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October 1970

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INFRARED DIFFERENCE-FREQUENCY GENERATION
USING A TUNABLE DYE LASER

by

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This research was supported in part by the Advanced Research Projects Agency of the Department of Defense, monitored by the Office of Naval Research under Contract No. N0014-67-A-0204-0040 and ARPA Order No. 322.

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ABSTRACT

Intense infrared radiation has been produced in the $3\mu - 4\mu$ region by generating the difference frequency between a wavelength-tunable dye laser and a Q-switched ruby laser. Saturation of the conversion of ruby photons to sum-and-difference frequencies was observed on a single pass through a phase-matched LiNbO_3 crystal. The system appears to be attractive as a simple, highly-efficient, and broadly-tunable infrared source.

Considerable interest exists in producing intense, wavelength-tunable radiation throughout the optical spectrum. Although notable success has been achieved in the near-ultraviolet, visible, and near-infrared using dye lasers¹ and parametric oscillators², the results of experiments at infrared wavelengths have been less spectacular.

Tunable far-infrared difference frequencies have been produced by Faries, Gehring, Richards, and Shen³, following earlier theoretical and experimental work by Laine⁴, Zernike, and Berman⁵, Yajima and Inoue⁶ and others⁷. To date, these efforts have been directed to the far-infrared ($\lambda > 100\mu$). In addition to difference-frequency generation, two important methods of using the Raman effect to produce infrared radiation have been reported recently. R. Pantell⁸ and co-workers at Stanford have used non-linear crystals and intense ruby light to yield simultaneous Raman-shifted visible light and infrared difference radiation. The resulting infrared radiation is continuously tunable over the spectral region 50 - 250 μ . Patel and Shaw⁹ have used a magnetically-tunable Raman shift to convert the output of a 10.6 μ CO₂ laser into a coherent beam between 10.9 μ and 13.0 μ with substantial efficiency. These two methods appear very promising.

Our efforts have been directed to producing intense infrared emission in the 2 μ - 20 μ spectral region. The optical system is shown in Fig. 1. The output of an unpolarized Q-switched ruby laser is divided into two parts by a beamsplitter. One beam is used to pump a wavelength-tunable dye laser. The output of the dye laser is then combined with the second part of the ruby beam by a dichroic mirror and the composite radiation is directed into a phase-matched LiNbO₃ crystal.

A silicon filter is used to block completely all emerging radiation at the ruby and dye wavelengths, while passing a substantial fraction of the infrared difference frequency. A liquid-nitrogen cooled PbSe detector was used to measure the infrared output, with appropriate attenuators being used to avoid saturating the detector.

The results of our initial experiments are presented in Figs. 2 and 3. The infrared power is seen to be (a) linearly proportional to the ruby power entering the crystal, and (b) independent of the power of the dye laser beam above a well-defined threshold.

The LiNbO_3 crystal was rotated to achieve phase-matching at an angle

$$\theta_{\text{IN}} \approx \sin^{-1} \left[\frac{n_o^p - n_o^d + (n_o^d - n_o^i)(\lambda_p/\lambda_i)}{n_o^p - n_e^p} \right]^{1/2}$$

where θ_{IN} is the angle from the optic axis at which phase-matching occurs (as measured inside the crystal). The superscripts (p, d, i) on the index of refraction, n , denote values at the pump, dye, and infrared wavelengths, respectively. Subscripts o and e refer to the ordinary and extraordinary indices of refraction. The incident radiation made a single pass through the nonlinear crystal; no optical cavities were employed. The experiment was conducted at ambient temperatures which were subject to random excursions of 3 - 5°C, with no apparent effect on the stability of the output amplitude.

The experimental data shown in Figs. 2 and 3 were obtained with a concentrated DTTC Iodide/DMSO dye solution in a transversely-pumped dye laser. The dye laser output could be varied from about 0.84μ to

0.89 μ by using a diffraction grating as the rear cavity reflector. Other dyes are available which cover the entire wavelength region 0.72 μ - 1.2 μ . An output mirror of 30% reflectivity was used on the dye cell. The spectral width of the dye laser radiation was roughly 3 - 5 cm^{-1} , although significantly narrower lines are feasible. Approximately 6 KW of infrared power was produced at the detector for an initial power of 4 MW of total unpolarized ruby radiation. We estimate that the conversion of available ruby photons (of the correct polarization) to infrared photons substantially exceeded 1%, in spite of visible curvature of the polished crystal faces. The spectral width of the infrared emission was less than 10 cm^{-1} . Because the dye laser operates only during the ruby pulse, synchronization of the two laser pulses is assured.

