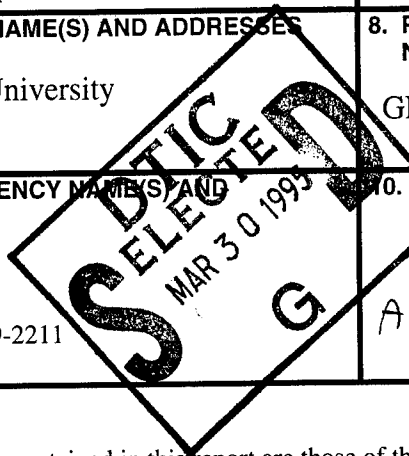


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**New Material for High Average Power
Infrared Generation: QPM-DB GaAs**

Final Report
for the period
1 September 1992 to 30 November 1995

Principal Investigator
Professor Robert L. Byer, Applied Physics Department

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Abstract

We have demonstrated diffusion-bonded-stacked GaAs to have good potential for high average power infrared generation. Diffusion bonding is the formation of a crystalline bond between two single-crystal wafers. Quasi phasematching is achieved in a stack of wafers of alternating orientation with the goal of retaining the low loss and high damage threshold of bulk single-crystal material. In this program we have substantially reduced loss per interface from 2% to 0.5% at 2 μm . This is adequate for single-pass applications in harmonic, sum and difference frequency generation, but further reduction of loss is required for resonant cavity applications such as optical parametric oscillation. We have performed studies of bonding techniques, surface preparation, optical damage thresholds, and numerical modeling of nonlinear frequency conversion. We are continuing this investigation with new characterization facilities and bonding apparatus and techniques developed in this program.

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**New Material for High Average Power
Infrared Generation: QPM-DB GaAs**

Table of Contents

Form 298	i
Title Page	ii
Abstract	iii
Table of Contents	iv
I. Introduction	1
II. Research Results	18
III. Scientific Personnel Supported by this Contract	26
IV. List of all Publications Supported by this Contract	26

New Material for High Average Power Infrared Generation: QPM-DB GaAs

Robert L. Byer

I. INTRODUCTION

We are developing the diffusion-bonded stacked (DBS) structure for quasi-phasematched interactions to meet the need for high power nonlinear conversions in the infrared. In our preliminary investigations, we have compared optical and thermal properties of some potential DBS materials. Theoretical projections of device performance were compared for DBS GaAs and ZnSe and birefringent crystals ZnGeP₂ and AgGaSe₂ for both second-harmonic generation (SHG) of 10- μ m radiation and 2- μ m pumped optical parametric oscillators (OPO's). We are refining bonding processes for GaAs and have initial diffusion bonding results for ZnSe. We have fabricated and tested DBS GaAs structures for SHG, demonstrating that the crystal orientation is conserved during the bonding process, and that the nonlinear generation of the individual layers sums coherently. These studies indicate that DBS materials have potential for application in high-average-power OPO's.

High-average-power coherent sources are needed throughout the infrared, especially in the 3 to 5- μ m region. Nonlinear frequency conversion of existing lasers can provide these sources. Optical parametric oscillators (OPO's) are attractive nonlinear devices because they can provide output tunable over a wide range; however, currently available infrared (IR) nonlinear materials, e.g. the chalcopyrites AgGaS₂, AgGaSe₂, and ZnGeP₂, are limited by low surface damage thresholds, large absorption coefficients, or low thermal conductivity^{1,2}. GaAs and ZnSe have high thermal conductivities and low absorption coefficients, and are widely used for windows and mirrors in high power IR laser systems. These cubic crystals also have large second order nonlinear susceptibilities; however, they cannot be birefringently phasematched, and, therefore, have not been used in practical frequency conversion applications.

An alternative to birefringent phasematching is quasi phasematching (QPM)^{3,4,5}, where a periodic modulation of the nonlinear susceptibility compensates for the phase velocity mismatch between the interacting waves. Stacks of discrete plates at Brewster angle have

been used to quasi phasematch second harmonic generation (SHG) in GaAs^{6,7} and CdTe⁸, but reflection and scattering losses associated with the many interfaces in the air-spaced layers precluded wide-spread application.

To eliminate the air-semiconductor interface problems, we diffusion bonded adjacent layers of GaAs. Diffusion-bonded stacked (DBS) GaAs devices were fabricated, and SHG of 10.6 μm has been demonstrated^{9,10}. Our goal now is the demonstration of a DBS optical parametric oscillator (OPO). We have investigated the different elements necessary in the realization of a mid infrared DBS OPO, and report on the progress in theory, bonding, devices and nonlinear testing.

