

# Grating cavity dual wavelength dye laser

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**Abstract:** We report simultaneous dual wavelength dye laser emission using Littman-Metcalf and Littrow cavity configurations with minimum cavity elements. Dual wavelength operation is obtained by laser operation in two optical paths inside the cavity, one of which uses reflection in the circulating dye cell. Styryl 14 laser dye operating in the 910 nm to 960 nm was used in a 15%:85% PC/EG solvent green pumped with a Q-switched doubled Nd<sup>3+</sup>:YAG laser.

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**OCIS codes:** (050.1950) Diffraction gratings; (140.2050) Dye lasers; (140.4780) Optical resonators.

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## 1. Introduction

Recently, laser systems emitting at two wavelengths has drawn the attention for different applications [1–4], including the implementation of such systems in terahertz sources. Terahertz waves, also known as far infrared, submillimeter waves, or T-rays, cover the electromagnetic spectrum between 0.1 and 10 THz (30  $\mu$ m to 3 mm). Until recently, the study in this range of the electromagnetic spectrum has been limited due to the lack of manmade sources. There are a few ways to obtain terahertz sources [5]: ultrashort laser pulses pumping semiconductor antennas [6], semiconductor quantum cascade lasers [7], direct multiplication of high frequency sources [8], nonlinear mixing of laser sources [9,10] or Raman shift of infrared gas lasers. Historically, the first terahertz source was obtained by nonlinear difference frequency generation of a ruby laser in quartz [11]. However, it was not until the development of time domain spectroscopy, using ultrashort lasers pumping of semiconductor antennas [12], that an intense study in this region of the electromagnetic spectrum began.

In order to perform spectroscopy in the frequency domain a narrowband tunable light source at the wavelengths of interest is required. Of particular interest are terahertz sources using nonlinear effects. They present the ability to synthesize and control directly the frequency of the radiation, resulting in a tunable narrowband emitted wave. This has been obtained using diode lasers or fiber lasers [13–21], unfortunately low power terahertz radiation is produced. Dual wavelength sources for terahertz generation are often found at long wavelengths, where terahertz wavelength separation is easily achieved. Dual wavelength sources based on dye lasers were studied extensively before terahertz sources were conceived [22–26]. Recently, interest in such sources employing a variety of configurations such as multiple flow cells, gratings or mirrors, has emerged.

In this paper we present experimental results, to the best of our knowledge, of a dual wavelength dye laser with minimum optical elements using either Littrow [27] or Littman-Metcalf [28] configuration. We present a model based on the total internal reflection in the flow cell for one of the wavelengths to obtain the conditions for the dual emission.

## 2. Dye laser characterization

We studied the laser dye Styryl 14 (5,6-dichloro-2[8-(p-dimethylaminophenyl)-2,4-neopentylene-1,3,5,7-octatetraenyl]-3-ethylbenzothiazolium perchlorate; or Benzothiazolium, 5,6-dichloro-2-[[3-[4-(dimethylamino)phenyl]-1,3-butadienyl]-5,5-dimethyl-2-cyclohexen-1-ylidene] methyl]-3-ethyl perchlorate; LDS 950 in the Exciton catalog) [29,30]. The dye was dissolved in an 85% ethylene glycol (EG) and 15% propylene carbonate (PC) solution. This dye fluoresces in the vicinity of 940 nm. It was optically pumped by a frequency doubled

$\text{Nd}^{3+}$ : YAG pulsed laser (Quantel Q-switched, 10 Hz, 6 ns) using a 50 mm potassium dihydrogen phosphate (KDP) crystal.

The dye circulated uniformly on a 20 mm long quartz cell over a 2 mm aisle between a Teflon separator and the transparent front window used as pumping window (Radiant Dyes Laser Acc. GmbH, RDFC20). The pump beam was expanded to illuminate the length of the cell and focused using a cylindrical lens on a 15 mm width beam spot. We used both Littrow and Littman-Metcalf configurations. In both cases a gold coated 1200 lines/mm grating was used as tuning element.

