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High-efficiency, high-power difference-frequency generation of 0.9–1.5 μm light in BBO

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An efficient method for generation of high energy pulsed ir light between 0.9 and 1.5 μm is described. The technique uses difference frequency mixing of pulsed, visible dye and Nd:YAG laser light in a 10 mm long BBO crystal. Quantum efficiencies of up to 23% and ir pulse energies up to 4.5 mJ are demonstrated. The low shot-to-shot fluctuations of difference frequency generation in BBO make this technique an attractive alternative to the conventional optical parametric oscillator or Raman shifting methods that are currently used to access this spectral region.

1. Introduction

Dye laser systems are by far the most common source for tunable, narrow band, high intensity, pulsed light needed in much of modern optical research. Their fundamental tuning range is, however, presently limited to the range between the near uv (> ≈320 nm) and the near ir (typically 900 nm). To generate shorter wavelength light, the output of the dye laser can be efficiently frequency doubled or multiplied in nonlinear media or it can be Raman shifted [1–3]. For the generation of mid ir light, it is common to generate the difference frequency between a tunable dye laser and a fixed frequency laser in nonlinear crystals like $LiNbO_3$ [4–6]. Alternatively, the output of the dye laser can be Raman shifted [1,2] or the ir light can be directly generated in an optical parametric oscillator (OP)) [7–13]. Since difference frequency generation (DFG) requires phase matching in a suitable nonlinear crystal, restrictions apply to the range of wavelengths that can be readily generated. For this reason, the interesting range from 0.9 to 1.5 μm is, at present, mainly covered by Raman shifting and OPO's. However, both of these generation processes are highly nonlinear and exhibit a threshold behavior. Consequently, a very intense pump source is needed and the output is likely to exhibit considerable shot-to-shot fluctuations. In addition, frequency narrowing of the OPO output is not yet easily done even to the levels commonly found in commercial pulsed dye lasers [14].

Since these problems are not encountered in DFG, it is of interest to find a simple scheme to generate intense, narrow band, tunable, nanosecond pulses of light in the 0.9 to 1.5 μm range starting from visible dye laser radiation. Inspection of the relevant parameters of β-BaB_2O_4 (BBO) [15] immediately shows that this crystal should be well suited for difference frequency mixing of the dye laser output with the fundamental light (1.064 μm) of the Nd:YAG laser. The phase matching curve calculated for type I mixing according to the well-known parameters of the Sellmeier equations is shown in fig. 1. To achieve the desired ir range (0.9–1.5 μm), dye laser output only from the easily accessible 490 to 625 nm region is required. Furthermore, the range of phase matching angles is small enough to permit coverage of this whole wavelength region by a single crystal. The proposed mixing scheme would even permit generation of wavelengths far outside the investigated range. For example, a dye laser wavelength of 422.3 nm would lead to a generated wavelength of 700 nm at a phase matching angle of 27.14° and a dye laser at 694.7 nm would produce light at 2.00 μm with a phase

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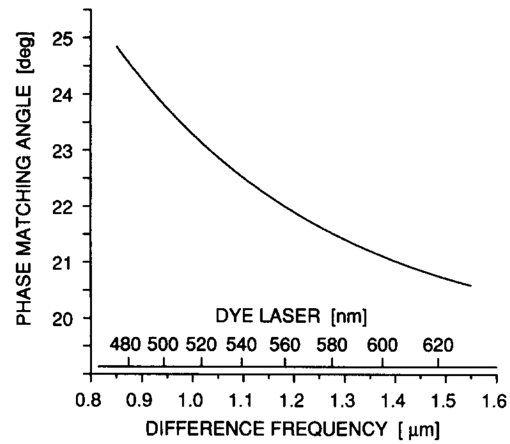

Fig. 1. Calculated phase matching angle for type I difference frequency generation of near ir light from a visible dye laser (e polarization) and the fundamental of a Nd:YAG laser (1.064 μm; o polarization).

matching angle of 20.05°. The high damage threshold of BBO should also allow the use of high intensities and thus yield high efficiency in the nonlinear DFG process.

To our knowledge, no experimental demonstration of difference frequency generation in BBO with nanosecond lasers has been reported. It has been used for femtosecond pulses [16] and implicitly the principle has been proven in BBO OPOs, where the wavelength of the idler was in the desired range [10–12,14]. However, the above mentioned difficulties have so far prevented the use of the these devices in spectroscopic applications. In this communication we show that difference frequency generation in BBO is in fact very efficient and leads to an extremely simple and useful light source for the near ir. We demonstrate this experimentally by difference frequency mixing the output of a Rhodamine 6G dye laser with part of the fundamental output of the Nd:YAG laser used to pump the dye laser.

