DIFFERENCE FREQUENCY GENERATION BY OPTICAL MIXING OF TWO DYE LASERS IN PROUSTITE *

C.D. DECKER † and F.K. TITTEL
Department of Electrical Engineering, Rice University, Houston, Texas 77001, USA

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Mid-infrared radiation tunable from 5.82–7.25 μ has been generated as the difference frequency between the outputs of two independent dye lasers mixed in a proustite crystal set at the phase matching angle. Dual dye laser optical mixing is compared to previous mixing schemes, and is shown to have certain advantages over ruby laser–dye laser mixing and optical parametric oscillator signal–idler mixing in proustite. An extension of the tuning range for ruby–dye mixing to 6.47 μ is also reported.

Tunable coherent infrared radiation has been produced in several laboratories by mixing a ruby laser and a tunable dye laser in a nonlinear crystal, generating the mid-infrared difference frequency [1–4]. The reported wavelength ranges covered by such techniques to date are 3–5.65 μ and 10.1–12.7 μ. Bhar et al. [5] have recently mixed the signal and idler waves (1.87–2.47 μ wavelength) from a proustite optical parametric oscillator (OPO) in a second proustite crystal to generate low power ($\approx$ 200 μW) difference frequency radiation tunable from 7.8–11.9 μ. This technique is attractive since it theoretically allows a broad range of infrared wavelengths to be generated, but it cannot produce high power infrared radiation due to the low power ($\approx$ 50 W) of the mixing signal and idler waves.

In this letter we report an extension of the tuning range for ruby laser–dye laser (hereafter ruby–dye) mixing, and we present a new dual dye laser mixing scheme which we feel has certain advantages over either of the two previous arrangements for frequency mixing. We previously reported [4] the generation of high power difference frequency radiation tunable from 3.20–5.65 μ by mixing the output of an organic dye laser with its ruby pump in a proustite crystal. This tuning range has been extended in the present work to 6.47 μ by generating 5.32–6.47 μ radiation as a result of mixing a Q-switched ruby laser with a 1,3,3,1′,3′,3′-hexamethyl-2,2′-indotricarbocyanine iodide (HEXA) dye solution laser and with a 3,3′-dimethyl-2,2′-oxatricarbocyanine iodide (OXA) dye solution laser in a 9.5 mm length proustite crystal. The experimental arrangement is the same as that reported in ref. [4], except that the proustite crystal is cut so that the normal to the crystal face makes an angle of 26° to the optic axis. Radiation tunable from 5.32–5.95 μ is generated by mixing the ruby radiation with 7984–7860 Å radiation from the HEXA dye solution laser, with peak infrared powers varying from 110 W at 5.32 μ to 60 W at 5.95 μ. Similarly, 5.70–6.47 μ radiation is produced using 7905–7778 Å radiation from the OXA dye solution laser, with peak powers ranging from 80 W at 5.70 μ to 10 W at 6.47 μ.

Ruby–dye mixing in proustite is limited by the existence of a rather low damage threshold for proustite at the ruby wavelength (for Q-switched pulses, about 3 MW/cm^2). The damage threshold at 1.06 μ, by comparison, is in excess of 12 MW/cm^2 [6]. Local heating due to optical absorption appears to be the predominant

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† Present address: GTE Sylvania, Electro-Optics Organization, P.O. Box 188, Mountain View, California 94040, USA.

damage mechanism [7]. The mid-infrared difference frequency power produced by optical mixing is directly proportional to the intensity of the high frequency pump (ruby) radiation incident upon the crystal, so the existence of an upper limit for the acceptable pump intensity limits the infrared power that may be generated. By utilizing pumping radiation in the 7450–7800 Å wavelength range, available from an OXA dye solution laser [8], much higher pump intensities can be tolerated by the proustite crystal without damage. High power, narrow linewidth radiation from such a ruby laser pumped OXA dye laser may then be mixed with longer wavelength radiation from a second dye laser to generate infrared radiation.

Dual wavelength output from a single dye cell has been obtained by several researchers [9–11] using a special dye laser cavity configuration. The outputs from such a dual frequency laser have been mixed in a nonlinear crystal to generate far-infrared radiation [11]. The maximum wavelength separation between the two spectral components of a single dye solution laser is about 300 Å, however, so a single dye system cannot be used to produce the two mixing beams for generating radiation at wavelengths less than about 15 μ.

For this reason two independent dye lasers are required for the generation of mid-infrared radiation by optical mixing of two dye laser beams. Both dye lasers may conveniently be pumped by a common excitation source, which insures synchronization of the two output pulses. The polarizations of the two independent dye lasers can be chosen at will through the use of a linearly polarized ruby laser pump, an appropriate beam splitter to provide the two pumping beams, and properly selected and oriented dye laser cavity elements.