Lasing action was achieved in Littrow configuration (Fig. 1) in which the cavity was formed between the first diffracted order of the grating back feed into the flow cell and the 10% Output Coupler (OC), stable due to the gain lens in the dye [31]. The grating was placed at 150 mm from the end window of the circulating cell. The wavelength tunability was achieved by rotating the diffraction grating. Because of the grating diffracted angles, the grating was placed at a very acute angle. The output was monitored and measured from the flow cell in the opposite side of the diffraction grating.

The best laser output measured at 950 nm was obtained using Styryl 14 dye with a molar concentration of  $8.4 \times 10^{-4}$  M on the 15%-85% PC-EG solution. At this concentration we obtained a laser characteristic curve with a threshold of 1.06 mJ and a slope of 0.864 mJ/mJ. The largest conversion efficiency obtained was of 0.82% at 25 mJ pumping energy.

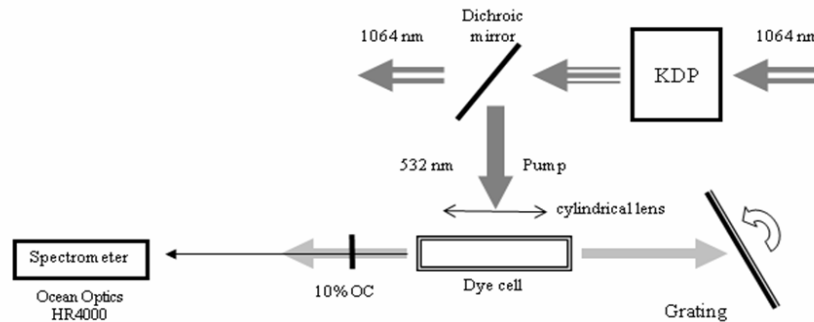


Fig. 1. Dye laser in Littrow configuration. Tuning is achieved by rotating the grating. OC: Output coupler.

Lasing in the Littman-Metcalf configuration (Fig. 2) was obtained with the cavity formed between the first diffracted order of the grating retroreflected by a 100% dielectric mirror in the 950 nm region and a 10% Output Coupler (OC) placed at 100 mm from the cell. The distance between the retroreflecting mirror and the end of the cell was 200 mm. Again, the grating was placed at an acute angle in order to achieve the retroreflection and the output was monitored and measured from the flow cell in the opposite side of the diffraction grating. The wavelength tuning was obtained rotating the 100% retroreflector mirror and the laser was stable due to the gain lensing in the gain medium.

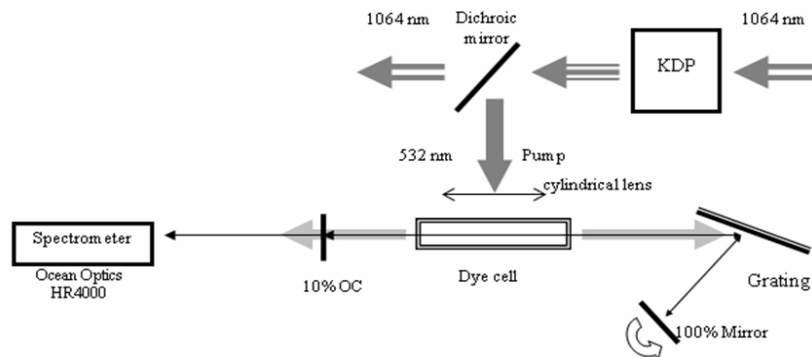


Fig. 2. Dye laser in Littman-Metcalf configuration. Tuning is achieved by rotating the 100% mirror. OC: Output coupler.

Using the Littman-Metcalf configuration with Styryl 14 dye at the molar concentration mentioned above ( $8.4 \times 10^{-4}$  M) we obtained laser output from 910 nm to 960 nm by rotating the tuning retroreflector mirror approximately four degrees (Fig. 3).