A. Quasi Phasematching

QPM is achieved by a periodic modulation of the nonlinear susceptibility which compensates for the phase velocity mismatch between the interacting polarization waves, allowing efficient interactions in the absence of phase-velocity matching. Optimally, the modulation of the nonlinear susceptibility is a sign reversal, with a period Λ equal to twice the coherence length, L_c . The coherence length is the distance over which a π phase difference develops between the fundamental and the harmonic polarization waves, and is related to Δk , the wave vector mismatch due to material dispersion, by

$$L_c = \pi/\Delta k, \quad \text{where} \quad \Delta k \equiv k_1 - k_2 - k_3 = (n_1\omega_1 - n_2\omega_2 - n_3\omega_3)/c,$$

n is the refractive index, k is the wave vector, c is the speed of light in vacuum, and subscripts refer to quantities evaluated at the three interacting frequencies; we use the convention $\omega_1 > \omega_2 \geq \omega_3$. For efficient QPM, one of the spatial harmonics $K_m = 2\pi m/\Lambda$ of the periodically modulated medium must lie close to Δk , where m is an integer and is the order of the quasi phasematching. If this condition holds, quasi-phasematched nonlinear interactions behave similarly to conventionally phasematched nonlinear interactions, but with an effective wave vector mismatch $\Delta k_Q = \Delta k - K_m$, and an effective nonlinear susceptibility d_m equal to the amplitude of the m th Fourier component of the nonlinear susceptibility. For odd-order QPM ($m = 1, 3, \dots$) with 50% duty cycle, $d_m = 2d_{eff-b}/m\pi$, where d_{eff-b} is the effective nonlinear coefficient for the homogeneous bulk material. Detailed analyses of QPM appear in Refs.[4,5,11].

Developing appropriate means to periodically modulate the nonlinear susceptibility is the limiting step for practical devices for QPM interactions. Typical coherence lengths in

the visible are several microns and in the IR are several tens to hundreds of microns. Success in the creation of periodic domain structures in bulk and thin film oxide ferroelectrics has led to rapid progress in QPM for visible and near IR interactions (see Refs. in ⁵), but the transparency range of these materials precludes their use in the mid IR and far IR. Techniques for patterning the orientation of semiconductor films are emerging¹²; however, thin films are limited in their power handling capabilities. At present the only technique for producing bulk quasi-phasematched semiconductor media appears to be stacking rotated crystal plates. DBS structures permit the reduction of losses below those observed for air-spaced plates. While thin plates (250- μm) of LiB_3O_5 have been optically contacted with good results for QPM interactions¹³, the high indices of refraction of III-V semiconductors lead to significant losses for optical contacting wafers with practical polishing tolerances⁶.

B. Practical Devices

The established high power lasers in the infrared are: Neodymium at 1 μm , and CO_2 at 10 μm . Holmium and Thulium based lasers are also being developed at 2 μm . There exists a variety of semiconductor lasers in the infrared; however, their current lower powers prevents their use in high conversion nonlinear applications. We have identified options to produce wavelengths in the 3-5 μm region working from these sources: a 10- μm pumped SHG, a 1- μm pumped OPO, a 1- μm pumped OPO which then pumps a difference frequency generator (DFG) or OPO, and a 2- μm pumped OPO. Each of these devices utilizes DBS materials. SHG is simple to demonstrate; however, the resulting wavelengths are limited by existing sources. Similarly, DFG demonstration is also relatively simple, but limited for wavelengths unless pumped by an OPO. OPO's can provide an enormous range of wavelengths; however, gain and loss is critical. There must be sufficient gain to allow the OPO to reach threshold during the pulse for a pulsed pump, or the loss must be low enough to permit the OPO to reach threshold below the damage threshold of the crystal in the continuous regime. The first DBS OPO demonstrated will probably be a singly resonant OPO (SRO) in the pulsed regime.

We need to produce both fixed wavelength and tunable sources. QPM tunability can be achieved in three different ways. As with birefringent crystals, DBS structures may be tuned by temperature and angle; however, this only provides coverage over narrow-spectral regions. DBS crystals may also be fabricated with intrinsic tunability. If the layers are wedged, the crystal may be tuned by translating it perpendicular to the incident beam, thus

increasing or decreasing the coherence length. The wedged structure may provide greater tuning range than either angle or temperature tuning, and it does not introduce alignment problems such as those associated with rotating the crystal.

There are many developmental stages in the realization of a DBS OPO. We need to choose a material with appropriate optical characteristics, develop a diffusion bonding process for that material, fabricate a device with modulated nonlinear domains and verify that we do not degrade the domain orientation or the crystal quality during the fabrication process, demonstrate nonlinear conversion for SHG and DFG, and finally demonstrate nonlinear conversion for parametric oscillation and amplification where low loss is critical.

