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Second harmonic and sum frequency generation with a double-wavelength dye laser

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With the use of a double-wavelength flash-lamp pumped dye laser two second harmonics and the sum frequency were generated in a KDP crystal. The double-wavelength generation was obtained with the use of a Fabry-Pérot etalon in the laser cavity.

1. Introduction

The frequency generated in a dye laser depends on the kind of the dye, the solvent, the dye concentration, the mixture ratio (in the case of a two-component active medium), and on the dispersive properties of the laser cavity. A large number of dyes for the laser generation of light ranging from the near ultraviolet to the near infrared have been known since many years [1]. The broad gain curve of a laser dye enables the frequency tuning across a broad spectral region by the use of prisms, gratings, Fabry-Pérot etalons or filters.

The applicability of a dye laser can be extended to the whole UV region by the second (or higher) harmonic frequency generation. The harmonic frequency can be also tuned by the fundamental frequency tuning as well by the adjustment of the phase matching angle, when a nonlinear crystal is used. The same may be done by another method sometimes more efficient [2], i.e. the sum frequency generation by mixing the frequency of a dye laser with the frequency of another dye laser [3, 4], pumping pulses or their harmonics [5–9], cw pumping laser [10], as well as with the frequency of an additional laser [11]. All these methods require at least two lasers. A generation of four second harmonics and six sum frequencies with the use of one laser has been recently observed for four He-Ne IR fundamental laser frequencies [12, 13].

The broad gain curve of a laser dye enables also a simultaneous generation of two wavelengths in a laser cavity with high Q for two frequencies. This is realized in many different ways, usually by the use of two dispersive elements in the cavity, e.g. [14–16].

The simultaneous generation of two wavelengths (even separated by tens nm) can be obtained with a Fabry-Pérot etalon in the laser cavity, provided that the gain curve of the laser dye overlaps the free spectral range of the etalon. For large spectral separation of the generated wavelength it is advisable

to use two-component active media. Such a kind of laser and one nonlinear crystal allow to generate two fundamental and two second harmonics and the sum frequency.

The purpose of this work was to generate second harmonics and the sum frequency with one double-wavelength flash-lamp pumped dye laser.

2. Experiment

To carry out the investigations a dye laser was used with a 2×10^{-4}M methanolic solution of rhodamine 6G and (for more significant separation of the generated bands) a mixture of 2×10^{-4} M methanolic solutions of rhodamine 6G and uranin as the active medium. The solution passed through a 10 cm long, 2 mm I. D. glass cell, at a flow rate optimized to prevent both heating effects and the turbulence of the solution. The active medium was pumped with two linear, air-filled flash-lamps supplied with the energy of 150 J. The flash-lamps between which a dye cell was inserted were wrapped together with a reflector of aluminium foil. The 35 cm long cavity was terminated by two flat, broad-band dielectric mirrors with 100 and 90% reflectivities. All optical flats inside the cavity were slightly wedged to prevent uncontrolled etalon effects. In order to obtain the double-wavelength generation a Fabry-Pérot etalon was inserted into the cavity. A few μm thick mica flake without any reflecting coatings was used as the etalon. To prevent resonances arising from the birefringence of mica the laser beam was polarized by a glass plate placed in the cavity at the Brewster angle. The mica flake was experimentally positioned so as to obtain a relatively clear spectrum. In order to generate two bands with about the same energy the etalon was tuned by rotation around its vertical axis.

The second harmonics and the sum frequency were generated at room temperature in a 1 cm long KDP crystal with z cut of 60°10′. The phase matching angle was controlled by tilting the crystal placed outside the cavity. The KDP crystal was used in a typical arrangement, e.g., [12].

All spectra were recorded with the Zeiss PGS 2 grating spectrograph. The fundamental beam was focussed within the KDP crystal with a 10 cm focal-length lens. The crystal was placed close to the fully open spectrograph slit at a distance of about 3 m from the output mirror of the dye laser.

The pulse energy of the two fundamental bands selected with interference filters was measured with a calorimetric type meter. The relative intensities as well as the pulse width were measured with the use of a RFT P 12 FQ 51 (S-23 photocathode) photomultiplier and the Schlumberger OCT 559 oscilloscope. The relative intensities were evaluated with the use of filters with known transmission, taking into account the spectral response characteristic of the photomultiplier. The second harmonics and the sum frequency were selected by adjustment of the phase matching angle. For efficiency measurements the

crystal was placed close to the output mirror. The fundamental beam was focussed within the crystal also with the 10 cm focal-length lens.

3. Results

The laser generated simultaneously two bands in a pulse with the halfwidth of about 1 μs. For the rhodamine 6G (R6G) solution and a 15.5 nm free spectral range etalon these bands were centred at about 587.0 nm and 602.5 nm and in each band the pulse energy was 0.9 mJ. A 19 nm separation of the generated bands was obtained while using a mixture of rhodamine 6G and uranin (R6G : U) solution in 5 : 3 ratio. These bands were centred at about 579.0 nm and 598.0 nm and in each band the pulse energy was 0.7 mJ.