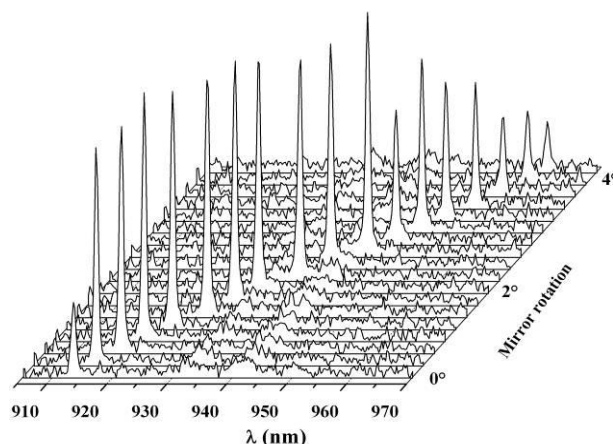


Fig. 3. Wavelength tuning of the dye laser using Styryl 13 on a 15%:85% PC-EG solution with a  $8.4 \times 10^{-4}$  M concentration pumped by 532 nm 4 nsec pulses on Littman-Metcalf configuration.

### 3. Dual wavelength operation

As mentioned earlier, contrary to the standard use of the Littrow and Littman-Metcalf configurations, we used the light leaving the output to characterize the emission of the system. Consequently, we observed while lasing at some positions of the grating a double spot coming from the laser (Fig. 4a). Both spots departed from each other approximately  $1^\circ$  to  $5^\circ$ . The initial assumption was that it was a reflection within the dye flow cell. When feeding the light into a spectrum analyzer using a collecting lens, both spots were measured simultaneously and a dual wavelength lasing operation was observed (Fig. 4b).

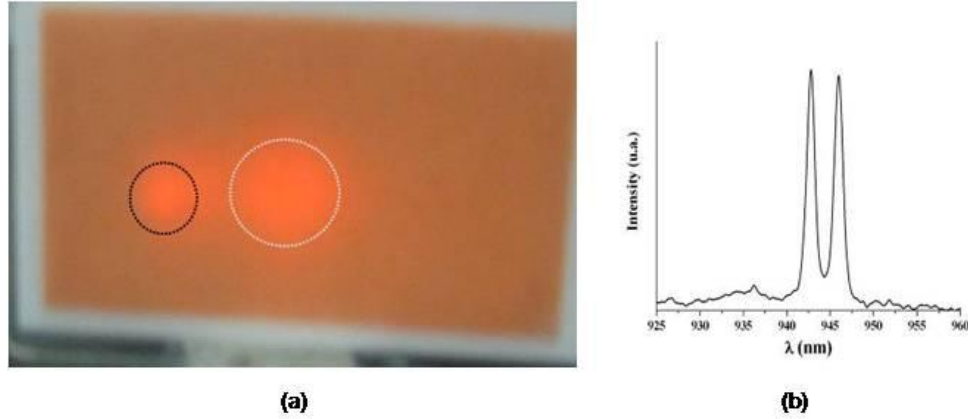


Fig. 4. (a) Double lasing spot observed on a fluorescent card placed 300 mm from the flow cell using Littman-Metcalf configuration. The left spot (black circle) is smaller than the right spot (white circle). (b) Spectrum obtained when both spots were measured simultaneously in the spectrum analyzer showing dual wavelength operation.

We considered different possibilities to explain the dual wavelength operation and reached an explanation by examining the trajectories of the light inside the flow cell. Besides the standard grating cavity, there is a stable ring cavity tuned at different wavelengths, explaining the dual wavelength operation. The ring trajectory is formed between the output coupler and the rear reflecting element using total internal reflection (TIR) on the interface between either the flowing dissolved dye ( $n = 1.42$ ) and the quartz cell ( $n = 1.46$ ) or the quartz cell and the air ( $n = 1$ ) surrounding the cell. We were able to observe this behavior due to the distance between reflecting elements and the flow cell, combined with the relatively long length of the cell.

#### Littrow configuration

We first turn our attention to the Littrow configuration [27], where the grating is normally used as retroreflector on the first diffraction order for some specific wavelength (Fig. 5). Longer wavelengths are diffracted to a larger angle and the opposite will occur for smaller wavelengths. The cavity is stable because the gain lensing occurs on the laser medium.

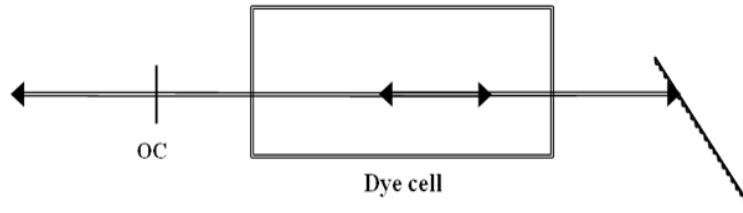


Fig. 5. Dye laser cavity in Littrow configuration. OC: Output coupler.