In order to confirm the infrared output wavelength and demonstrate the flexibility of the tuning arrangement, we scanned the 3.5 μ absorption band of an 0.2 mm thick sheet of polyethelene. The result is shown in Fig. 4. The data points are compared to an IR spectrometer tracing obtained on a standard dual-beam instrument. The wavelength shown for each data point was computed as the difference wavelength between the measured dye laser output and the ruby output at 0.6943 μ . The discrepancy at 3.69 μ is real; the spectrophotometer apparently did not resolve the transmission peak.

The present experimental system is limited to the spectral region below 4.5 μ by the transmission characteristics of LiNbO_3 . Proustite ($\text{Ag}_3\text{As}_5\text{S}_3$) and pyragyrite (Ag_3SbS_3) appear promising as useful non-

linear mixing media which will extend the available wavelength coverage to at least $13\mu^{10,11}$. The ruby second harmonic (0.347μ) and a continuously tunable dye laser operating from $0.4\mu - 0.7\mu$ would be capable of covering the wavelength region from 0.7μ to 2μ .

It should be noted that the available radiation at the dye laser wavelength is also increased by the nonlinear difference frequency generation. Both the near-infrared and infrared beams are available for spectroscopic purposes.

We are indebted to R.M. Hilton and the Air Force Cambridge Research Laboratories for supplying the LiNbO_3 crystal we used. This work was supported by the Advanced Research Projects Agency and Office of Naval Research.

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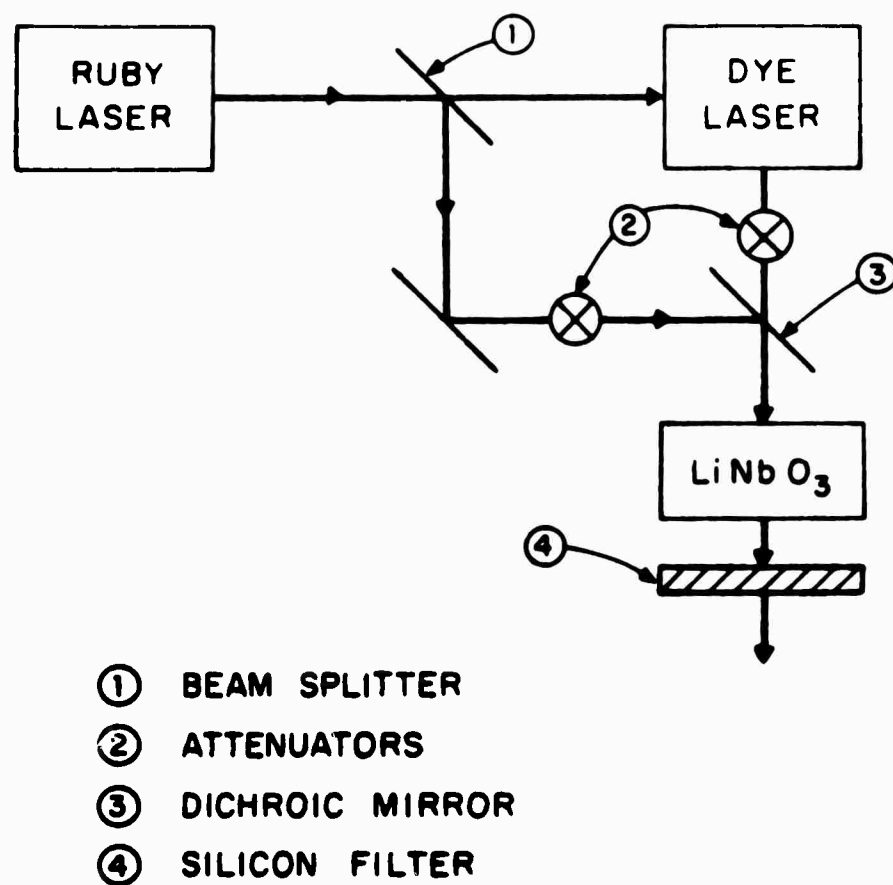


Figure 1. Experimental system for generating difference frequencies.

