TUNABLE LASERS

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We have produced continuously-tunable infrared radiation between 4μm and 21 μm by mixing the outputs of a ruby laser and a ruby-pumped dye laser in a single rotationally-twinned ZnSe crystal. The peak output power of 5 watts at 4μm is approximately 250 times larger than the power obtained from an untwinned ZnSe crystal of comparable length. This enhancement is attributable to randomly-spaced rotational twin planes perpendicular to the direction of propagation. Regularly-spaced twins should produce an enhancement proportional to the square of the number of twin planes.

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20. Abstract Continued

Preliminary work suggests absorption due to two photon carrier generation may be responsible for the early saturation of the IR conversion efficiency with pump laser intensity.

Twelve $\bar{4}3_m$ crystals are noted for possible application for 10μ frequency doubling using regularly spaced rotational twins to make them phase-matchable. Additional crystals suitable for frequency-doubling Nd-YAG lasers into the blue-green spectral region have been identified.

Further characterization of the wavelength tuning regions of new-infrared wavelength tuning regions of near-infrared laser dyes has been completed. The effects of optical pumping geometry have been studied for ruby-pumped dye lasers.

TUNABLE LASERS

Technical Report No. 1

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TUNABLE LASERS

ARPA Contract No. N00014-67-A-0204-0092

Technical Report No. 1

For period ending December 31, 1974

Summary

The production of tunable high power infrared radiation has numerous important applications, including laser induced chemical reactions, isotope separation, and high resolution spectroscopy. Our goal is to produce a powerful wavelength-tunable infrared laser system capable of operating throughout the 2μ → 20μ region of the spectrum.

Our work has emphasized nonlinear optical methods of transforming the high spectral brightness of fixed frequency and tunable dye lasers into regions of the infrared previously devoid of suitable high power wavelength tunable sources.

We have successfully demonstrated the use of rotationally-twinned nonlinear crystals to achieve periodic phase matching for ruby laser-dye laser difference frequency generation. The tuning range spanned the infrared from 3μ to 21.5μ using ZnSe. An enhancement of 250 in the infrared power was achieved with ZnSe containing randomly spaced twins. Experiments have been initiated to investigate the possibility of producing regularly-spaced rotational twins; regular twins should allow kilowatts of power to be generated across the entire 2μ-21μ region of the infrared.

A preliminary study of the saturation characteristics of the mixing efficiency suggests that free carrier absorption due to carriers produced by two photon excitation by the ruby and dye lasers is the cause of this saturation. This effect is of considerable importance as it may be responsible for the lower than expected conversion efficiency of proustite and other materials which have band gap energies less than two times the energy of the pump laser photons.

We have identified 12 suitable $\bar{4}3_m$ crystals for frequency-doubling CO_2 lasers using rotational twins to achieve periodic phase matching. These materials are also suitable for other nonlinear mixing experiments involving powerful laser pump sources in the 3μ-20μ region.

Further characterization of the wavelength tuning regions of near-infrared laser dyes has been completed. The effects of optical pumping geometry have been studied for ruby-pumped dye lasers.

TUNABLE LASERS

ARPA Contract No. N00014-67-A-0204-0092
Technical Report No. 1
For period ending December 31, 1974

Research Program

1) We have continued our investigation of the use of twin planes for reversing the sign of the nonlinear polarization in Zinc Selenide. In particular we have finished our initial study of the use of naturally twinned Zinc Selenide in a difference frequency generation experiment using a ruby laser and ruby-pumped dye laser. Particular attention has been given to absorption caused by carriers created by two-photon absorption.

2) A preliminary study of the suitability of a variety of $\bar{4}3_m$ materials for 10.6μ second harmonic generation has been made.

3) A series of commercially available dyes have been tested on an end pumped ruby-excited dye laser. This series gives coverage in the region between .834μ to 1.081μ. The latter figure is the longest wavelength achieved with a ruby-pumped dye laser.

Accomplishments

A) I.R. Difference Frequency Generation. A paper Enhanced Nonlinear Optical Effects in Rotationally Twinned Crystals by C. F. Dewey Jr. and L. O. Hocker has been submitted for publication by Applied Physics Letters. This paper reports difference frequency generation

in twinned ZnSe, work done during this reporting period and also the final six months of ARPA Contract DAHC04-71-0049. A preprint of this paper is included as Appendix I of this report.