As one of the reflectors closing the cavity is a diffraction grating, used as a tuning element, the diffracting orders coming from a diffracting element are governed by the equation:

$$\Lambda(\sin \theta_i + \sin \theta_m) = m\lambda \quad (1)$$

where  $\lambda$  is the wavelength,  $\Lambda$  is the diffraction grating period,  $\theta_i$  is the incident angle with respect to the normal, and  $\theta_m$  is the reflected angle with respect to the normal for the  $m$ 'th order. For the first diffraction order the incident and diffracted angles are the same.

Now, we observe the second cavity responsible for the dual wavelength operation. As mentioned above, it is formed between the output coupler and the rear reflecting element

using total internal reflection (TIR) on the interface between either the flowing dye and the quartz cell or the quartz cell and the air surrounding the cell. In Fig. 6 an exaggerated version of the trajectory followed by the different beams is presented, in which the trajectory adjustment due to different refractive indices have been neglected for simplicity. There is only one wavelength which has the same incident and reflected angles in the output coupler and satisfies Eq. (1) simultaneously.

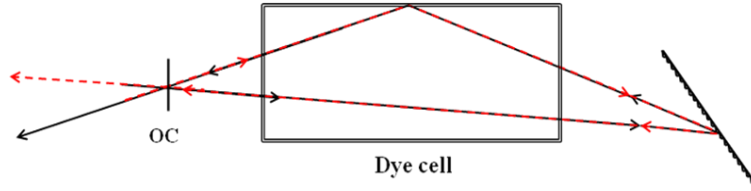


Fig. 6. Ring cavity through the flow cell using TIR and the external reflecting elements. OC: Output coupler.

If we change the distance between the cell and the diffraction grating, the angles needed to close the cavity necessarily change in order to accommodate the lasing action, therefore changing the wavelength of the ring cavity (Fig. 7).

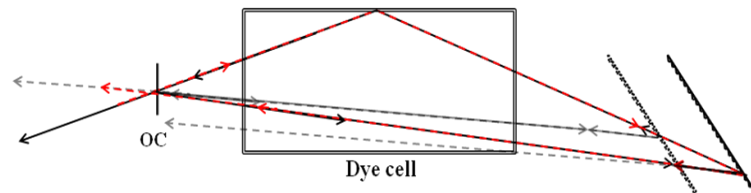


Fig. 7. Ring cavity at different flow cell – diffraction grating distances. The angles required to form a stable ring cavity are different depending on the distance; in the figure the stable angles for the shortest distance (gray line) are not the same as if the grating is farther apart (red/black line), thus the angle selected by the grating is different. OC: Output coupler.

This angle accommodation does not occur for the linear Littrow configuration (Fig. 5) which remains the same independent on the flow cell-grating distance. Therefore, we can tune the wavelength separation between both wavelengths by controlling the flow cell and the diffraction grating ( $d_{cm}$ ), and tune the overall lasing system by rotating the grating. Consequently, the new operating wavelength emerges the laser at a slightly different angle from the other. The longer wavelength is achieved on the ring cavity. We can therefore have simultaneously two stable cavities which have lasing action in different volumes in the flow cell; therefore they can simultaneously emit light in spite of having a homogeneously broadened gain medium as the dye (Fig. 8).

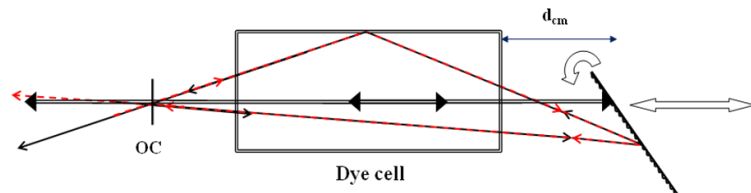


Fig. 8. Dual wavelength laser in Littrow configuration. Different wavelengths are obtained using the linear (double line) and ring (single line) trajectories depicted in the figure. Tuning is achieved by rotating the grating. Wavelength peak separation is achieved by controlling the separation ( $d_{cm}$ ) between the grating and the flow cell. OC: Output coupler.