To demonstrate the feasibility of such a dual dye laser mixing approach to the production of mid-infrared radiation, two independent dye lasers have been constructed. Fig. 1 shows the experimental mixing arrangement. The output from a 4 MW Q-switched ruby laser is separated into its horizontally and vertically polarized components by a Glan-laser prism. The horizontally polarized component of 1.5 MW power pumps an efficient narrowband dye laser (dye laser $\neq$1) described elsewhere [8]. The vertically polarized component with a power level of 2.5 MW pumps a similar dye laser (dye laser $\neq$2) which has been designed to predominantly emit vertically polarized radiation. Pumping of both dye lasers is accomplished by quasi-longitudinal pumping with each dye laser beam oriented at an angle of 2° with respect to the ruby laser beam.

The cavity for dye laser $\neq$2 consists of a 27% reflectance front mirror, a 10 mm path length quartz dye cell, a coupling mirror which transmits the ruby radiation into the cavity while reflecting the dye laser beam, and a 60° prism which deflects the dye laser beam in a vertical plane where it strikes a 1200 gr/mm diffraction grating blazed for 5000 Å. The grating grooves are oriented horizontally so as to obtain maximum efficiency for radiation of a vertical polarization. The dye cell is set at an angle of 30° to the vertical so that the entire dye laser cavity predominantly transmits vertically polarized dye laser radiation. With such dye laser cavities, the pumping arrangement shown in

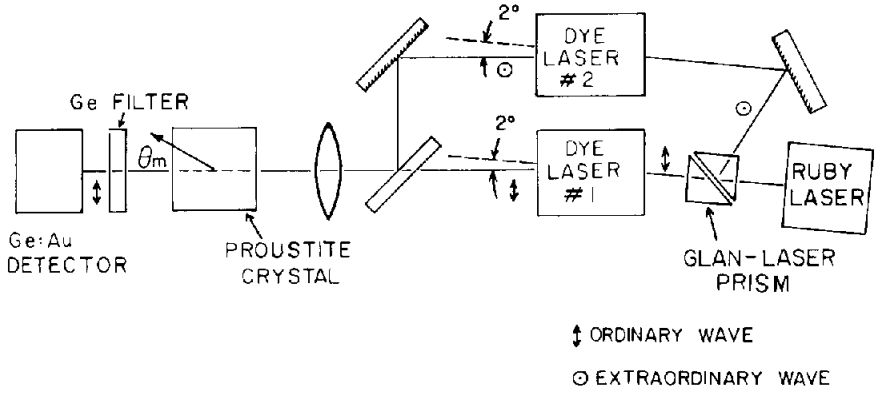

Fig. 1. Experimental arrangement for difference frequency generation by dual dye laser optical mixing.

fig. 1 results in a horizontally polarized output from dye laser ≠1 and a vertically polarized output from dye laser ≠2.

Orthogonal polarization of the two mixing dye lasers is important for optical mixing in proustite, since it allows type 1 phase matching (pump: extraordinary wave; signal and generated infrared idler: ordinary waves) to be used. Type 1 matching is more efficient than type 2 phase matching in proustite by a factor of four [12].

Dye laser ≠1 produces nominally 120 kW dye laser pulses for a solution of 7×10^{-5} M HEXA in DMSO and approximately 80 kW pulses for a similar solution of 3, 3′-diethyl-2, 2′-(4, 5, 4′, 5′-dibenzo)-thiatricarbocyanine iodide (DIBENZO) in DMSO. The spectral linewidth of the laser emission was determined to be 2.5 Å. Dye laser ≠2 produces 200 kW pulses in the wavelength range of 7410–7950 Å for a solution of OXA in DMSO, with a spectral linewidth of 4.4 Å. The difference in spectral linewidth between the two dye lasers is a consequence of the fact that dye laser ≠2 uses a 1200 gr/mm diffraction grating which is less dispersive than the 1800 gr/mm grating used in dye laser ≠1. The two dye laser beams are recombined by a dichroic reflector and are focused onto the 26° cut proustite crystal by a 30 cm focal length lens. Broadband AR coatings are applied to the faces of the proustite crystal so that the entrance face transmits approximately 90% of 7500–9000 Å radiation, while the exit face transmits 90% of 6–10 μ radiation.

Mid-infrared difference frequency radiation is generated by mixing the outputs from the two dye lasers in the proustite crystal which has been angularly tuned to the appropriate phase matching angle for the mixing wavelengths. The difference frequency radiation is detected by a calibrated, liquid nitrogen-cooled, high speed (time constant < 2 nsec) Ge:Au detector after filtering out the dye laser radiation with a Ge filter. This detector is most sensitive to 2.5–6 μ radiation, with response dropping sharply beyond 6 μ.

By using 7520 Å wavelength pumping radiation from dye laser ≠2, difference frequency radiation tunable from 5.82–6.60 μ is generated by frequency mixing with 8635–8487 Å signal radiation from dye laser ≠1 using DIBENZO as the laser dye, while 6.60–7.25 μ radiation is produced by mixing with 8487–8390 Å signal radiation from a solution of HEXA. A Keithley model 105 pulse amplifier is used to amplify the lower power signals from the Ge:Au detector so that they can be displayed on a Tektronix 7904 oscilloscope. Fig. 2 shows typical oscillographs of the mixing dye laser pulses and the amplified difference frequency signal. The slight asymmetry of the difference frequency pulse is due to the response of the amplifier.