2. Experimental configuration

The experimental setup is schematically shown in fig. 2. An injection seeded Q-switched Nd:YAG laser (Continuum NY 81 [17]) was used to pump the dye laser (Quanta Ray PDL1 [17]; Rhodamine 6G dye) and to provide the 1.064 μm signal wave for the mixing process. To pump the dye laser 190 mJ (40%) of the frequency doubled output of the Nd:YAG laser was used, resulting in 52 mJ of tunable light at the maximum of the dye curve. The output beam was steered onto a dichroic beam combiner with an uncoated fused silica prism. The residual fundamental output of the Nd:YAG laser was first attenuated by reflection with a partially reflecting mirror ($R=25\%$) and subsequently by a dielectric mirror whose reflectivity could be tuned between 50% and 90% by changing the angle of incidence. The two beams were combined with a dichroic mirror and carefully overlapped. Finally, a telescope consisting of uncoated $f=150$ mm and $f=-50$ mm lenses was employed to collimate the beams. This was necessary to generate both the high intensities needed in the nonlinear mixing process and to accomodate the 5 mm aperture of the BBO crystal.

The diameters (intensity decreased to $1/e^2$) of the collimated dye laser and 1.064 μm beams were measured to be 2.4 mm and 2.1 mm, respectively. Due to the Fresnel losses on the various optical surfaces, the maximum pulse energies of the two beams at the entrance surface of the BBO crystal were decreased to 39 mJ and 42 mJ, resulting in peak intensities of 115 MW/cm^2 and 100 MW/cm^2, respectively. The pulse lengths were measured to be 7.5 ns and 7.9 ns, respectively, with a smooth temporal profile due to the injection seeding of the Nd:YAG laser. To ensure perfect temporal overlap of the two laser pulses the 1.064 μm beam was optically delayed by placing the BBO crystal 3.3 m downstream from the laser, whereas the simultaneous 532 nm output, used to pump the dye laser, was steered over the shortest path possible, as was the dye output.

The polarization of the dye laser and 1.064 μm beam were vertical and horizontal, respectively. This is appropriate for the desired type I phase matching in the BBO crystal with the dye laser as pump wave and the Nd:YAG laser as signal wave. The BBO crystal used in the present experiment was 10 mm long, 6.5 mm high along the extraordinary axis and 5 mm wide along the ordinary axis, cut at an internal angle of 28.5° and AR coated with a single layer of MgF_2 for minimum reflection at 615 nm. The crystal was mounted with its ordinary axis horizontal on a precision rotation stage for adjustment of the phase

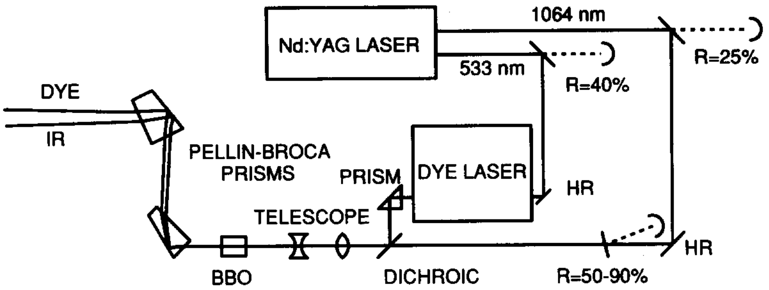

Fig. 2. Experimental setup for efficient difference frequency generation of ir light by nonlinear mixing of the output of a dye laser and the fundamental output of a Nd:YAG laser. The dye laser is pumped by part of the frequency doubled output of the Nd:YAG laser and its output is superimposed on a dichroic mirror with part of the residual fundamental light of the Nd:YAG laser. Subsequently both beams are telescoped down by a factor of 3 and directed through the BBO crystal which is mounted on a precision rotation state. Finally, the various wavelengths are separated in two Pellin-Broca prisms.

matching angle around an axis perpendicular to the direction of beam propagation and parallel to the table top.

The resulting tunable ir light (idler wave) was also horizontally polarized. Therefore, it suffered only negligible losses on the surfaces of the two fused silica Pellin-Broca prisms used to separate the three waves exiting from the BBO crystal. The energy of the generated idler wave was measured with a pyroelectric detector (Sensor Physics Model 510 [17]).

3. Method of alignment and experimental results

The detector of small amount of ir light is considerably more difficult than the detection of either uv or visible radiation. Since both the overlap of the two beams and the correct phase matching angle have to be adjusted simultaneously in the DFG process, we devised a procedure that allows a nearly complete separation of these two adjustments and detection in the uv and visible during the alignment.