Each wavelength departs from each other depending on the grating period and distances between the flow cell and the grating, explaining the double spot observed (Fig. 4a). In reality, there are three spots but two of them are close enough to be indistinguishable and appear as a larger spot. Both cavities are stable due to gain lensing on the gain medium.

### Littman-Metcalf configuration

In a slightly more elaborated way, the Littman-Metcalf configuration [28] has an equivalent behavior for the dual wavelength operation. In this configuration the first diffracted order from the grating is retroreflected into the incident path using a mirror (Fig. 9).

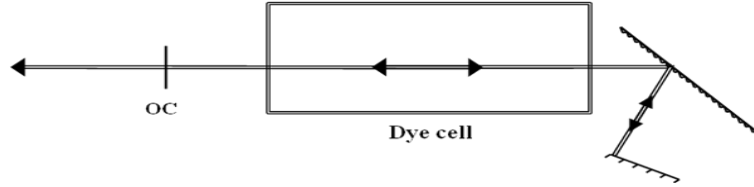


Fig. 9. Dye laser cavity in Littman-Metcalf configuration. OC: Output coupler.

The second cavity responsible for the dual wavelength operation is once again formed between the output coupler and the rear reflecting element using total internal reflection (TIR) on the interface between either the flowing dye and the quartz cell or the quartz cell and the air surrounding the cell. In Fig. 10, an exaggerated version of the trajectory followed by the different beams is presented, in which the trajectory adjustment due to different refractive indices have been neglected for simplicity. There is only one wavelength which has the same incident and reflected angles in the output coupler and satisfies Eq. (1) simultaneously using the pivoting point in the reflecting mirror.

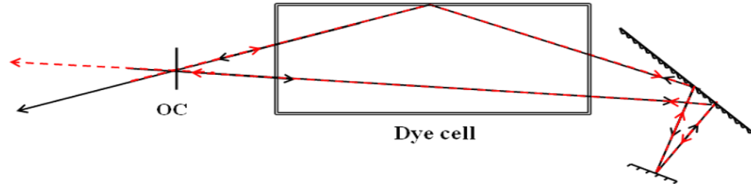


Fig. 10. Ring cavity through the flow cell using TIR, the diffraction grating and a mirror reflecting the first diffracting order. OC: Output coupler.

If we change the distance between the reflecting mirror and the diffraction grating, the angles needed to close the ring cavity necessarily change in order to accommodate the lasing action, therefore changing the wavelength of the ring cavity (Fig. 11). Similar behavior can be obtained by rotating the diffraction grating, but this will also change the wavelength of the linear Littman-Metcalf cavity, which is not a convenient option.

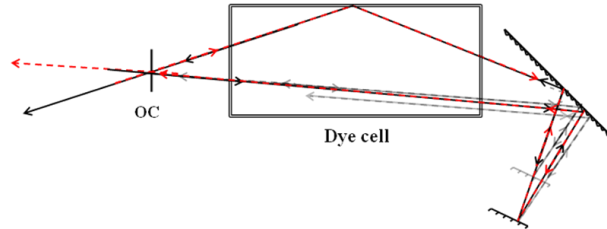


Fig. 11. Ring cavity at different diffraction grating-mirror distances. The angles required to form a stable ring cavity are different depending on the distance; in the figure the stable angles for the shortest distance (gray line) are not the same as if the mirror is farther apart (red/black line), thus the angle selected by the grating is different. OC: Output coupler.

Because we change the mirror-grating distance, leaving the grating unaltered, this angle accommodation does not occur for the linear Littman-Metcalf configuration (Fig. 9) which remains the same independent on the mirror-grating distance. Therefore, in this case we can tune the wavelength separation between both wavelengths by controlling the mirror and the diffraction grating ( $d_{cm}$ ), and tune the overall lasing system by rotating the grating. Consequently, the new operating wavelength emerges the laser at a slightly different angle

than the other. Once again, the longer wavelength is obtained in the ring cavity. We can therefore have simultaneously two stable cavities which have lasing action in different volumes in the flow cell. Therefore, simultaneous dual wavelength emission occurs even when a homogeneously broadened gain medium as the dye is used (Fig. 12).