Work has continued on the power saturation effect mentioned in the last paragraph of page 7 of Appendix I. This effect is of substantial importance as theory predicts that the conversion efficiency in the difference frequency generation experiment should improve as the input beams are more tightly focussed into the sample until either the bulk or the surface damage threshold of the material is reached. It was found that difference frequency generation appeared to saturate and even diminish as the power level was raised above 10 Mw/cm^2, long before the bulk or surface damage threshold for the material was reached. We feel that this saturation is due to absorption of the infrared radiation by the carriers produced by two-photon absorption of the ruby and dye beams. Since the absorption should be proportional to the carrier density; by measuring the conductivity of the sample during the laser pulse we can obtain a measure of the absorption. Figure 1A shows the conductivity of a ZnSe Crystal versus Ruby laser power density. The initial linear rise of the curve is probably due to one photon absorption from impurity levels. At power levels above about 1 Mw/cm^2 the conductivity increases as the square of the laser power and is accordingly due to a two-photon process.

The dependence of the radiated difference frequency power on the total Ruby laser and dye laser power density can be determined readily. Let us consider separately the two limiting cases of the carrier life-

time (τ) being much shorter than the laser pulse width (δ) and the carrier lifetime being much longer than the laser pulse width. In both cases we will consider the laser power outputs to be constant and vary the degrees of focussing. In the case with no absorption P_{IR} the infrared difference frequency power detected would be linearly dependent on the total laser power density P.

For the case $\tau << \delta$ the carrier density in the sample is local power density at the instant, and accordingly this absorption coefficient (γ) is given by

$$\gamma = a_1 P + a_2 P^2 \tag{1}$$

The infrared power outside the crystal of length L is then given by

$$P_{IR} \propto \int_0^L \frac{P}{L} e^{-(L-x)(a_1 P + a_2 P^2)} dx \tag{2}$$

We assume a square wave laser pulse so E_{IR}, the total I.R. energy produced is proportional to P_{IR}. Integrating (2) then gives

$$E_{IR} \propto P_{IR} \propto \frac{P}{(a_1 P + a_2 P^2)L} \left[1 - e^{-(a_1 P + a_2 P^2)L}\right] . \tag{3}$$

We know that $a_1 P \tilde{=} a_2 P^2$ at 2 Mw/cm^2 from Figure 1A, and accordingly besides the proportionality constant, the only unknown in equation (3) is a_2. We chose that value of a_2 that gives the peak of P_{IR} for a P of

10 Mw/cm^2. Equation (3) thus normalized is plotted in Figure 1B.

For the case $\tau >> \delta$ the carrier density is proportional to the time integral of the local power density; accordingly

$$\gamma(t) = \int_0^t a_1 P(t')dt' + \int_0^t a_2 P^2(t')dt' \qquad (4)$$

and

$$E_{IR} = \int_0^\delta \int_0^L \frac{P(t)}{L} e^{-(L-x)\gamma(t)} dx\, dt \quad . \qquad (5)$$

If we assume a square wave laser pulse, and recognize that the a_2P^2 term will dominate the absorption, equations (4) and (5) can be integrated to give

$$E_{IR} \propto P \int_0^\delta \frac{[1 - e^{-La_2P^2t}]}{La_2P^2t} dt \qquad (6)$$

This integral has a series solution

$$E_{IR} \propto \frac{P_o}{La_2P_o^2} \sum_{J=1}^{\infty} \frac{(La_2P_o\delta)^J}{J \cdot J!} \qquad (7)$$

Choosing the proportionality constant and $a_2\delta$ to give the peak energy out at the same laser input power as in the first case gives the dashed curve in Figure 1B. The sum in equation (7) converges slowly for $a_2P_o\delta >> 1$. For example the point at 30 Mw/cm^2 required 60 terms to give a 1% accuracy.