1. Materials

Useful DBS materials must have low absorption for the interaction frequencies, and have an adequate nonlinear coefficient. They need to be diffusion bondable, and have a workable coherence length for the desired interaction. For high peak power and high average power, the crystals need a high damage threshold and large thermal conductivity, respectively.

Table I compares the optical and thermal properties of III-V and II-VI semiconductors. Values for many of the properties, *i.e.* nonlinear coefficient and absorption, vary by factors of three or more depending on the references. We have chiefly used four different references^{14,15,16,17} comparing the results obtained for the same materials.

The ideal material will depend on the application. The dispersion properties of some potential DBS materials are shown in Fig. 1. These curves were generated using dispersion equations given by Pikhtin and Yas'kov¹⁸. The regions of low dispersion near the center of the curves indicate the useful spectral region for that material. The flatter the dispersion curve, the longer the coherence length, and the fewer bonds necessary for a given length of crystal. The coherence lengths for different nonlinear interactions determine the feasibility of the DBS structure. The longer the L_c , the easier the wafer is to polish and handle, and the more tolerant the final device is to absolute thickness errors.

TABLE I. PROPERTIES OF SOME NONLINEAR SEMICONDUCTORS

properties	GaAs	GaP	InSb	InAs	InP	ZnSe	β -ZnS
nonlinear coefficient (d_{14}) (pm/V)	83 ¹⁴	37 ¹⁴	500 ¹⁵	300 ¹⁵	140 ¹⁵	50 ¹⁵	25 ¹⁵
transmission range (μm)	1-16	0.6-11	8-25	3.8-7	1-14	0.5-22	1-11
dn/dT ($^{\circ}\text{K}$) @ 10 μm	1.5×10^{-4}	1.0×10^{-4}	1.5×10^{-4}	2.9×10^{-4}	8.2×10^{-5}	6.8×10^{-5}	4.1×10^{-5}
$(1/L)dL/dT$ ($^{\circ}\text{K}$) ¹⁾	5.7×10^{-6}	5.3×10^{-6}	5.0×10^{-6}	5.3×10^{-6}	4.5×10^{-6}	7.0×10^{-6}	6.7×10^{-6}
Thermal conductivity K (W/m $^{\circ}\text{K}$)	52	110	36	50	70	18	27
n @ 10 μm	3.27	2.96	3.95	3.42	3.05	2.41	2.20
α (cm $^{-1}$) @ 10 μm	0.01	0.21	0.009	20	<0.1	0.0005	0.15

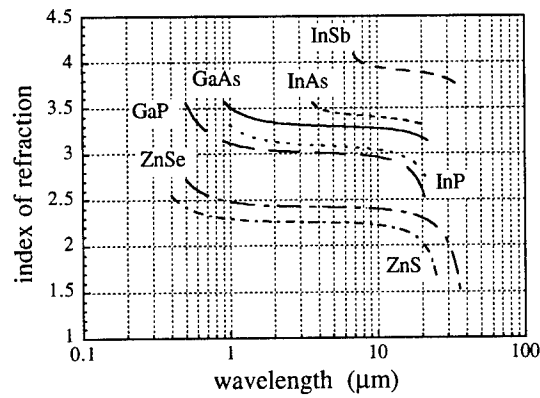


Fig. 1. Dispersion curves for III-V and II-VI semiconductor materials.

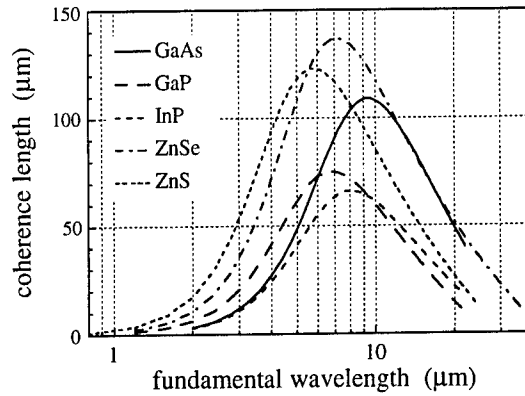


Fig. 2. Coherence lengths for second harmonic generation of the mid IR in GaAs, GaP, InP, ZnSe, and ZnS.

Coherence lengths for SHG, calculated from the dispersion relationships, are shown as a function of fundamental wavelength in Fig. 2. The materials GaAs, ZnSe, and ZnS are of particular interest because of their longer coherence lengths.