The phase matching angles for the second harmonics and the sum frequency generation were calculated for $oo \rightarrow e$ interaction from the known relations [17]:

$$\theta_m^{2\omega} = \arcsin\left[\frac{(n_o^{\omega})^{-2} - (n_o^{2\omega})^{-2}}{(n_e^{2\omega})^{-2} - (n_o^{2\omega})^{-1}}\right]^{1/2},$$

and

$$\theta_m^{\omega_1+\omega_2} = \arcsin\left\{\frac{[(n_o^{\omega_1}/\lambda^{\omega_1} + n_o^{\omega_2}/\lambda^{\omega_2})\lambda^{\omega_1+\omega_2}]^{-2} - (n_o^{\omega_1+\omega_2})^{-2}}{(n_e^{\omega_1+\omega_2})^{-2} - (n_o^{\omega_1+\omega_2})^{-2}}\right\}.$$

The refractive indices were calculated from Sellmeier's relation with respect to the values obtained by ZERNIKE [18]. The wavelengths of the second harmonics, sum frequencies, as well as the phase matching angles for the R6G and the R6G: U lasers are listed in table. The calculated phase matching angles listed in this table show a relatively good agreement with the used 62°10′ KDP

Table. Wavelengths of second harmonics, sum frequencies and phase matching angles for the rhodamine 6G (R6G) laser and the laser with the rhodamine 6G: uranin (R6G : U) mixture

	R6G		R6G : U	
	λ [nm]	Θ_m	λ [nm]	Θ_m
$2\omega_1$	293.5	62°27′	289.5	63°52′
$\omega_1 + \omega_2$	297.3	61°24′	294.2	62°
$2\omega_2$	301.25	60°02′	299.0	60°42′

crystal. The spectra of fundamental beams, the second harmonics and the sum frequency for the R6G and the R6G: U lasers are presented in figs. 1 and 2, respectively. The successive spectra were obtained by tilting the KDP crystal around the phase matching angle with a step of about 23′, which corresponds to about 15′ of the refraction angle.

The efficiency of the frequency conversion was pretty low, of the order of $5 \times 10^{-4} - 1 \times 10^{-3}$.

In these investigations no attention was paid to gain high efficiency and narrow spectra.

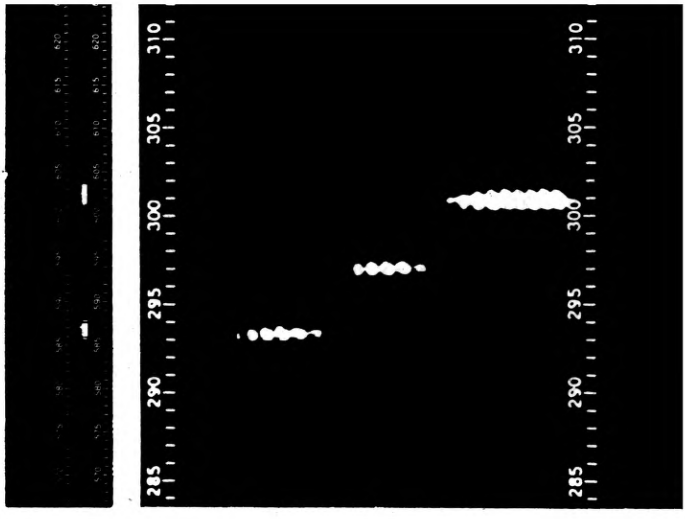

Fig. 1. Spectra of the fundamental frequencies, second harmonics and the sum frequency of the rhodamine 6G laser

4. Conclusions

This simple arrangement with only one dye laser and one nonlinear crystal is a source of two fundamental frequencies and a second harmonic or sum frequency. The phase matchable bandwidth calculated from Miller's expression [19] with respect to Zernike's refractive indices [18] for $\lambda^{\omega} \approx 600$ nm,

$$\delta\lambda^{\omega} = \pm \frac{1.39\lambda^{\omega}}{2\pi l} \left(\frac{\partial n_o}{\partial \lambda^{\omega}} - \frac{1}{2} \frac{\partial n_e^{2\omega}}{\partial \lambda^{2\omega}} \right)^{-1}$$

is about 0.3 nm. The values of the same order can be obtained by using similar expressions for the second harmonic and the sum frequency of ZERNIKE and MIDWINTER [20]. So, the occurrence of both second harmonics and the sum frequency at one orientation of the crystal does not seem to be possible at the widely separated fundamental wavelengths, even at the focussed, convergent beam. The proper UV band can be obtained by the adjustment of the phase

matching angle, while the desired wavelength – by the choice of the active medium and the etalon. This simple method seems to be applicable to other double-wavelength, both pulsed and cw dye lasers.

This inexpensive type of radiation source may find many applications in investigations of multiphoton processes.

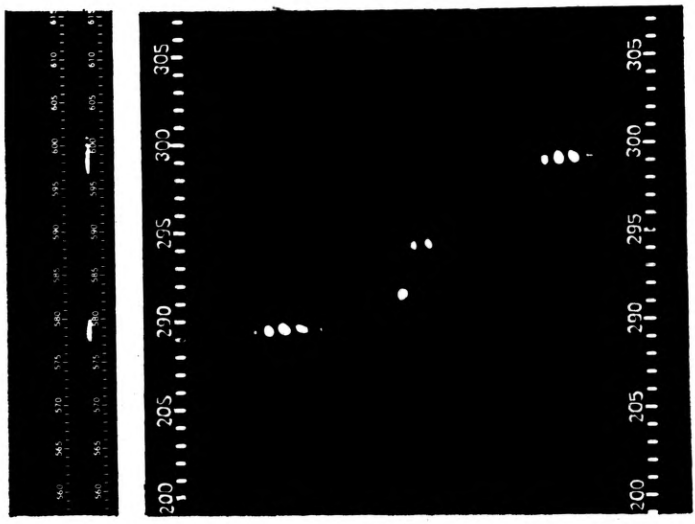

Fig. 2. Spectra of the fundamental frequencies, second harmonics and the sum frequency of the laser with the rhodamine 6G: uranin mixture

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Генерирование второй гармоники, а также суммовой частоты с помощью красильного лазера на двойную длину волны

В кристалле KDP были генерированы вторые гармоники, а также суммовая частота излучения красильного лазера, накачиваемого лампами-вспышками, генерирующего две частоты. Одновременная генерация двух полос была достигнута при использовании эталона Фабри-Пэро.