It is interesting that the two limits both predict a peak in the I.R. energy, in qualitative agreement with that observed in the laboratory. However, other experiments will be required to determine conclusively whether the carrier absorption is responsible for the early saturetion of the I.R. conversion efficiency.

B) $\bar{4}3_m$ Crystals Suitable for 10.6μ Frequency Doubling. Twelve $\bar{4}3_m$ materials were chosen for their appropriateness for 10.6μ frequency doubling. These materials along with their transmission ranges, refractive indices, 10μ doubling coherence length and nonlinear coefficients ere shown in Table I. The figure of merit shown in the last column is proportionel to the 5μ power one would expect to generete in if crystals were made of eech material with a fixed number of twins appropriately spaced. This tends to emphasize those materials that have long coherence lengths for the 10μ frequency doubling process. The unusual combination of a long coherence length and a high nonlinear coefficient makes GaSb look very promising for this process. Experimental work on these crystals awaits availability of suitable crystals.

C) Experiments on ruby laser pumped dye lasers have been conducted to determine the effects of dye laser pumping geometry on the tunable spectral range of each of several near infrared dyes. Three cavity geometries were compared: side-pumped, end-pumped, and angle-pumped. Comparison was made to flashlamp-pumped (with an elliptical cavity) operation reported by Webb et al. of Eastman Kodak. For Dye #940 (Kodak #IR-125), the results were as follows:

	Tuning Range (nm)	
Ruby Laser Pumped	In DMSO	In H_2O-Surfactant
Angle pumped	840-940	---
Side pumped	---	860-940
End pumped	847-967	860-930
Flashlamp Pumped		
Elliptical cavity	924-952	---

Grating-tuned operation to wavelengths as long as 1081 nm has been achieved.

TABLE 1

$\bar{4}3_m$ - Td MATERIALS

Material	Transmission Range (μ)	Refractive Index n	Coherence Length 10μ Doubling L_c (μ)	Nonlinear Coefficient* x 10^{12} mks d	Figure of Merit $\frac{d^2}{n^3} L_c^2$
GaSb	2 → 20μ (1	3.8 (5	134 (5	630 (5	130
CdTe	1 → 28 (3	2.7 (3	235 (8	86 (8	21
InAs	4 → 22 (1	3.5 (5	53 (5	420 (5	11.6
GaAs	1 → 17 (1	3.3 (5	104 (5	190 (5	10.9
ZnTe	.6 → 25 (9	2.7 (2	290 (8	47 (8	9.4
InP	.9 → 20 (6	3.5 (6	170** (6	170 (2	3.8
CuI	.5 → 20 (7	2.2 (7	450 (7	11 (7	2.3
CuBr	.5 → 22 (7	2.0 (7	370 (7	11 (7	2.1
ZnSe	.6 → 22 (3	2.5 (3	126 (8	40 (8	1.6
GaP	.6 → 12 (4	3.0 (5	46 (5	110 (5	0.95
AlSb	1.0 → 20 (6	3.3 (6	115** (6	50 (2	0.92
CuCl	.5 → 17 (7	1.9 (7	170 (7	9.5 (7	0.38

* Rel to GaAs Ref. 5 except for InP and AlSb.