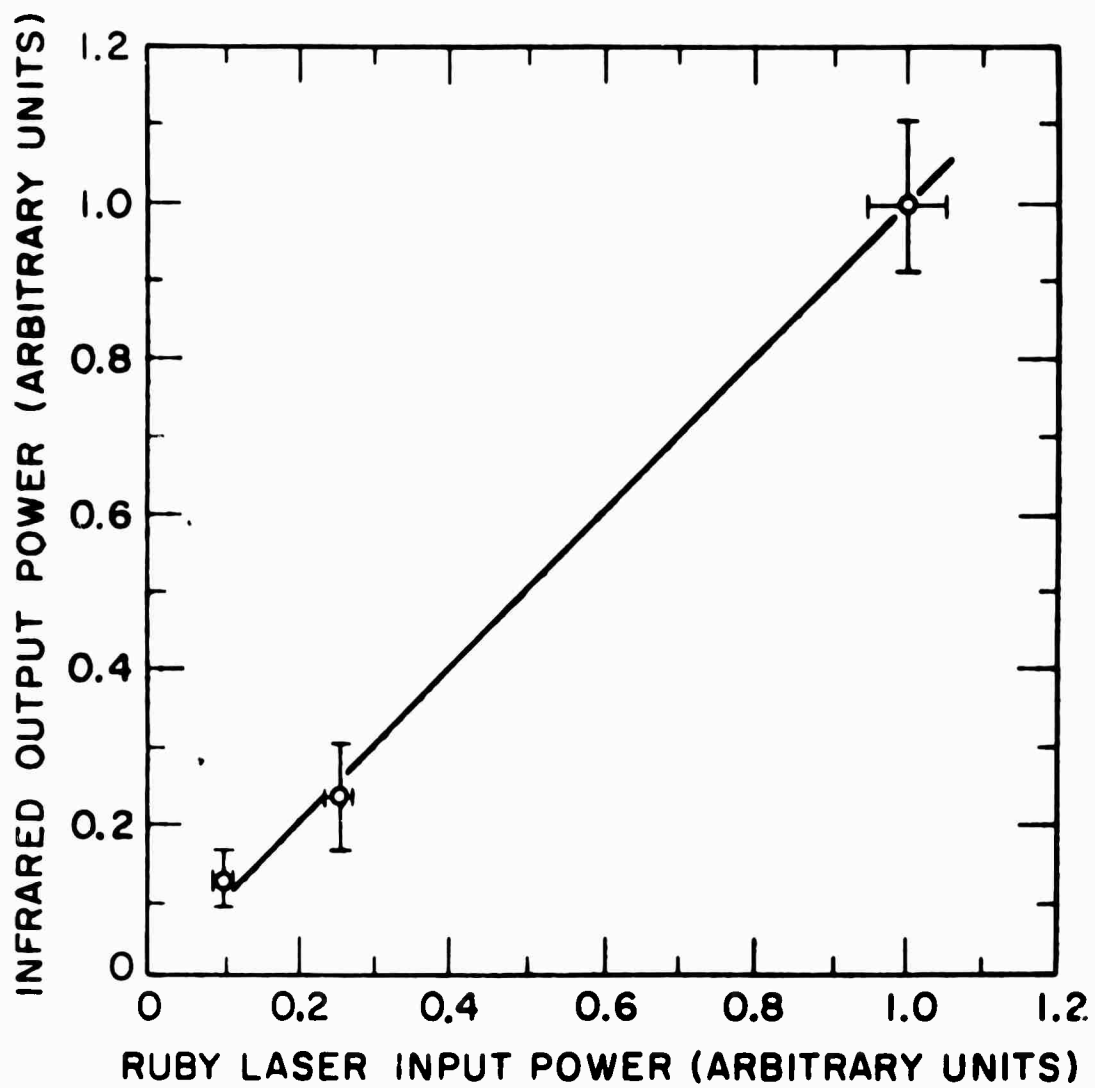


Figure 2. Infrared output power as a function of ruby laser excitation.

