# High-power broadly tunable difference-frequency generation in proustite*

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High-power wavelength-tunable coherent radiation has been produced by generating the difference frequency between the output from a tunable narrow-linewidth ruby-pumped infrared dye laser and a $Q$-switched ruby laser in a proustite crystal. Peak infrared powers in the kilowatt range and wavelength tunability from 3.20 to 5.65 $\mu$ have been achieved. The experimentally determined infrared power tuning curves are compared to those predicted by theory for interacting Gaussian light beams.

Considerable emphasis has recently been placed upon the production of wavelength-tunable radiation in the midinfrared for spectroscopic and pollution detection applications. Semiconductor diode lasers[1] are capable of producing ultranarrow linewidths, but possess a very limited tuning range (typically less than 10 $cm^{-1}$), mode-hopping discontinuities in the tuning curves, and low power (a few tens of microwatts cw). The spin-flip Raman laser[2] has achieved powers of the order of 100 W with a narrow bandwidth, but its tuning range is limited by the magnitude of the applied magnetic field and emission is restricted to a wavelength band near the pump laser wavelength. Both of the above infrared sources require liquid-helium temperatures for operation.

Difference-frequency generation through the mixing of two laser beams in a phase-matched nonlinear crystal is capable of producing kilowatt-level powers, and by choosing one of the mixing lasers to be a wavelength-tunable organic dye laser, considerable tuning ranges in the midinfrared can be achieved. Dewey and Hocker[3] first demonstrated the usefulness of this approach by mixing radiation from a DTTC iodide organic dye laser with its 4-MW ruby laser pump in a $LiNbO_3$ crystal. They achieved peak powers of several kilowatts in the 3- to 4-$\mu$ range, but the $LiNbO_3$ crystal restricted generation to wavelengths less than 4.5 $\mu$. Hanna *et al.*[4] attempted to circumvent this restriction by utilizing a proustite crystal as the mixing medium. Generation of radiation from 2.5 to 13 $\mu$ is theoretically possible in appropriately oriented proustite crystals. Using a single-mode 290-kW ruby pump and a cryptocyanine dye laser, Hanna's group produced tunable radiation from 10.1 to 12.7 $\mu$ with 100 mW peak power, and a 6-W component at 4.9 $\mu$. Meltzer and Goldberg[5] produced infrared radiation tunable from 4.1 to 5.2 $\mu$ by mixing a DTTC iodide dye laser with its ruby pump in a lithium iodate crystal internal to the laser cavity. This arrangement is useful for enhancing infrared generation when limited pump power is available or when the nonlinear crystal possesses moderately low nonlinear coefficients. They obtained 100-W peak-power infrared pulses for a 4-MW ruby pump.

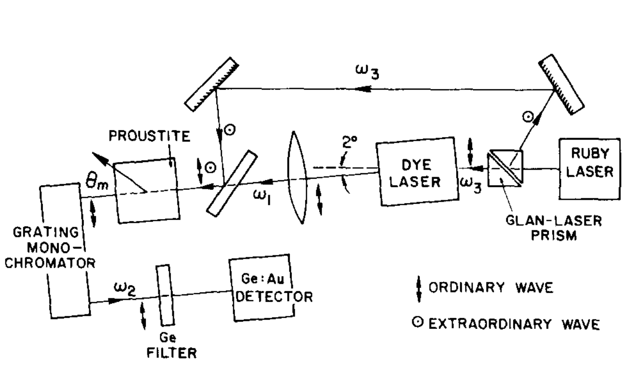


FIG. 1. Experimental arrangement for difference-frequency generation.

In this letter we report the production of infrared radiation continuously tunable from 3.20 to 5.65 $\mu$ with powers in the kilowatt range. The experimental arrangement is shown in Fig. 1. The ruby laser is $Q$ switched using a solution of cryptocyanine in acetonitrile and is internally apertured to a beam size of 2-mm diameter. This laser oscillated in the $TEM_{00}$ mode with a peak output power of approximately 900 kW in a 24-nsec pulse. The linearly polarized ruby beam is split into two beams of orthogonal polarization by a Glan-laser prism. The approximately 500-kW power of the horizontally polarized component pumps an efficient narrow-linewidth organic dye laser described elsewhere.[6] A longitudinal pumping configuration is used, with the dye laser beam making an angle of 2° with the pumping ruby beam. The

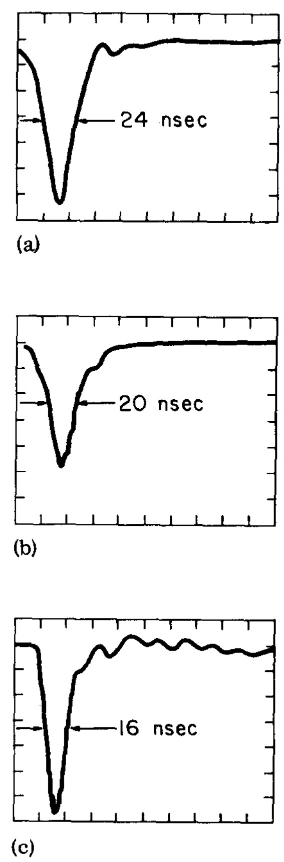


FIG. 2. Temporal output characteristics for (a) ruby laser, (b) dye laser, and (c) difference-frequency signal. Time scale is 20 nsec/div.

