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Tunable broadband pulsed dye laser

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This Letter describes the construction of a simple dye laser to be pumped by a Q-switched Nd:YAG laser. This system is used to create excited states at a time scale of ~10 ns, to be detected by means of time-resolved EPR[1] or transient absorption spectroscopy. Neither of these techniques requires the spectral bandwidth of the dye laser to be particularly narrow, but spurious wavelengths must remain absent. For our applications multimode variations within the spectral envelope from shot to shot are not important and the dye laser was not designed to minimize these variations (e.g., a long cavity), which would have been necessary if applied to CARS, for example.[2] Recently a pulsed dye laser was reported[3] with a conversion efficiency of up to 20%, however tunability was achieved by changing the dye and/or its solvent. We preferred continuous tunability within the spectral range of the dye used.

Apart from the main wavelength, the output of a short cavity pulsed dye laser contains a background of spurious wavelengths, originating from spontaneous emission,[4] in particular when pumped with high energy pulses from a Q-switched Nd:YAG laser (e.g., at 20 pulses/s: 100 mJ at 532 nm, 60 mJ at 355 nm). Therefore, the concept of an oscillator–amplifier dye laser was used.

To gain maximum throughput efficiency of the dye laser, use was made of the polarized nature of the pump laser output by employing optical elements with such a geometry that all faces of these elements have the Brewster angle with respect to the laser beam. This results in 100% deflections at the air–glass boundary. Also, 100% internal reflections were used. Fused silica Brewster cut elements were preferred because they have a long life expectancy and are relatively inexpensive compared to many different customer-specified high power mirrors (up to 100 MW/cm^2). In the dye laser, described in this Letter, only a few standard broadband mirrors were required.

The output of the Nd:YAG laser is horizontally polarized at all wavelengths (1064 nm, 532 nm using a 90° quartz rotator, and 355 nm). A gull wing prism wavelength separa-