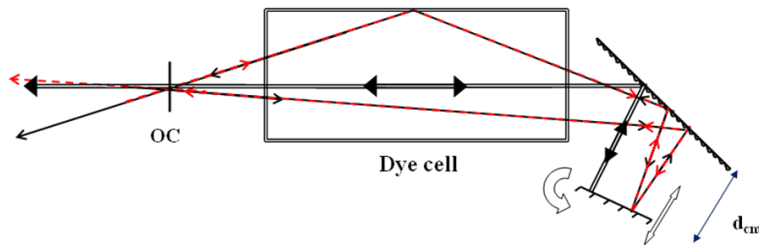


Fig. 12. Dual wavelength laser in Littman-Metcalf configuration. Different wavelengths are obtained using the trajectories depicted in the figure: linear (double line) and ring (continuous black/red line). Tuning is achieved by rotating the retroreflecting mirror and the wavelength separation is controlled by the separation between the grating and retroreflecting mirror ( $d_{cm}$ ).

As in the Littrow configuration, each wavelength departs from each other depending on the grating period, the mirror-grating distance, and the separation between the grating and the retroreflecting mirror. In order to avoid competition from the Fabry-Perot cavity formed by the lateral windows, the flow cell is placed at an angle perpendicular to the cavity plane.

Therefore, the dual wavelength operation in a dual cavity using traditional linear configuration and a ring cavity using total internal reflection, employing a gain medium in either Littrow or Littman-Metcalf configurations, is possible based on the reflecting trajectories at different wavelengths. There are three exiting beams, two of them are close enough to be undistinguishable and they appear as the larger spot in Fig. 4. The longer wavelength is achieved on the ring cavity, while the shortest is obtained in the traditional linear cavity. In order to have both cavities passing thorough the dye cell, the OC must present a small tilt to accommodate for both trajectories. We can foresee that a ring cavity using a dispersive element as a reflecting element will have similar dual wavelength operation.

#### 4. Dual wavelength experimental observation

To perform a controlled observation of the dual wavelength operation of the laser we use the same experimental setup as the one described in the first section. We placed the dye flow cell at a small angle, perpendicular to the ring cavity plane to avoid Fabry-Perot contribution of the lateral windows of the cell. We also place the OC at a small angle with respect to the flow cell plane. The dual wavelength operation is tunable by rotating the grating in the Littrow configuration (Fig. 13a) or the retroreflecting mirror in the Littman-Metcalf (Fig. 13b) configuration. The dual wavelength tuning range on Littrow configuration, rotating the feedback grating close to  $4^\circ$ , was between 910 nm to 960 nm. Rotating the retroreflecting mirror in the Littman-Metcalf configuration close to  $2^\circ$  the tuning achieved was from 900 nm to 970 nm. The characteristic curve for the dual wavelength operation presents a similar slope and threshold to the one obtained in single wavelength experiments.



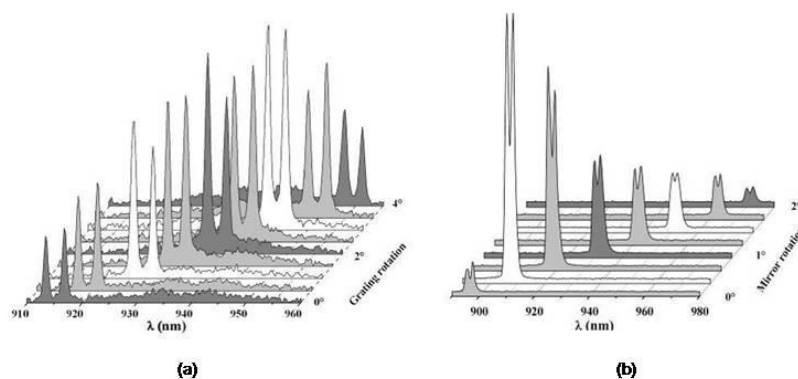


Fig. 13. Dual wavelength tunability of the dye laser using Styryl 14 on a 15%:85% PC-EG solution with a  $8.4 \times 10^{-4}$  M concentration pumped by 532 nm 4 nsec pulses on (a) Littrow and (b) Littman-Metcalf configurations.