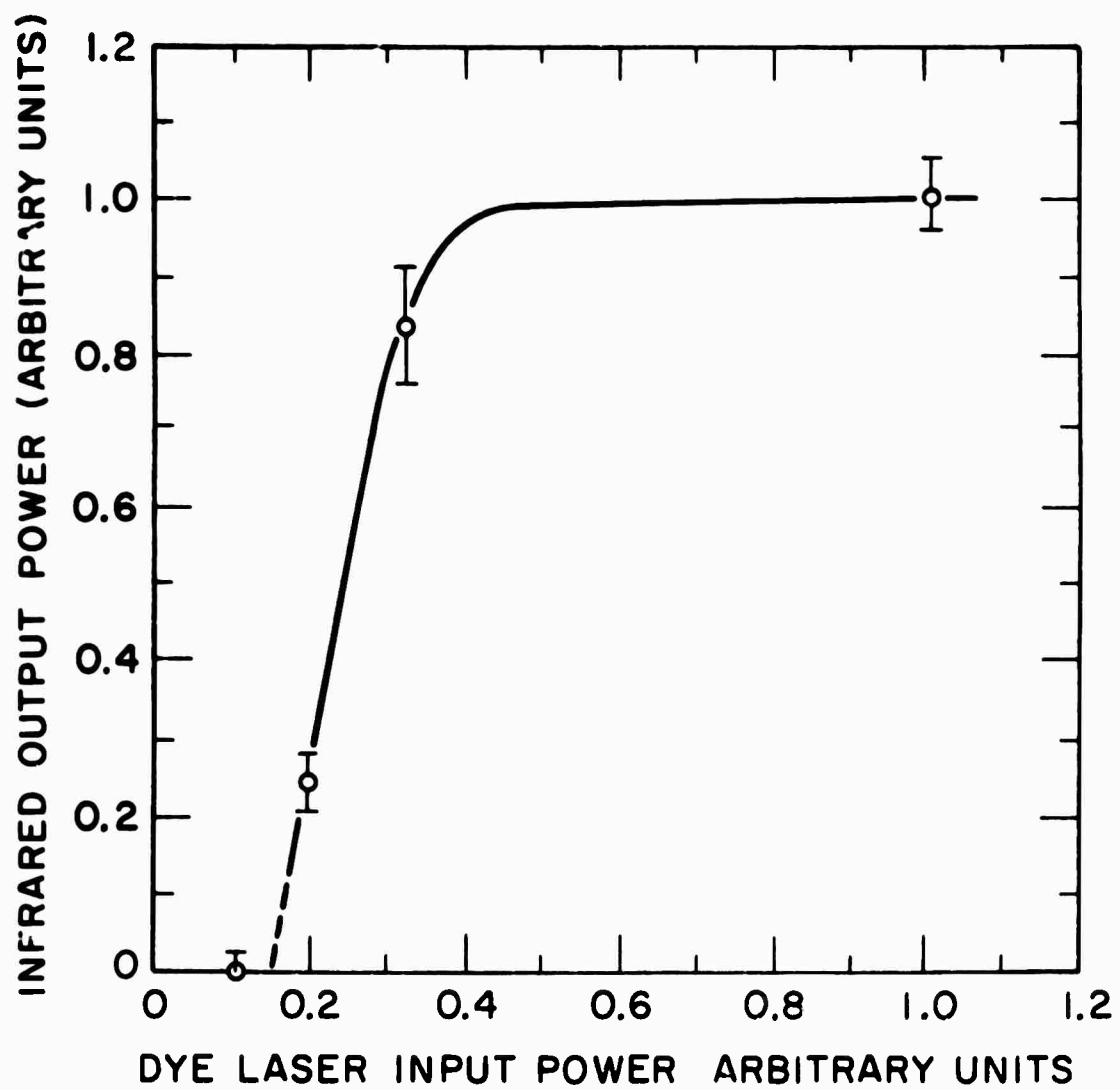


Figure 3. Infrared output power as a function of dye laser excitation.