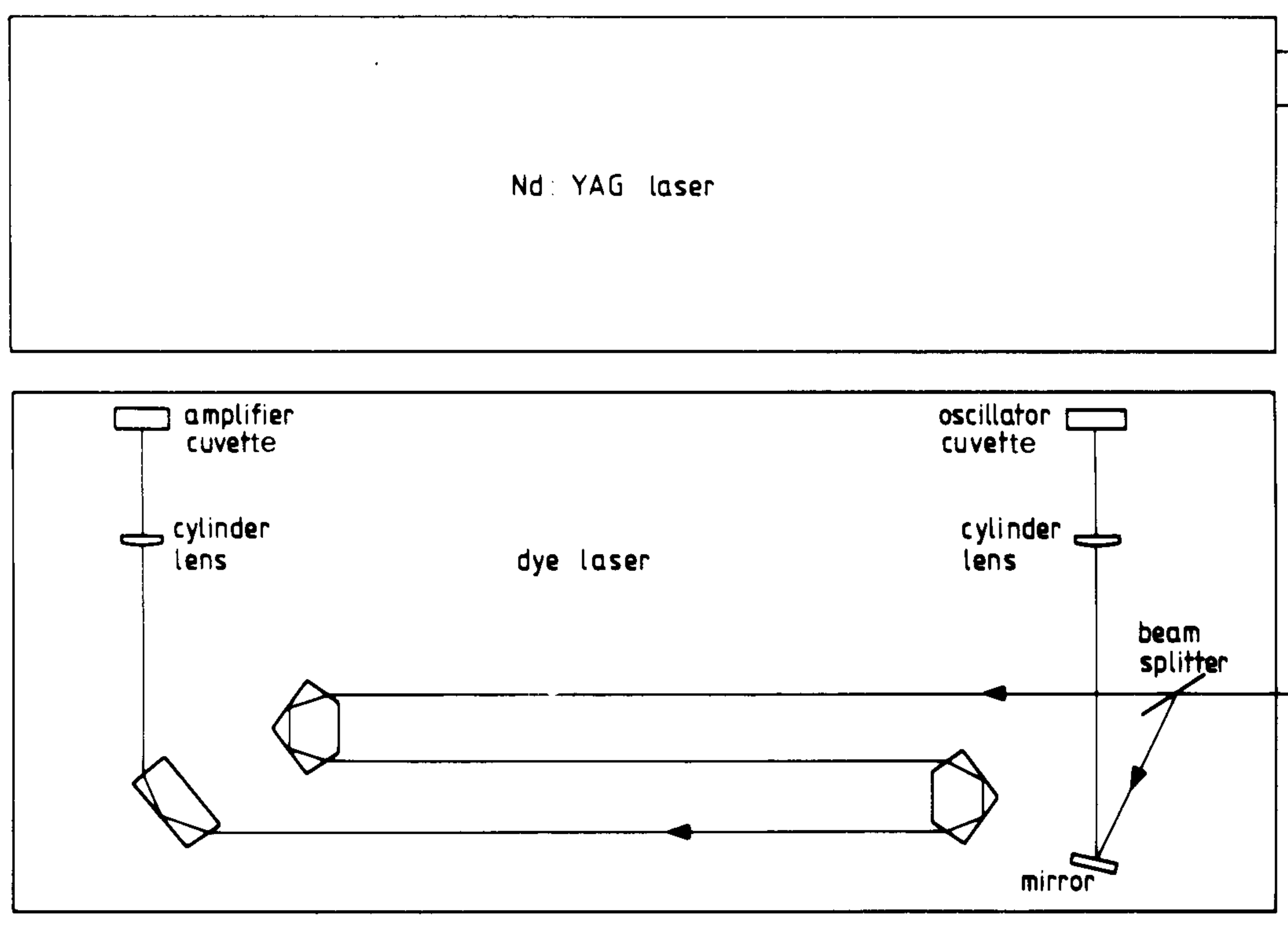

Fig. 1. Layout of the pump scheme.