The first step is to take advantage of the fact that the fundamental Nd:YAG laser radiation can be doubled in the BBO crystal at a phase matching angle of 22.78°. This is very close to the angle of 21.91° expected for DFG between the optimum dye laser wavelength of 564.0 nm and the 1.064 μm. This near coincidence is not fortuitous, since the difference frequency mixing of 532 nm light with 1.064 μm light to produce 1.064 μm light requires the same phase matching angle as the reverse process of doubling 1.064 μm light to produce 532 nm light. Furthermore it can be seen from fig. 1 that the change in the phase matching angle between the DFG of 1.064 μm and 1.200 μm is not large. The green light from the doubling process can readily be found with high efficiency. This gives an experimentally determined value for the tilt of the BBO crystal.

As a second step the dye laser output can be sum frequency mixed with the 1.064 μm light to produce 368.7 nm light at a phase matching angle of 30.30°. For this process to occur, the polarization of the dye laser beam must be rotated by 90° to make it an ordinary beam. This is accomplished by inserting a quarter wave plate (with negligible wedge) into the beam. A large amount of uv light is then readily found. Since the efficient generation of the uv light depends on the presence of both beams and a good overlap, one can optimize the overlap by maximizing the power of the uv light generated. In addition, this constitutes a second measurement for the crystal angle.

With the overlap of the beams established and calibration of the proper angular setting, the detection of the near ir difference frequency between the dye laser and the 1.064 μm light is straightforward. At most, slight tweaking is needed to optimize the output at the idler wavelength. From 42 mJ of dye laser light (at 564 nm) and 33 mJ of 1.064 μm light 4.5 mJ of idler light were generated. This is a quantum efficiency of 23%. The size of the idler was 7 mm at 2 m distance from the crystal and its shape appeared smooth and round, quite unlike the situation in Raman shifting where strongly distorted beam shapes are normally found [2]. Changing the dye laser wavelength and consequent readjustment of the phase matching angle gave the tuning curve shown in fig. 3; for comparison, the tuning curve of the dye laser is also given. This result shows that difference frequency generation in BBO is a highly efficient process.

Additional measurements were performed to characterize the difference frequency generation in BBO:

(i) For a fixed crystal angle the wavelength of the dye laser had to be changed by ± 0.9 nm to decrease the generated idler energy to 50%. This relatively low sensitivity should allow easy tracking of the phase matching angle during a scan of the dye laser and therefore the wavelength of the idler.

(ii) At fixed dye laser wavelength, the tilt angle of the crystal was altered and it was found that a 1.2 mrad change decreased the idler energy from 4.5 to 1.5 mJ. This value corresponds with the subjective impression that the phase matching angle can be adjusted manually without much difficulty. It allows the use of a moderately precise rotation stage for the DFG process in BBO. The divergence of the two input beams, by contrast, was on the order of 0.5 mrad.

(iii) The pulse energy of the 1.064 μm light was varied and the dependence of the generated idler energy measured. The result of this measurement is shown in fig. 4. Down to a signal energy of about 13 mJ no significant change in the output was observed. To understand this result, one must first realize that

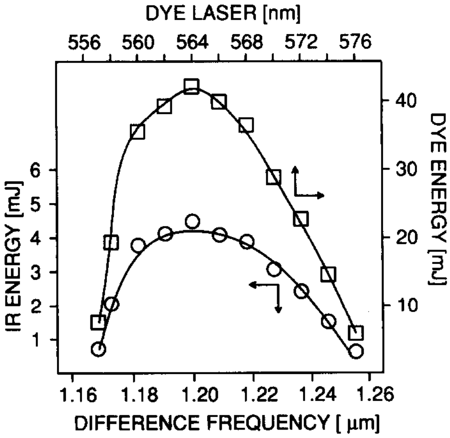

Fig. 3. Tuning curves for the output of the Rhodamine 6G dye laser (□) and the ir light (○) generated by mixing the dye laser output in a BBO crystal with 33 mJ of 1.064 μm light from the Nd:YAG laser.