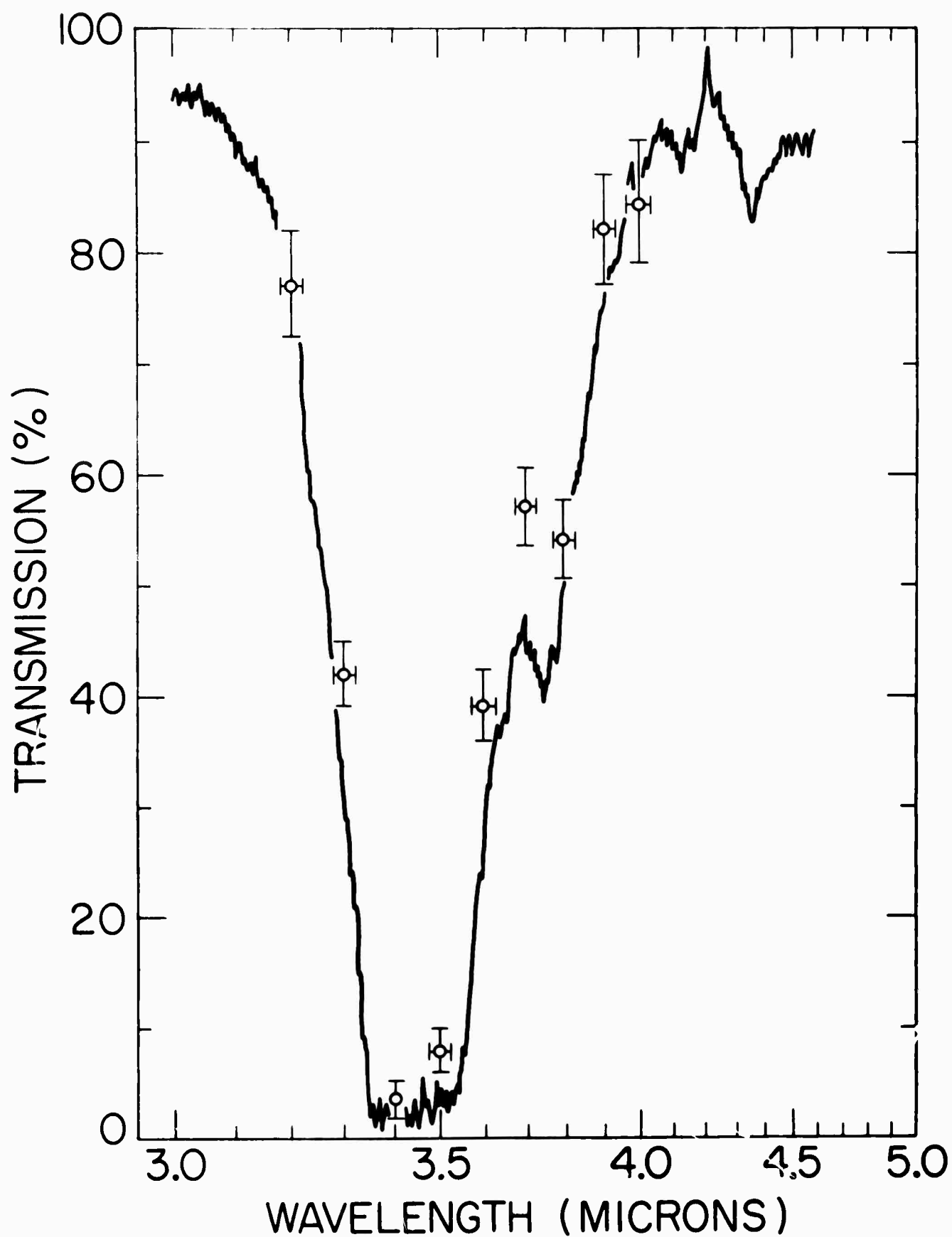


Figure 4. Absorption spectra of polyethelene. Solid curve: conventional dual-beam spectrophotometer. Data points: measured transmission of infrared emission from crystal with wavelength computed from the measured dye and ruby laser wavelengths.

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1 ORIGINATING ACTIVITY (Corporate author) M.I.T., Cambridge, Mass.		2a REPORT SECURITY CLASSIFICATION Unclassified	
		2b GROUP	
3 REPORT TITLE Infrared Difference-Frequency Generation Using a Tunable Dye Laser			
4 DESCRIPTIVE NOTES (Type of report and inclusive dates)			
5 AUTHOR(S) (Last name, first name, initial) C. Forbes Dewey, Jr. and Lon O. Hocker			
6 REPORT DATE October, 1970		7a TOTAL NO. OF PAGES 11	7b NO. OF REFS 11
8a. CONTRACT OR GRANT NO. b. PROJECT NO. ONR Contract No. N0014-67-A-0204-004 c. (ARPA Order No. 322) d.		9a ORIGINATOR'S REPORT NUMBER(S) Fluid Mechanics Laboratory Publication No. 70-11 9b OTHER REPORT NO(S) (Any other numbers that may be assigned this report)	
10. AVAILABILITY/LIMITATION NOTICES Distribution Unlimited			
11. SUPPLEMENTARY NOTES		12 SPONSORING MILITARY ACTIVITY Advanced Research Projects Agency Dept. of Defense and Office of Naval Research, Washington, D.C.	
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DD FORM 1 JAN 64 1473

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		ROLE	WT	ROLE	WT	ROLE	WT
	lasers infrared radiation difference-frequency generation dye lasers nonlinear optics						

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