** From index values.

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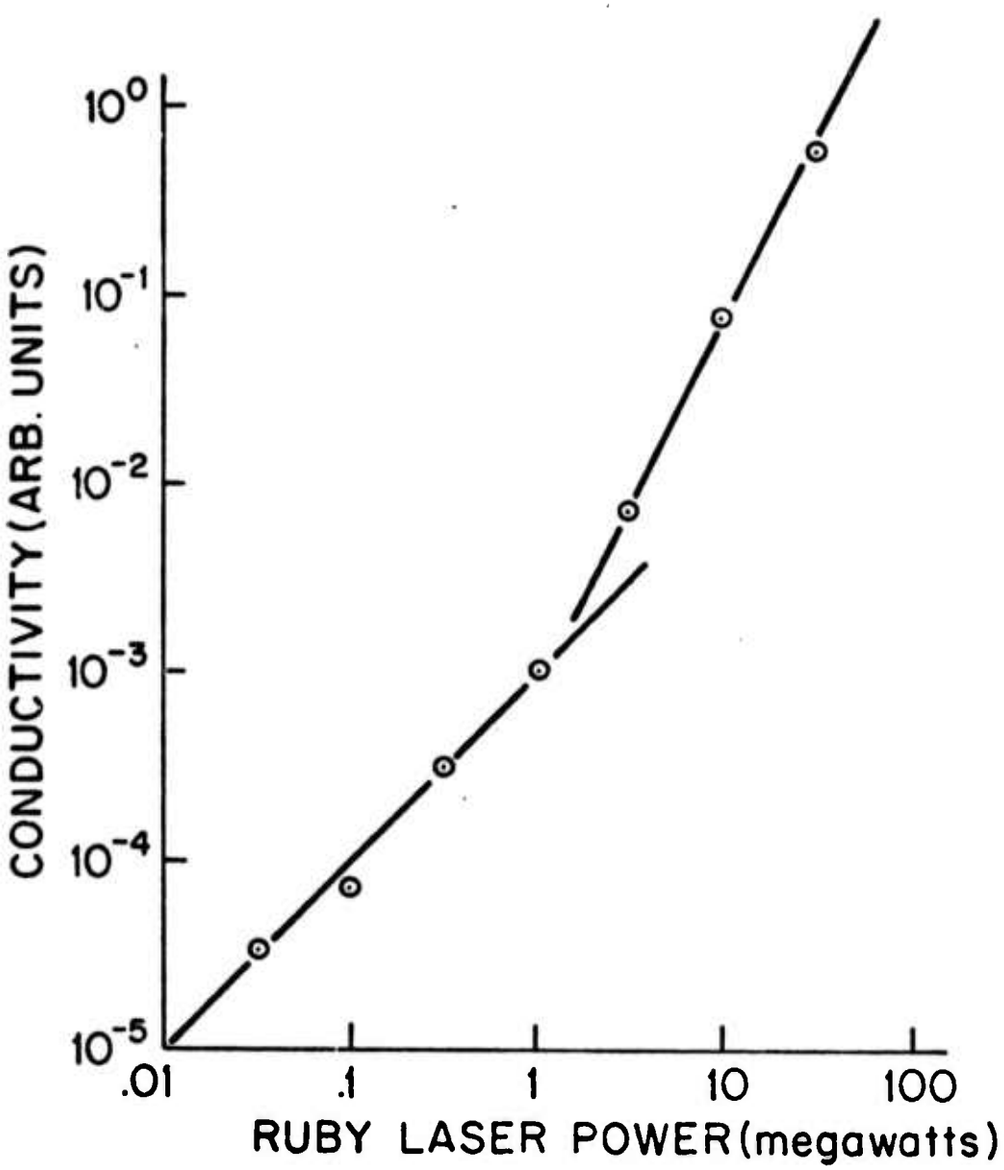

Figure 1A. Conductivity of ZnSe observed during laser pulse vs. Ruby laser power. Note the conductivity is proportional to P for low powers and P^2 for high power levels.