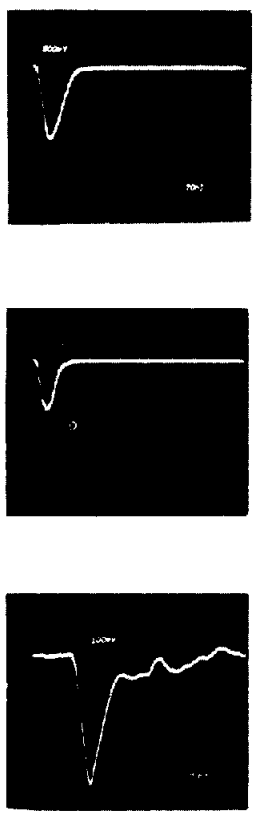

Fig. 2. Temporal characteristics of the mixing laser pulses (a) vertically polarized dye laser output (dye laser ≠2); (b) horizontally polarized dye laser output (dye laser ≠1); (c) difference frequency signal. Horizontal time scale is 20 nsec/div.

Difference frequency radiation of wavelength greater than 7.25 μ could not be detected due to the lack of responsivity of the Ge:Au detector at longer wavelengths. However, the experimental arrangement described in this paper is capable of generating difference frequency radiation out to at least 11 μ, but detection of such radiation requires a detector sensitive to 7–12 μ radiation.

Fig. 3 depicts the difference frequency power gen-

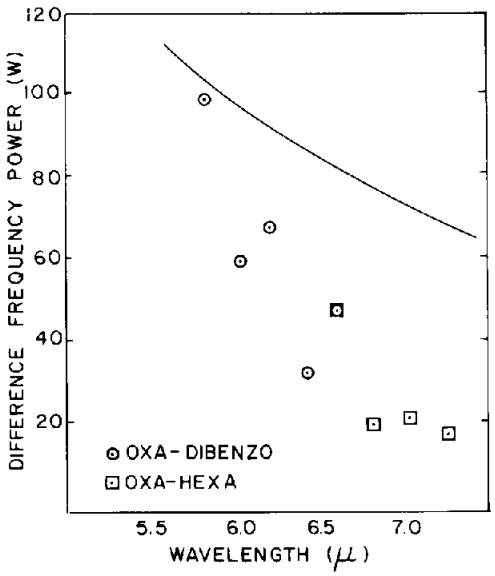

Fig. 3. Difference frequency power generated as a function of wavelength for dual dye laser optical mixing. Solid curve represents the theoretical prediction of infrared power generated by a 200 kW power pump beam of 4.4 Å linewidth mixed with a 120 kW power signal beam of 2.5 Å linewidth.

erated as a function of wavelength for dye laser–dye laser optical mixing. The relatively broad spectral linewidth of the pump radiation at 7520 Å, coupled with the narrower linewidth of the signal radiation from dye laser ≠1, degrades the power generated by dye–dye mixing since not all spectral components of the laser emissions will be phase matched.

Taking into account momentum mismatch due to the finite spectral linewidths of the two mixing dye laser beams, computer calculations based upon the assumption of lorentzian lineshapes for the laser spectral outputs predict the generation of difference frequency power ranging from 104 W at 5.82 μ to 65 W at 7.25 μ, as shown in fig. 3. The use of intracavity etalons to reduce the spectral linewidths of both lasers would result in a substantial increase in the infrared power generated. Assuming no momentum mismatch due to finite spectral breadth of the mixing laser beams (i.e., perfect phase matching), input powers of 200 kW and 120 kW for the pump and signal, respectively, would result in infrared powers varying from 1.68 kW at 5.82 μ to 1.01 kW at 7.25 μ. Powers equal to or greater than those produced by ruby–dye mixing are achieved even with the broader linewidth dual dye arrangement, however, since both mixing beams may be focused to extremely small beam waists for mixing without damage to the crystal. The use of such tight focusing increases the conversion efficiency for the mixing process.

The use of a tunable dye laser to provide the pump laser beam for optical mixing allows one to tailor experimental conditions so as to optimize certain parameters or to avoid physical limitations such as the low damage threshold in proustite. Generation of wavelengths unattainable by ruby–dye mixing in a crystal of a given cut can often be attained by dye–dye mixing since the phase matching angle for such mixing can be changed by several degrees by changing the wavelengths of the interacting beams. Phase matching with waves propagating normal to the crystal face can be achieved by wavelength tuning of both dye laser ≠1 and dye laser ≠2.

In summary, optical mixing of the outputs of two independent dye lasers has been shown to be an attractive alternative to previous frequency mixing schemes. The infrared power levels generated are comparable to those produced by ruby–dye mixing, and are far in excess of those obtainable by mixing the output beams of an OPO. Use of dual dye mixing theoretically allows the production of difference frequency radiation from about 5 μ to the far-infrared, assuming the existence of appropriate mixing media.

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