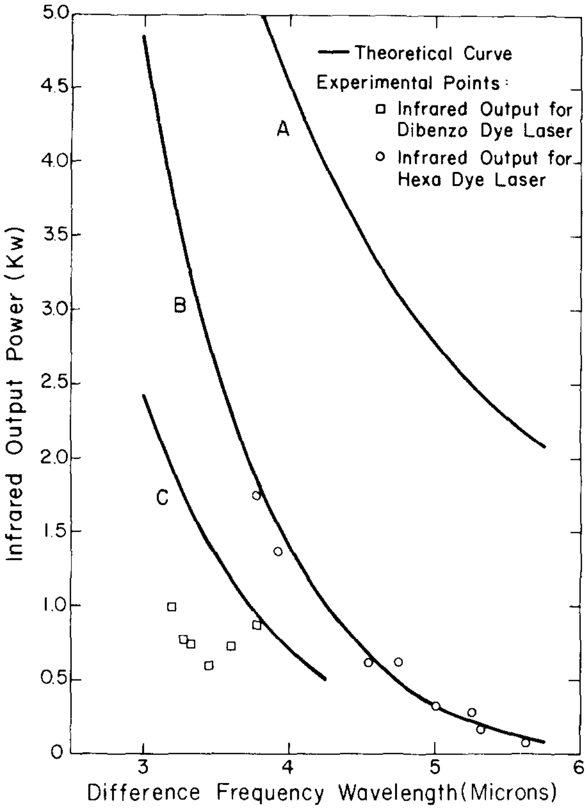


FIG. 3. Tuning plot of difference-frequency output power as function of wavelength. Curve A depicts the perfect phase-matching case ($\Delta k = 0$) for $P_1 = 40$ kW. Curve B shows $\Delta k \approx 4$ cm$^{-1}$ for $P_1 = 40$ kW, and curve C shows $\Delta k \approx 4$ cm$^{-1}$ for $P_1 = 20$ kW. $P_3 = 350$ kW in all three cases.

horizontally polarized output of this laser is recombined with the vertically polarized component of the ruby pump by a dichroic reflector. These two collinear beams impinge upon a 1-cm-long proustite ($Ag_3AsS_3$) crystal whose optic axis makes an angle of 38° to the crystal face. The crystal has been cut in such a manner that the nonlinear coefficients are additive.[7] Type 1 angular phase matching is used to achieve optimum parametric conversion. The infrared signal is generated at the difference frequency between the ruby and dye laser frequencies. Energy and momentum conservation for this process requires that $\omega_2 = \omega_3 - \omega_1$ and $\Delta \mathbf{k} = \mathbf{k}_3 - \mathbf{k}_1 - \mathbf{k}_2$, where $\Delta \mathbf{k}$ is the momentum mismatch and the subscripts 1, 2, and 3 refer to the dye laser, infrared difference frequency, and the ruby laser radiation, respectively. Wavelength tuning of the infrared radiation is achieved by simultaneous tuning of the dye laser and the corresponding phase-matching angle in proustite.

The ruby and dye laser radiation was monitored with silicon photodiodes (< 1-nsec time response). The mid-infrared difference-frequency radiation is detected by a calibrated liquid-nitrogen-cooled high-speed (time constant < 2 nsec) Ge:Au detector after filtering out the residual ruby and dye laser radiation with an antireflection-coated Ge filter. Figure 2 depicts the temporal behavior of these output signals. The infrared wavelengths were verified by interposing a grating monochromator between the proustite crystal and the Ge filter.

In the mixing experiments conducted, two different laser dyes were used. A $7\times10^{-5}M$ solution of 1, 3, 3, 1′, 3′, 3′-hexamethyl-2, 2′-indotricarbocyanine iodide (HEXA) in dimethyl sulfoxide (DMSO) produced nominally 40-kW dye laser pulses in the wavelength range 7900–8510 Å, and an $8\times10^{-5}M$ solution of 3, 3′-diethyl-2, 2′-(4, 5, 4′, 5′-dibenzo) thiatricarbocyanine iodide (DIBENZO) in DMSO produced approximately 20-kW pulses from 8500 to 8900 Å. Dye laser output power fluctuations of up to 35% under stable pump conditions are common as the laser is wavelength tuned over its effective lasing range.[6] The measured linewidth of the infrared dye laser was approximately 3 Å. Difference-frequency radiation tunable from 3.77 to 5.65 $\mu$ was achieved with the HEXA dye laser solution, and with the DIBENZO solution it is possible to extend the tuning range from 3.20 to 3.77 $\mu$.