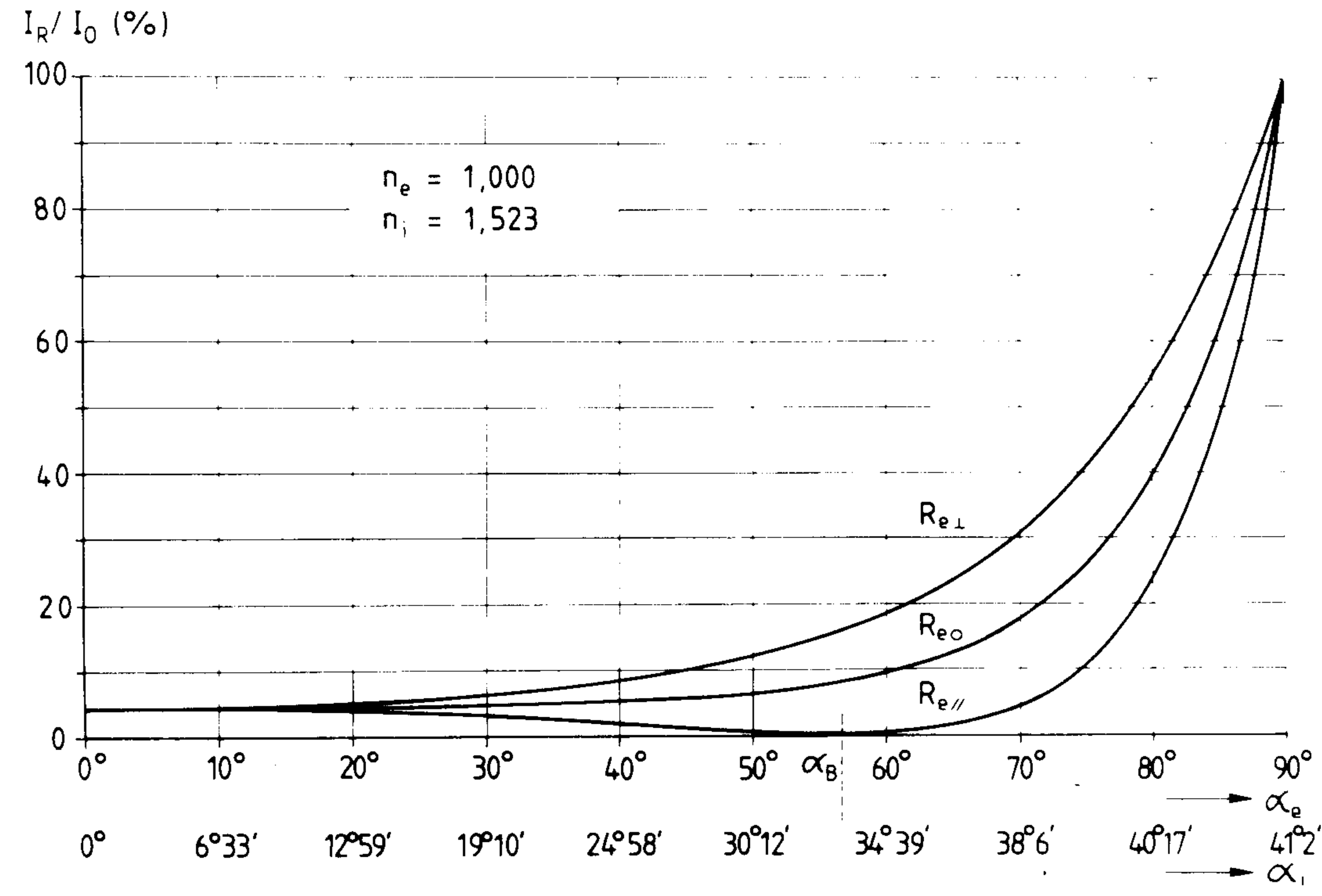

Fig. 2. Reflected fractions of light on the surface boundary of an air–glass transition for unpolarized light (R_{eo}), and light polarized with the electric vector parallel ($R_{e\parallel}$) and perpendicular ($R_{e\perp}$) to the plane of incidence. α_B is the Brewster angle.

tor is used to extract the desired output wavelength. The input and output faces of the two prisms are about at Brewster angle, resulting in negligible reflection losses. The output of the pump laser is directed toward the dye laser by two fused silica Brewster cut prisms (Fig. 1).

To pump the oscillator stage in the dye laser a small part of the main beam is split off, using a thin optical flat substrate (fused silica), which is tilted somewhat from the Brewster angle. The percentage of reflectance of this beam splitter is controlled by changing the angle of the substrate with respect to the direction of the main beam.[5] During alignment, the excitation power of the oscillator stage was chosen as low as possible, but high enough to assure stable operation (~1% of the incoming pump beam power).

The oscillator pump beam is directed to the oscillator cuvette through a fused silica cylindrical lens (f = 100 mm), using a standard broadband mirror (Melles Griot type 08MLB 003/341 or /343). The axis of the cylindrical lens could be rotated to control the overlap of the thin vertical image of the pump beam and the path of the dye laser beam in the cuvette. The focus line could also be adjusted.

The part of the pump beam that is not split off toward the oscillator stage is deflected toward the amplifier stage via an optical delay line, using fused silica Brewster cut elements. The time delay of the amplifier pump pulse is necessary to compensate for the time delay, caused by building up laser action in the oscillator stage.[6] The optical delay line could be optimized by changing the position of the reflecting elements.

Just as in the oscillator cuvette, a thin vertical image is focused in the amplifier cuvette by a cylindrical lens (lens and cuvette are identical to that in the oscillator stage). The cuvettes, the cylindrical lenses, and the Brewster cut elements are all uncoated. Standard flow cuvettes were used (Hellma 134F QS, 10 × 4 mm).

The Brewster angle (α_B) for the optical elements was calculated from $\alpha_B = \arctan \mu$, where μ denotes the refractive index from air to fused silica.[7] To calculate the Brewster angle of the elements in the pump scheme, a theoretical wavelength of 420 nm was chosen as a compromise between the refractive indices[8] at 355 and 532 nm of the Nd:YAG laser output. The reflection losses introduced by this mismatch between theoretical and practical excitation wavelength were still very small, for the region of close to zero reflection around the Brewster angle is rather broad (Fig. 2). Further, the angle of the elements with respect to the light beam could be optimized for minimal reflection.