Figures 3 and 4 show calculated tuning curves for difference frequency generation (DFG) in DBS GaAs and ZnSe. These tuning curves are more complicated than those for SHG because it is necessary to specify two input wavelengths for DFG. Here we assume that the input waves for DFG are the signal and the idler generated in an OPO pumped at 1.064 μm . The signal and the idler wavelengths are designated by λ_s and λ_i . The output wavelength from the DFG is λ_d . Because the coherence lengths are short, these devices would be more easily fabricated for third order QPM.

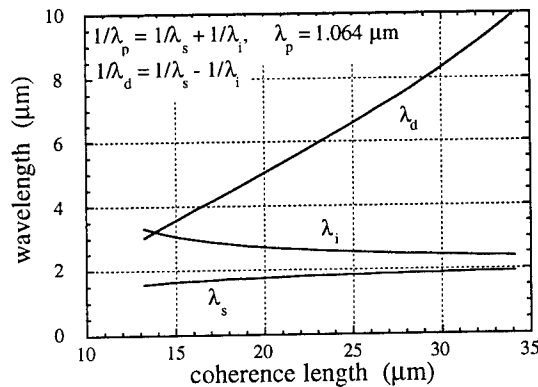


Fig. 3. Difference frequency generation in DBS GaAs for mixing the signal and idler from a 1.06- μm -pumped OPO. The output wavelength, λ_d , vs the coherence length is shown.

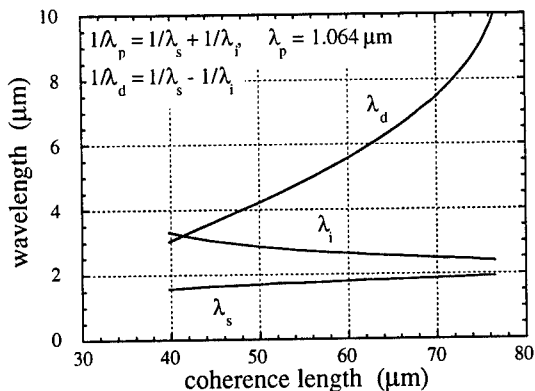


Fig. 4. Difference frequency generation, λ_d , for DBS ZnSe when pumped by the signal and idler from a 1.06- μm -pumped OPO.

For shorter wavelengths, approaching 1- μm , the dispersion increases for most of the semiconductors, and the coherence lengths decrease making it more difficult to fabricate devices; therefore, we have considered only the 2- μm pumped OPO option which we present in Figs. 5 and 6. For these calculations, we have assumed that the DBS OPO's are pumped by the output of a 1- μm pumped OPO operating at degeneracy.

We selected GaAs for the initial demonstration, because GaAs has the lowest absorption over the widest transmission window apart from ZnSe, and because inexpensive high-quality single-crystal wafers are commercially available. Although ZnSe shows promise due to its very low absorption, single crystal ZnSe is only now becoming available and is still very expensive.

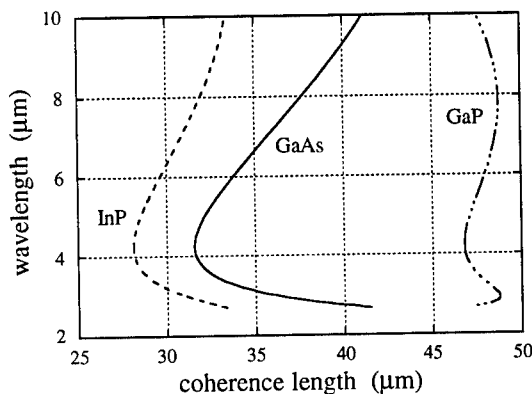


Fig. 5. Parametric oscillator tuning curves for III-V semiconductor compounds for a 2.13- μm pump wavelength.

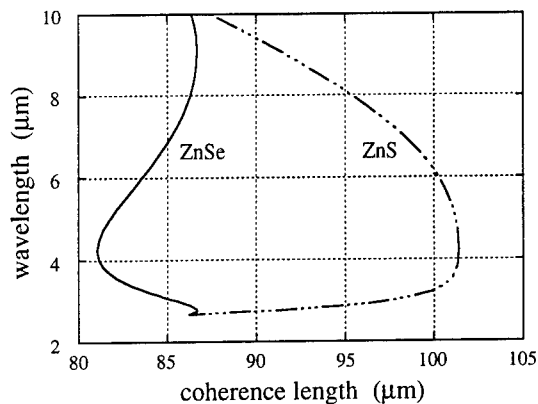


Fig. 6. Parametric oscillator tuning curves of II-VI compounds ZnSe and ZnS. The low absorption loss of ZnSe may allow very high average power parametric oscillator devices when pumped at 2.13 μm .