Taking into account the 2 mm free opening in the flow cell between the quartz window and the Teflon separator for the 20 mm long flow cell, angles between  $0.5^\circ$  and  $2.5^\circ$  are allowed for both configurations. Using Littrow configuration and changing the distance between the flow cell - diffraction grating ( $d_{cm}$ ) from 20 mm to 150 mm, we observed a peak separation variation from 1.0 to 3.4 nm in wavelength or 0.35 to 1.2 THz, respectively (Fig. 14b). At the same operating wavelength, using Littman-Metcalf configuration operating around 940 nm, varying the distance between the grating and the retroreflecting mirror ( $d_{cm}$ ) from 150 mm to 350 mm, we measured a wavelength separation from 1.4 to 5.6 nm in wavelength or 0.5 to 1.9 THz in frequency, respectively (Fig. 14b).

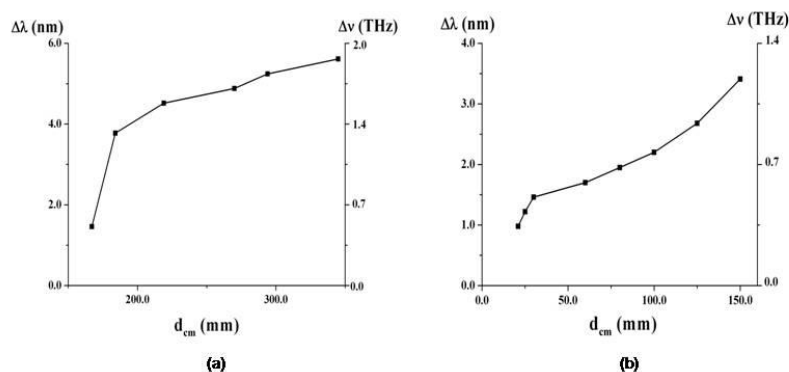


Fig. 14. Separation between wavelength in the dual wavelength operation with respect to the distance between the flow cell and the feedback element ( $d_{cm}$ ) for the (a) Littrow and (b) Littman-Metcalf configurations.

Because both wavelengths travel in different volumes inside the circulating cell, the gain medium is not shared between the different paths and simultaneous lasing is achieved in spite of the homogenous broadening of the Styryl 14 dye in the PC/EG solvent. To verify that simultaneous lasing action occurs in both wavelengths, we used a 3 mm x 3 mm x 3 mm c-cut Lithium Triborate (LBO) crystal. We measured, using the aforementioned fast scanning monochromator, the second harmonic generation of each of the signals and the sum frequency generation between them (Fig. 15). Sum frequency generation is observed only when both signals are simultaneously present, confirming simultaneous dual wavelength operation. This result is encouraging because the nonlinear effect is observed even when the system does not provide polarized output. This opens the possibility of nonlinear mixing for THz generation in the future using the setup presented in this paper.