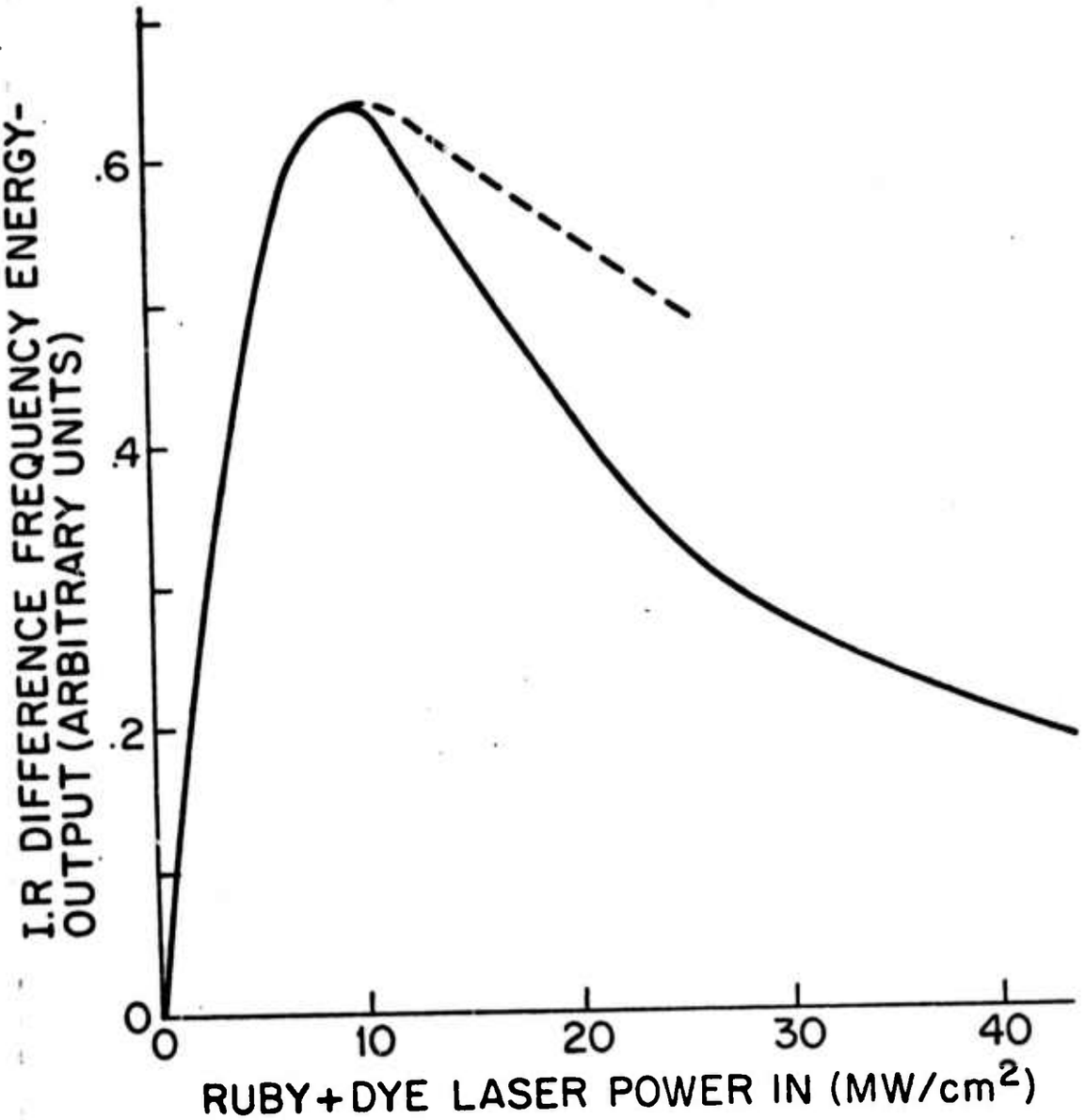

Figure 1B. Predicted dependence of IR difference frequency output on Ruby + Dye laser power for the cases: $\tau \ll \delta$ (solid curve) and $\tau \gg \delta$ (dashed curve). Both curves were normalized to give the peaks at 10 Mw/cm^2. The two curves overlap for power less than 10 Mw/cm^2.

APPENDIX I

ENHANCED NONLINEAR OPTICAL EFFECTS IN ROTATIONALLY TWINNED CRYSTALS*

by

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Abstract

We have produced continuously-tunable infrared radiation between 4μm and 21μm by mixing the outputs of a ruby laser and a ruby-pumped dye laser in a single rotationally-twinned ZnSe crystal. The peak output power of 5 watts at 4μm is approximately 250 times larger than the power obtained from an untwinned ZnSe crystal of comparable length. This enhancement is attributable to randomly-spaced rotational twin planes perpendicular to the direction of propagation. Regularly-spaced twins should produce an enhancement proportional to the square of the number of twin planes.

* Research supported by the Advanced Research Projects Agency under grants DAHC04-71-C-0049 and N0014-67-A-0204-0092.