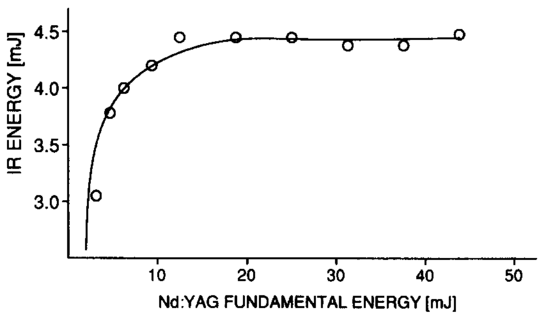

Fig. 4. Dependence of the generated ir light (1.20 μm) on the energy of the 1.064 μm signal input. The dye laser (pump) energy was 42 mJ at 560 nm.

for every idler photon produced one pump photon is split into a signal photon and an idler photon. Therefore, as the signal wave is amplified, the DFG process can also be viewed as optical parametric amplification (OPA) [18] of the signal wave due to the high intensity of the pump wave (dye laser). This concept was already realized in early work on DFG [19] and has recently been nicely demonstrated for DFG in $LiNbO_3$ [20]. Due to parametric amplification of the signal wave, its intensity will always increase to a maximum determined by overlap factors and other parameters of the experiment, independent (beyond a minimum level) of the signal input energy. Consequently the idler output will also be limited. To further corroborate this idea, the 1.064 μm light was attenuated to 5 mJ at the entrance of the crystal and the amount of 1.064 μm exiting the crystal was measured at full pumping by the dye laser. A parametric amplification of the signal wave by nearly a factor of 3 was indeed observed.

(iv) Since the DFG process is performed in saturation with respect to the signal input, very low shot-to-shot fluctuations are anticipated. The fluctuation on the input 1.064 μm beam was measured to be 2.35% (one standard deviation), that of the dye laser was 0.34% (probably due to saturation of the final amplifier), and finally the idler wave fluctuation was 3.4%. This value is much smaller than typical values for Raman shifting or direct generation of tunable ir light in an OPO.

4. Discussion

The experimental results presented in the preceding section show that difference frequency mixing in BBO allows the highly efficient generation of narrow band tunable ir light in the hitherto hard to access range of 0.9 to 1.5 μm. This was demonstrated using the output of a Nd:YAG pumped, visible dye laser and the residual fundamental output of the same Nd:YAG laser. Even though we have demonstrated this result only for Rhodamine 6G dye, there should be no doubt that neighboring dyes allow coverage of the whole range. Both the tuning curve shown in fig. 3 and recent work on $LiNbO_3$ [20] show clearly that the idler output in DFG depends nearly linearly on the pump energy as long as sufficient signal energy is supplied. The latter condition can be easily fulfilled since the fundamental output of the Nd:YAG laser is used as the signal wave. Somewhat lower output energy from dyes with smaller gain will therefore not drastically change the conversion efficiency. This is very different from alternative near ir generation processes, i.e. stimulated Raman shifting or optical parametric oscillation, where a fairly high threshold pump intensity is needed.

The observed quantum efficiency of 23% for the mixing process is already quite high but it could probably be increased further by optimizing the beam size and possibly by changing to a noncollinear mixing scheme with tangential phase matching. Thereby the effect of walk-off between the ordinary signal beam and the extraordinary pump beam in the BBO crystal could be lowered. We were not able to implement these changes due to the limited aperture of our crystal, which was purchased with a different application in mind.

Further increase in the amount of generated ir can also easily be obtained by suitable antireflection coating of the optics. Last but not least it might be advantageous to split off part of the fundamental Nd:YAG laser radiation before the frequency doubling needed to pump the dye laser. This is likely to provide a better spatial and temporal overlap than can be achieved with the depleted 1.064 μm beam left after efficient doubling.

The minimum length of BBO crystal needed for good conversion efficiency will have to be determined in future experiments and modeling.

5. Summary and conclusions

Difference frequency generation in BBO has been demonstrated to provide high power ns ir pulses in the range between 0.9 μm and 1.5 μm from a visible dye laser and the fundamental of Nd:YAG laser. A peak quantum efficiency of 23% has already been observed, and various possibilities to improve this value have been discussed. Due to the high damage threshold of BBO, scaling to even higher input energies seems quite feasible.

The frequency bandwidth of the generated tunable ir light should only be limited by the bandwidth of the dye laser if an injection seeded Nd:YAG laser is used. Therefore, the present scheme will readily allow the generation of ir light with a bandwidth on the order of 1 GHz with commercially available dye lasers. With high power amplified cw dye laser systems [21] a nearly Fourier transform limited bandwidth should be achievable, i.e. a bandwidth of about 100 MHz with a pump laser pulse length of 8 ns.

In summary, difference frequency generation in BBO seems to be the most convenient way to generate high power narrow band tunable ir light in the range from 0.9 to 1.5 μm. The relatively small sensitivity to changes of the crystal orientation and the input wavelength and furthermore the very low short fluctuations in the output, should prove that this scheme is an invaluable tool for optical experiments in this interesting wavelength range.

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