The oscillator stage of the dye laser consists of a standard broadband mirror (Melles Griot type 08 MLB 003/343 or / 341), four fused silica Brewster cut dispersion prisms,[6] a flow cuvette, and an output coupler (Zeiss FPE 015 nm, 30% reflectance, 3% wedge). The dye cuvette is about at the Brewster angle as a compromise for different optical boundaries. The cavity was kept as short as possible (~20 cm, resulting in ~1.5-ns round-trip time) and arranged in such a way that the laser beam passes the cuvette vertically (Fig. 3).

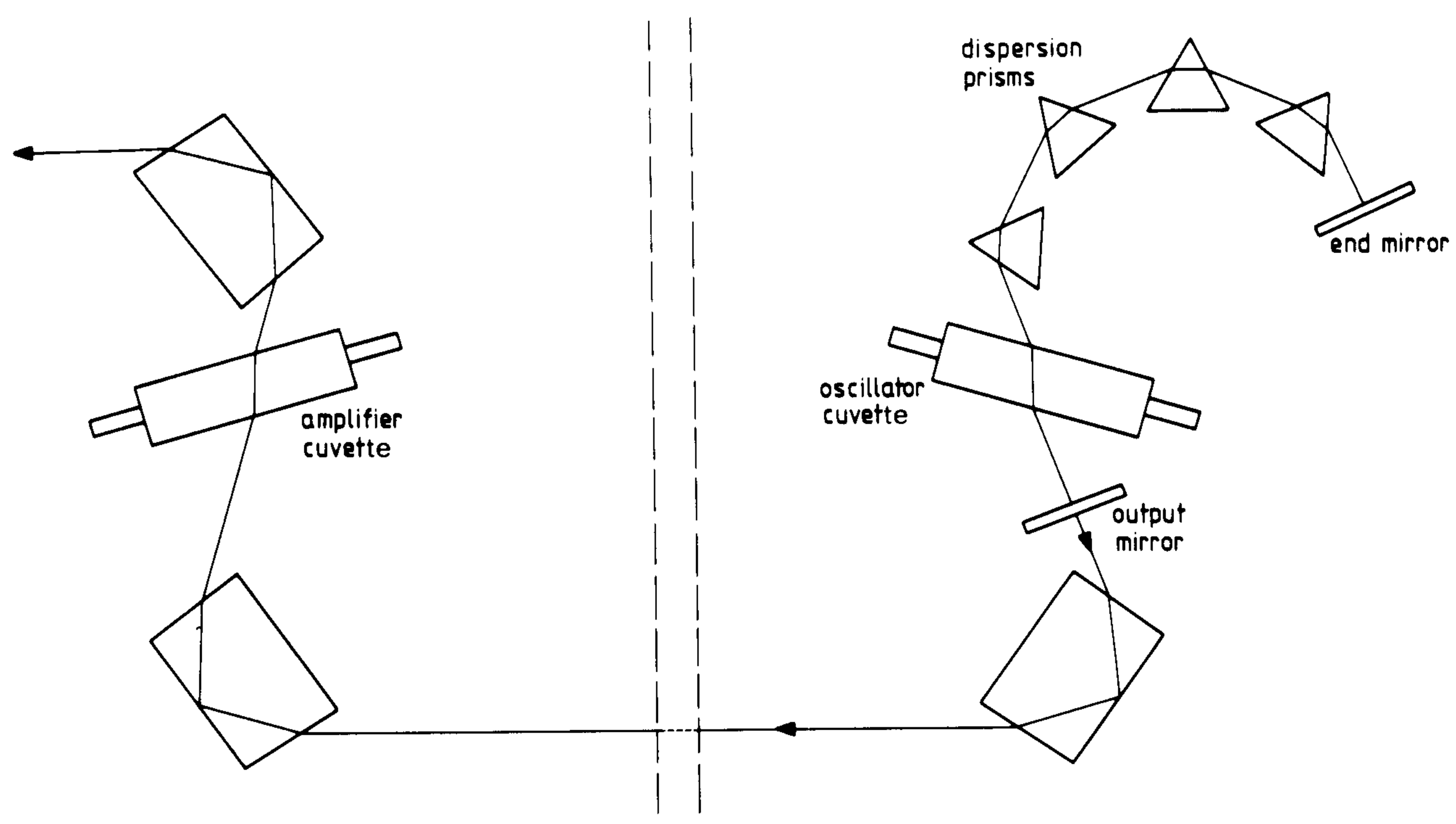

Fig. 3. Layout of the oscillator–amplifier scheme.