The use of nonlinear optical materials to transfer the high spectral brightness of a laser from one wavelength to another has been studied in great detail.[1,2] Frequency doubling, parametric oscillation, difference-frequency generation, and other higher order processes in nonlinear crystals have received particular attention. In order that these processes be efficient, the nonlinear crystal must be transparent at the wavelengths of interest, have a high damage threshold, possess high nonlinear coefficient, and be phase-matchable for the mixing processes employed. Experience has shown that the necessity to satisfy all four criteria simultaneously severely restricts the choice of acceptable materials.

The most common method of achieving phase matching is to utilize nonlinear crystals exhibiting birefringence such that, by appropriate orientation of the crystal axes relative to the direction of propagation of the optical beams, the birefringence exactly cancels the dispersion over the length of the crystal. An alternative approach was suggested by Bloembergen in 1962[3] and investigated experimentally by Boyd and Patel[4] in 1966. In cubic crystals which have high nonlinear coefficients and damage thresholds as well as large regions of optical transparency, phase matching can be achieved by using a number of thin plates of the material oriented such that the sign of the nonlinearity reverses from plate to plate. A corollary technique is to construct a composite

material exhibiting periodic variation of the nonlinear optical coefficient[5,6,7,8]. These approaches have not been exploited to any significant degree. It has proved difficult to produce a large number of thin elements having the proper thickness, orientation, and optical quality.

We have recognized that there is a naturally-occurring phenomenon in cubic crystals which may be employed to achieve periodic phase-matching. Rotational twin planes can be described as the intersection of two segments of a crystal which are related by a 60° rotation about the 111 axis. An optical beam propagating in the 111 direction in a cubic crystal containing rotational twins perpendicular to the direction of propagation will experience a reversal of the direction of the nonlinear polarization at each twin boundary. This arises because the nonlinear polarization rotates twice as fast as the linear polarization, but in the opposite direction; i.e., a 60° rotation of the crystal leads to a 180° rotation of the direction of the nonlinear polarization. Thus an electric field applied perpendicular to the 111 direction produces a nonlinear polarization whose component perpendicular to the 111 direction points in opposite directions on the two sides of a twin plane. The nonlinear optical properties of a crystal containing rotational twins are equivalent to those of a stack of optically-contacted and oriented plates

of thicknesses equal to the twin plane spacing. Consequently, periodic phase matching of the type originally suggested by Bloembergen[3] can be achieved in a single crystal containing regularly-spaced rotational twins.

For a finite coherence length the net polarization in an untwinned piece of material follows the circle[9] shown in Fig. 1a. Here E_i is that component of the radiated electric field in phase with the nonlinear polarization at the beginning of the sample and E_q is the quadrature component. The position, θ, on the circle is simply $\pi x/\ell_c$ where x is the distance into the sample along the direction of propagation, and ℓ_c is the coherence length. Fig. 1b illustrates the case where twin planes occur at ℓ_c, $2\ell_c$, $3\ell_c$, ... At each plane the incremental field reverses sign, but continues to dephase at the same rate. The dotted line shows planes at $x = \frac{1}{2}\ell_c$, $\frac{3}{2}\ell_c$, $\frac{5}{2}\ell_c$ ..., the plate separation suggested as most desireable by Bloembergen. Inasmuch as the magnitude, not the phase, of $\vec{E}$ determines the nonlinear optical power, these two spacings give nearly identical results.

Normal methods of crystal growth produce rotational twins with uneven spacing. For twin separations of the same order as ℓ_c, but of otherwise random lengths, the electric field changes in the manner of a complicated random walk as in Fig. 1c. If the spacing is large compared

to ℓ_c, the electric field changes according to Fig. 1d. The magnitude of the net electric field after traversing N twin planes of random spacing larger than about 0.5 ℓ_c is $|\vec{E}| \cong (NE_o)^{1/2}$, where E_o is the electric field produced by a slab one coherence length thick. Inasmuch as the power radiated by the nonlinear polarization is proportional to $|\vec{E}|^2$, the nonlinear optical power radiated from a randomly-twinned sample will be greater than that radiated by an untwinned sample one coherence length thick by a factor equal to N. A crystal with regular twins separated by an integral number of coherence lengths would exhibit an enhancement of radiated power proportional to N^2.