The experimental results are shown in Fig. 3, along with theoretical power tuning curves derived from a parametric analysis for interacting Gaussian light beams.[8] Curve A depicts the power tuning curve for the case of perfect phase matching ($\Delta k = 0$). For this case the difference-frequency power generated (in mks units) is given by

$$P_2 = \frac{\omega_2}{\omega_1} P_1 T_1 T_2 T_3 \frac{W_3^2}{W_1^2 + W_3^2} \sinh^2\!\left(\frac{gl}{2}\right), \tag{1}$$

where $g$ is the nonlinear gain parameter given by

$$g = \left[4\left(\frac{\mu_0}{\epsilon_0}\right)^{3/2} \frac{\omega_1\omega_2 d^2}{n_1 n_2 n_3} \frac{P_3}{\pi W_3^2}\right]^{1/2}, \tag{2}$$

and where $d$ is the effective nonlinear coefficient; $n_3$, $n_2$, and $n_1$ are the refractive indices of proustite at the ruby, infrared, and dye wavelengths, respectively; $P_3$ and $P_1$ are the ruby and dye laser powers incident upon the crystal; $W_3$ and $W_1$ are the radii of the ruby and dye beams (a 30-cm–focal-length lens is inserted in the dye laser path to match spot sizes at the crystal); $\epsilon_0$ and $\mu_0$ are the free-space permittivity and permeability; and $l$ is the crystal length. $T_3$ and $T_1$ are the power transmission coefficients for the entrance face of the crystal at the ruby and dye wavelengths, and $T_2$ is the power transmission coefficient for the exit face at the generated infrared wavelength. In our experiments $T_1T_2T_3 = 0.68$, and $W_1 = W_3 = 1.5$ mm.

Tuning curves B and C were computed for the case of a nonzero phase mismatch resulting from the finite linewidth of the dye laser radiation and the fact that the ruby and dye beams are not collinear within the crystal except for normal incidence. For this case the infrared power generated is given by

$$P_2 = \frac{\omega_2}{\omega_1} P_1 T_1 T_2 T_3 \frac{W_3^2}{W_1^2 + W_3^2} \frac{g^2}{4s^2} \sinh^2(sl), \tag{3}$$

where

$$s = \pm\tfrac{1}{2}[g^2 - (\Delta k)^2]^{1/2} \tag{4}$$

describes the gain parameter. Curve B is seen to be in good agreement with the experimental infrared powers obtained by using the HEXA dye laser. The agreement is less pronounced for curve C. We attribute this to the pronounced variation in the output power of the less-intense DIBENZO dye laser. The over-all power conversion efficiency from ruby pump to difference-frequency power was the order of 0.4% in the 4-$\mu$ region.

The ruby power incident on the crystal was approximately 350 kW. This corresponds to a power density of 10 MW/cm$^2$, which is close to the reported damage threshold for a coated proustite crystal.[9] Further increases in infrared output power are thus possible only by increasing the power of the dye laser or by decreasing $\Delta k$.

The theoretical half-power gain bandwidth[10] was calculated to be 7.5 cm$^{-1}$. The observed spectral width of the infrared emission is approximately 8 cm$^{-1}$. Infrared generation was limited to 3.20 $\mu$ at the short-wavelength extreme by the limit of wavelength tunability of the laser dyes used. Generation at the long-wavelength extreme was limited to 5.65 $\mu$ because of the extremely large external angle (16°) of the mixing beams to the crystal normal in order to achieve phase matching internal to the crystal. For this case angular phase mismatch coupled with decreasing parametric gain places a limit upon further wavelength tuning. However, by use of appropriately cut proustite crystals, tunability from 5.65 to 13 $\mu$ should be attainable.

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[1]G.A. Antcliffe and J.S. Wrobel, Appl. Opt. 11, 1548 (1972).
[2]C.K.N. Patel, E.D. Shaw, and R.J. Kerl, Phys. Rev. Lett. 25, 8 (1970).
[3]C.F. Dewey and L.O. Hocker, Appl. Phys. Lett. 18, 58 (1971).
[4]D.C. Hanna, R.C. Smith, and C.R. Stanley, Opt. Commun. 4, 300 (1971).
[5]D.W. Meltzer and L.S. Goldberg, Opt. Commun. 5, 209 (1972).
[6]C.D. Decker and F.K. Tittel, Opt. Commun. (to be published).
[7]K.F. Hulme, O. Jones, P.H. Davies, and M.V. Hobden, Appl. Phys. Lett. 10, 133 (1967).
[8]A. Yariv and J.E. Pearson, Progr. Quantum Electron. 1, 1 (1969).
[9]D.C. Hanna, B. Luther-Davies, H.N. Rutt, R.C. Smith, and C.R. Stanley, IEEE J. Quantum Electron. 8, 317 (1972).
[10]S.E. Harris, Proc. IEEE 57, 2096 (1969).