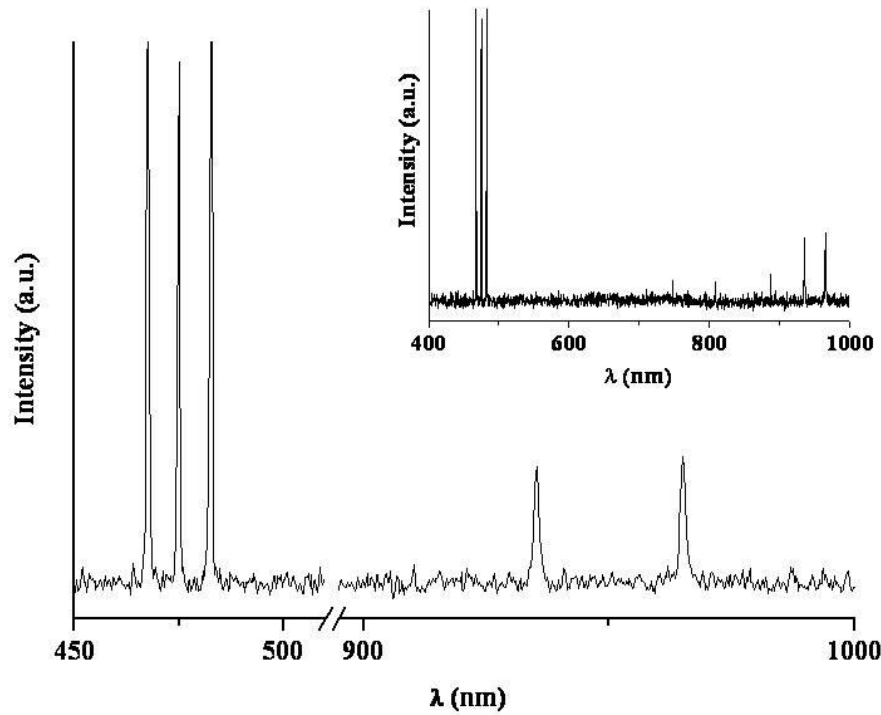


Fig. 15. Dual wavelength lasing in Littrow configuration ( $\lambda_1 = 935.3$  nm and  $\lambda_2 = 965.0$  nm) and the signal through a LBO crystal exhibiting second harmonic (467.6 nm and 482.5 nm) and sum frequency generation (479.9 nm). The inset shows the complete wavelength scan in which a long pass filter was used to reduce the fundamental signals strength.

We monitor the tuning and wavelength separation controlling the grating angle and the separation between the flow cell and the grating, for the Littrow configuration. For example, we choose the sum frequency generated at 475 nm by changing the separation  $d_{cm}$  and rotating the grating to compensate the detuning (Fig. 16).

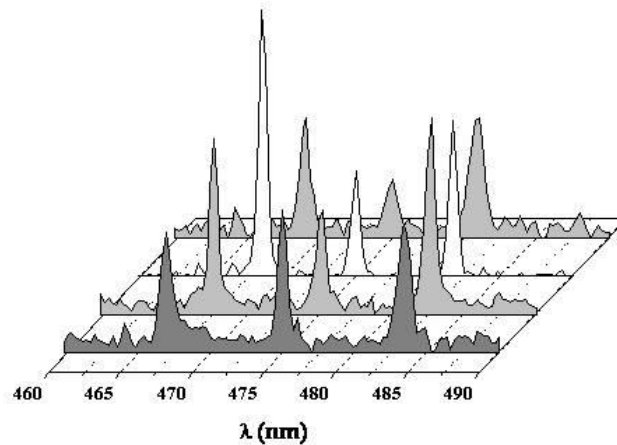


Fig. 16. Wavelengths scan for the dual wavelength laser second harmonic and sum frequency when changing the flow cell-grating separation and tuning the wavelength to maintain the sum frequency at a fixed wavelength.

## 5. Conclusions

In this work, we present a dual wavelength dye laser using a grating cavity in Littrow and Littman-Metcalf configurations operating in the near infrared. The dual wavelength operation is achieved by means of two beam trajectories in the cavity configuration: traditional linear grating cavity and ring grating cavity. On the ring grating cavity the reflections occur on an output coupler, TIR in the flowing cell window and a diffraction grating retroreflector. Two independent wavelengths exit the laser at slightly different angle, hence simultaneous operation is confirmed. The spectral separation among the two wavelengths can be controlled by varying the separation between the flow cell, the dispersing grating, and the retroreflecting element.

In particular, pumping Styryl 14 laser dye in a 15%:85% PC-EG solution with 4 nsec pulses of 532 nm  $\text{Nd}^{3+}$ :YAG doubled laser we obtain single wavelength operation tunable between 910 nm to 970 nm using a 1200 lines/mm grating with threshold close to 1.0 mJ and slopes close to 0.9 mJ/mJ with an effective conversion efficiency of 0.82% at 25 mJ pumping energy. Under these conditions, on dual wavelength operation, varying the flow cell-retroreflecting element separation, we obtained wavelength separation between 2 nm and 20 nm in either Littman-Metcalf or Littrow configurations, exiting at angles close to  $2^\circ$ .

Therefore, we have obtained a dual wavelength laser system using minimum cavity elements: pumping source, flow cell with a dye laser, output coupler, and a dispersive retroreflection element, either a diffracting grating or a diffracting grating combined with a mirror. We foresee that using the combined linear:TIR-ring cavities, and any dispersive element as a reflecting element, will have similar dual cavity and therefore independent laser dual wavelength operation. This type of system could be used as a THz source using difference frequency generation.

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