To demonstrate the enhanced nonlinear effects which can be obtained in twinned crystals, we have generated infrared difference-frequency radiation in a piece of randomly-twinned ZnSe. The experiment employed a Q-switched ruby laser and a ruby-pumped dye laser as the two mixing beams in a configuration originally reported by us in 1971 The ZnSe crystal contained about 150 parallel twin planes perpendicular to the 111 direction with separations greater than 2μm, of which about half were greater than 20μm. The separations are measurable as the twin planes can be easily identified from the side of the crystal (see Fig. 2). The coherence length for these experiments ranged from 2μm to 10μm depending upon which IR difference frequency was being generated. We would therefore expect about a factor of $\sim 10^2$ improvement over an untwinned sample one coherence length thick.

The laser dyes used were DTTC, DOTC, and DTDC[11] in DMSO and aqueous-surfactant[12] solutions. Difference-frequency generation was observed over the wavelength range 4μm to 21μm, with a peak power of about 5 watts at 4μm and about 0.5 watts at 21μm. The decrease of power at longer wavelengths is attributable in part to a decrease in dye laser power from DTDC mixtures, and in part to a decrease in the number of twin planes larger than $\sim 5\ell_c$ ($\ell_c \sim 30\mu m$ at a wavelength of 21μm). The ratio of infrared power to dye laser power, P_{ir}/P_d, with random twins is proportional to $N\ell_c^2/\lambda_{ir}^2$; for ruby laser-dye laser difference-frequency generation in ZnSe, ℓ_c increases nearly linearly with λ_{ir} so that P_{ir}/P_d would be expected to be approximately independent of wavelength if the effective value of N were constant.

When an untwinned ZnSe crystal was substituted for the twinned mixing crystal, a peak power of 1 nW was observed at 4μm. The untwinned specimen was slightly wedged so that no Maker fringes were observed. The infrared power from an untwinned plane-parallel slab at a fringe maximum would therefore be 2 nW, or 250 times smaller than that measured using the twinned crystal. This value is in reasonable agreement with the enhancement factor of 150 predicted for a random walk process with 150 twins larger than $0.5\ell_c$.

Confirmation that the observed signal was indeed the difference frequency was done in the usual manner--by tuning the infrared output wavelength through the narrow passband of a multilayer dielectric filter, observing absence of the signal when either the ruby or dye beam was blocked, and seeing the disappearance of the signal when the IR wavelength was tuned into the ZnSe absorption edge at 22μm.

All of the above experiments were performed with a surface power density of about 10 MW/cm^2. Using this power density, an overlap area of 0.1 cm^2, the nonlinear coefficient of Soref and Moos[13], and the coherence length appropriate to 4μm mixing, we find that the untwinned crystal produced about 1/4 of the power predicted by theory. Such discrepancies are frequently observed in mixing experiments of this type. In this experiment, increasing the power density by focusing did not increase the IR power even though the input power density was substantially below bulk or surface damage thresholds. This could be explained if the IR power was being absorbed by free carriers produced by the intense ruby and dye beams. Carriers can be produced either by direct excitation of bound impurities or by two-photon excitations across the band gap.[14,15,16] Further investigation of this effect and its relation to second-harmonic generation is in progress.[17]

In conclusion, the effectiveness of using rotational twinning as a means for changing the phase of the nonlinear polarization inside the crystal has been demonstrated in ZnSe. Practical systems capable of producing large nonlinear optical conversion efficiencies must await the development of techniques for producing twins with controlled spacing. Additional cubic crystals which show promise for nonlinear mixing processes with both regularly and irregularly spaced twins include GaSb, CdTe, InAs, GaAs, ZnTe, InP, GaP, ZnS and the copper halides. We have also found that free carriers produced by two-photon absorption in some crystals may present a lower power limit for difference-frequency generation than either bulk or surface damage thresholds.