A shorter cavity results in more round trips during the 12-ns pump pulse, decreasing the bandwidth of the output. In general, also a lower output percentage of the cavity will increase the spectral purity. The reflectivity of the output mirror and the pump power to the oscillator were determined experimentally by using output mirrors with various percentages of reflection and by varying the pump power: 30% reflectivity and ~1% pump power resulted in reliable operation using a 12-ns width of the pump pulse and a 1.5-ns round-trip time in the cavity.

Using the dispersion theory for multiple prism arrays as discussed by Duarte and Piper[9,10] a theoretical linewidth of the dye laser output of ≃1.5 nm/mrad was calculated. Thus, with an intracavity beam divergence of some millirads a linewidth of a few nanometers is expected; the exact value depending on multipass effects, beam geometry, etc.

The polarization direction of the oscillator light is forced to be horizontal in the cuvette (like the pump beam) by the many faces at Brewster angle within the cavity. Thus, an optimum condition is created for dye molecules with transition dipole moments of excitation and stimulation (and stimulated emission) along the same axis.[6]

Rotation of the excited dye molecules will reduce the stimulated emission. However, the width of the pump pulse, the round-trip time in the cavity, and the rotation correlation time of the dye molecules dissolved in an alcohol (which is estimated to be less than some tens of picoseconds) make this process negligible at the given photon flux.

The output beam from the oscillator was directed toward the amplifier cuvette using two Brewster cut elements. In the second cuvette the light path through the dye solution is vertical again. After passing this dye cuvette the amplified laser beam is deflected horizontally, again using a Brewster cut element, turning the output polarization to vertical. All Brewster angles in the oscillator–amplifier were calculated using a refractive index of silica at 520 nm as a compromise between the blue and red dyes.

The dispersion prisms were mounted together at one block and were prealigned. The oscillator wavelength could be tuned by the end mirror controls. In this way, tuning is uncalibrated, but for most applications this is only a small disadvantage, because once running, the laser system is operated for a long period at the same wavelength. With minor modifications calibrated wavelength selection is possible. The output wavelength was measured using a beam splitter and a 0.25-m monochromator.

Of the deflection elements in the pump scheme only those just before the cylindrical lenses have a two-axis adjustment. All other elements are only partly adjustable. The two 90° deflection elements between pump laser and dye laser were mounted on prism tables. All components of the dye laser and pump scheme were attached to 10-mm thick anodized alumina plates, assembled perpendicularly.

The dye solution was circulated through both cuvettes by means of a centrifugal pump and was cooled by a glass heat exchanger and 2-liter stock water at ambient temperature.

Testing the dye laser, several dyes were used, all of which were dissolved in methanol. Most frequently, a mixture of Rh6G and Rh640 (1:1) was used (Lambda Physik), lasing around 610 nm. With this dye mixture, pumping at 532 nm, 33 mJ/pulse, and 21-Hz repetition rate from a JK Laser HY200, a conversion efficiency of over 10% was reached, quite comparable with values from commercial dye lasers. The amplification of the light pulse from the oscillator was found to be about 200 times. The bandwidth at 609 nm was 3-nm FWHM, as measured with a SPEX minimate 0.25-m monochromator (1-nm resolution), in good agreement with the theoretical expected value. No attempts were made to measure the percentage of amplified spontaneous emission.[11–13] The dimensions of the assembled dye laser are 30 × 20 × 120 cm (width, height, and length, respectively). For the dye laser described here, all fused silica (Suprasil I) components were custom-made by the Optics Group of the University of Amsterdam.

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