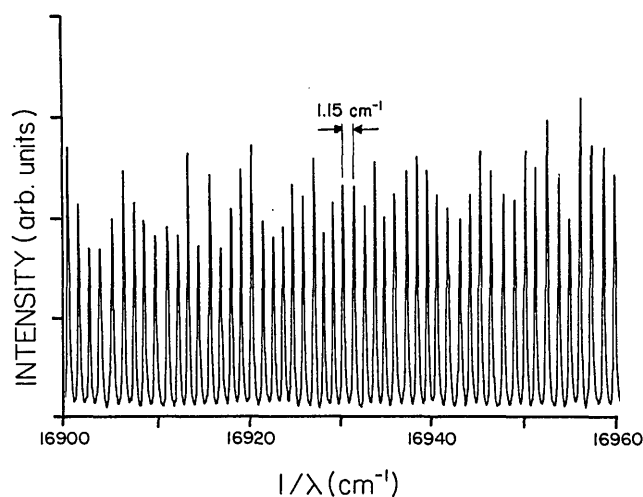
resolution. The distance between dye cell and output mirror is not especially important. For the most efficient operation, however, the mirror is kept relatively close to the cell (i.e., 1–5 cm).

In our original prototype a 50-mm wide, 600-l/mm ruled diffraction grating (Bausch & Lomb, 35.63-05-460, 54°6' blaze) was used in high order. For narrowband operation, however, the feedback efficiency for this grating was rather low. After trying several other gratings we settled on a 50-mm wide, 1800-l/mm holographic grating (PTR Optics, TF-26) which at $\theta_0 = 89.2°$ gave a net one pass efficiency of 5% ($\lambda = 632.8$ nm in first order). Since the grating is used twice per pass, the net efficiency is roughly the square of the grating efficiency, and consequently a proper choice of grating is essential. At present, we are searching for more efficient gratings. Observe that in selecting a grating for this laser, there are other considerations besides feedback efficiency. For example, it is important to avoid gratings that feed back any light toward the cell (i.e., independent of the tuning mirror) within the gain profile of the dye (Littrow case). Also note that there is a potential problem of multiple reflections between the grating and tuning mirror which can be eliminated by making certain that the rulings are not exactly perpendicular to the laser's optical axis (i.e., the grating is slightly rotated about its normal).

The tuning mirror is fully reflecting (aluminum coated) and as wide as the entire diffracted order which is reflected back onto the grating (50 mm). This mirror is mounted in a precision rotatable mount since it is the tuning element of the laser.

Once aligned (see alignment procedure in Appendix) our test laser was characterized by a spectral halfwidth-at-half-maximum of 1.25 GHz = 0.04 cm^{-1} = 0.013 Å at 600 nm (see Fig. 3). The power output was 10 kW in a 6-nsec pulse. The linewidth measurement

Fig. 4. The transmission through a fixed Fabry-Perot etalon (1.15-cm^{-1} free spectral range) as the laser is swept over a 60-cm^{-1} interval. The sole scanning element is the tuning mirror which in this case is rotated about 3 mrad. The sweep time is 10 min.

laser is aligned it is quite stable and does not require further adjustments over a period of days or longer. The initial alignment of this laser is unusually simple and relatively insensitive to pump laser focus and grating position. The alignment is, however, quite sensitive to the path through the dye cell as determined by the output mirror (see Appendix).

There are many possible variations to this grazing incidence pulsed dye laser design which we have tried that could improve its performance and versatility depending upon the specific application:

(1) A curved concave cylindrical mirror could be used to replace the flat turning mirror in long cavities to prevent losses due to vertical beam divergence.

(2) A Littrow mounted grating could be used in place of the tuning mirror to increase the dispersion and consequently reduce the laser linewidth.

(3) Intracavity etalons may be inserted to select particular modes of short cavities or to passively narrow output from long cavities.

(4) A totally reflecting mirror can replace the output mirror in which case the 0th order grating reflection is used for output coupling.

Note added in proof: It has recently come to our attention that I. Shoshan, N. Danon, and U. Oppenheim of the Technion-Israel Institute of Technology have developed a similar laser. A description of their work appears in J. Appl. Phys. 48, 4495 (1977).

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Appendix

In aligning this or any laser it is helpful to have a convenient way of estimating the laser performance. The method that we used was to observe visually the fringe pattern when laser light, which is diffusely reflected from a white card, is viewed through a Fabry-Perot etalon mounted on the end of a telescope. The etalon plate has a free-spectral-range of 1.15 cm^{-1} and a finesse of 20 (Molectron DL026C). The telescope is five power and focused at infinity.

To set up the laser, first obtain the one pass spot caused by the feedback from the output mirror. Adjust the mirror vertically to center the spot on the region of spontaneous emission and adjust the mirror horizontally so that the spot is as close as possible to the illuminated face of the dye cell. Next, slide the grating into the beam at grazing incidence so that most of the spot is intercepted. (One can see how much of the grating is being used by observing the 0th order reflected beam.) Find the strongest diffraction order and reflect it back to the grating with the tuning mirror. Adjust the mirror vertically to improve the output. Finally, observe the laser output with the Fabry-Perot etalon and adjust the output mirror horizontally until a single narrow line is obtained.

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