Acknowledgements

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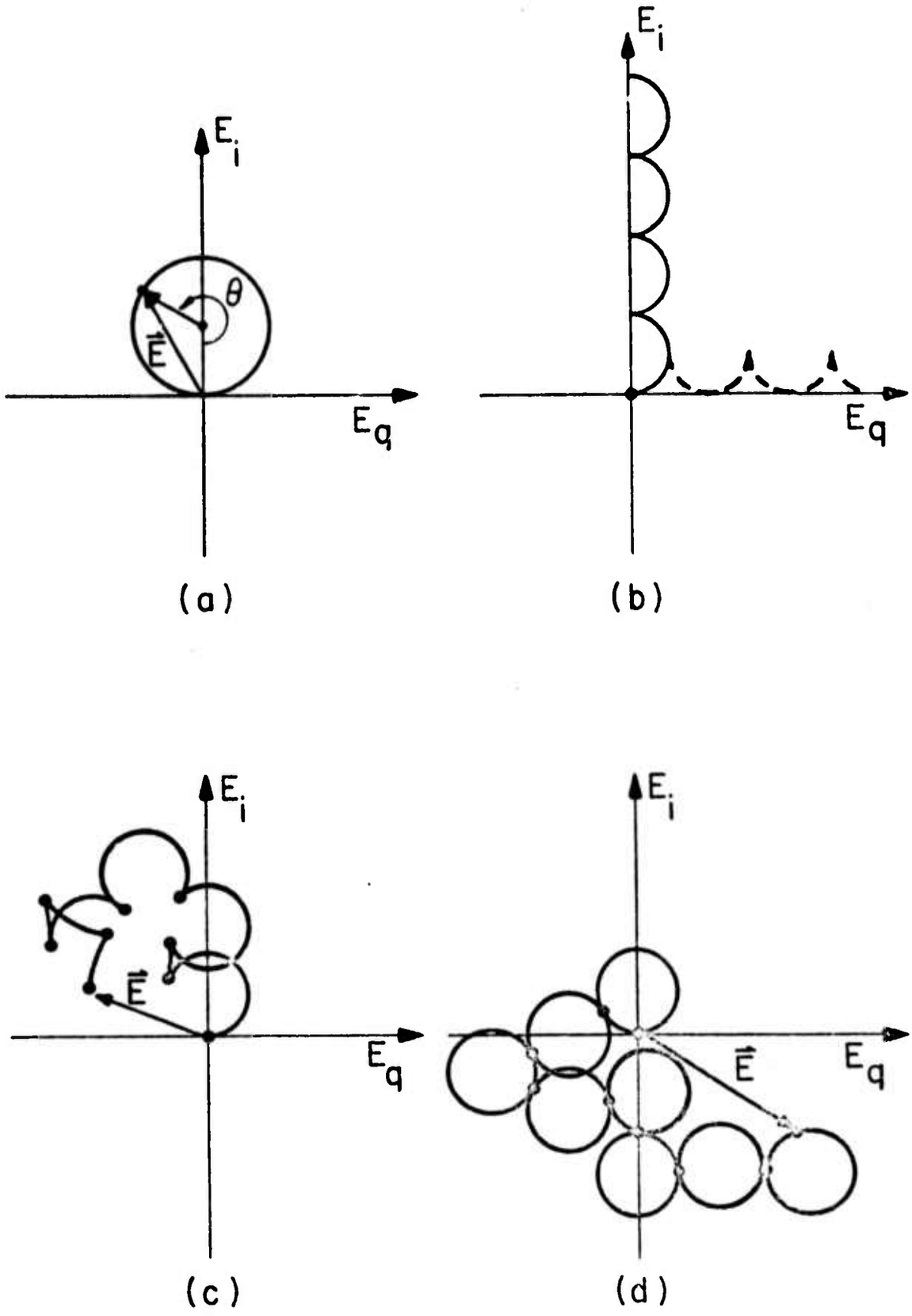
FIGURE CAPTIONS

Figure 1

Plot of amplitude and phase of the net difference frequency electric field for: a) untwinned crystal $\Theta = \frac{\pi x}{\ell_c}$; b) regularly twinned crystal with twin spacing = ℓ_c; c) randomly-twinned crystal with twin spacing < ℓ_c; d) randomly twinned crystal with twin spacing > ℓ_c. The distance along the path from the origin is proportional to the distance into the crystal.

Figure 2

Magnified section of the ZnSe crystal showing the twin planes. The 111 direction is in the plane of the figure and perpendicular to the twins.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100μm