2. Diffusion Bonding

Diffusion bonding enables permanent bonding between surfaces without disturbing the crystalline structure, and without introducing another material (*i.e.* glue) between the surfaces. Adjacent surfaces are brought into close contact under pressure, and heated in an appropriate atmosphere to allow diffusion across the interface, creating a monolithic structure which is stable when exposed to thermal gradients (unlike optical contacting) and essentially indistinguishable from a bulk crystal. Diffusion bonding has previously been used both for joining dissimilar semiconductors for optoelectronic devices^{19,20,21,22,23,24}, and for fabricating laser slabs with nonuniform doping^{25,26}. For a DBS crystal there is ideally no difference in linear optical properties between the bond and the bulk material, nor any change of index of refraction at the interfaces.

There are two different mechanisms or stages in diffusion bonding. The first is the diffusion process: two surfaces are placed very closely together permitting electrons and atoms to move from one surface to the other. This process is not dependent on pressure, but is dependent on temperature and time. The atoms cannot move unless there are adjacent vacancies, and they are equally likely to diffuse in any direction. The second stage involves mass transport. The wafer is deformed by pressure to force all the surfaces into contact. The point at which this deformation occurs is called the yield point, and depends on temperature, pressure, and the number of dislocations²⁷. This deformation will increase the stress, and the number of dislocations in the crystal. It is not yet known how this will affect the optical properties of a DBS structure.

We have experimentally diffusion bonded a variety of undoped and lightly doped GaAs wafers. They were all mechanical grade and polished on both sides. The wafers were diced into 1-cm² pieces. The pieces were cleaned thoroughly with trichloroethane, followed by acetone and finally methanol; they were then stacked in a boron nitride holder between graphite spacers. Pressure was applied with a 1-kg weight, and the assembled stack was placed in an oven with a 5% H₂ and 95% N₂ atmosphere. The temperature was ramped to 840° C over 1 hour. After maintaining that temperature for two hours, the oven was cooled to room temperature over approximately 8 hours.

Stacks with 2 through 9 layers were bonded into monolithic units. The samples cleaved along the crystal planes leaving the bonded surfaces intact. We were able to bond wafers regardless of their dopings, their alignment of the crystalline axes, and their orientations: {100} to {100}, {110} to {110} and {110} to {100}. The exterior surfaces of the stack, which were in contact with the graphite spacers, were noticeably degraded after the processing. We did not repolish these surfaces, as the resulting uncertainty in the layer thicknesses would have hampered analysis of the SHG results.

We have succeeded in diffusion bonding polycrystalline ZnSe under similar conditions to GaAs. We expect polycrystals and single crystals to have similar bonding characteristics. When single crystal ZnSe growth is better characterized, we will be able to transfer the DBS technology developed for GaAs to ZnSe. The key parameters in diffusion bonding are: surface preparation, applied pressure, temperature and time. We are currently studying the quality of the bonds as a function of these parameters. By considering the melting points, vapor pressures, surface mobilities and yield points, we should be able to transfer bonding processes from one compound to another.

3. DBS Nonlinear Structure

Both GaAs and ZnSe are $\bar{4}3m$ crystals where the maximum nonlinear coefficient is for radiation polarized along a $\langle 111 \rangle$ direction. The {110} wafers were chosen for the nonlinear studies because they provide the maximum effective nonlinear coefficient for propagation normal to the input face. Adjacent wafers were rotated by 180° to alternate the sign of the effective nonlinear coefficient.

Figure 7 shows a schematic of the basic DBS structure. Figure 8a shows a structure with {100} oriented cap layers. For this orientation, there is no effective nonlinear coefficient at normal incidence. Since the thickness of these cap layers is not important, this structure

could be repolished, without compromising the conversion efficiency. The DBS structure can be built into laser output couplers, as shown in Fig. 8b. This would reduce interface loss. This configuration would be especially suited to SHG of a CO₂ laser, because the DBS structure could be made of the single crystal version of the polycrystalline coupler material. Figure 8c shows the wedged DBS structure which allows wavelength tuning by translation instead of by rotation. Caps could be added to this structure eliminating any overall linear wedge, thus avoiding beam steering effects. The wedged DBS structure would permit larger tuning regions than comparable birefringent crystals because the

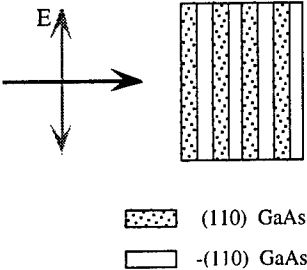


Fig. 7. Basic DBS Structure in GaAs showing the layers of alternating nonlinear coefficients

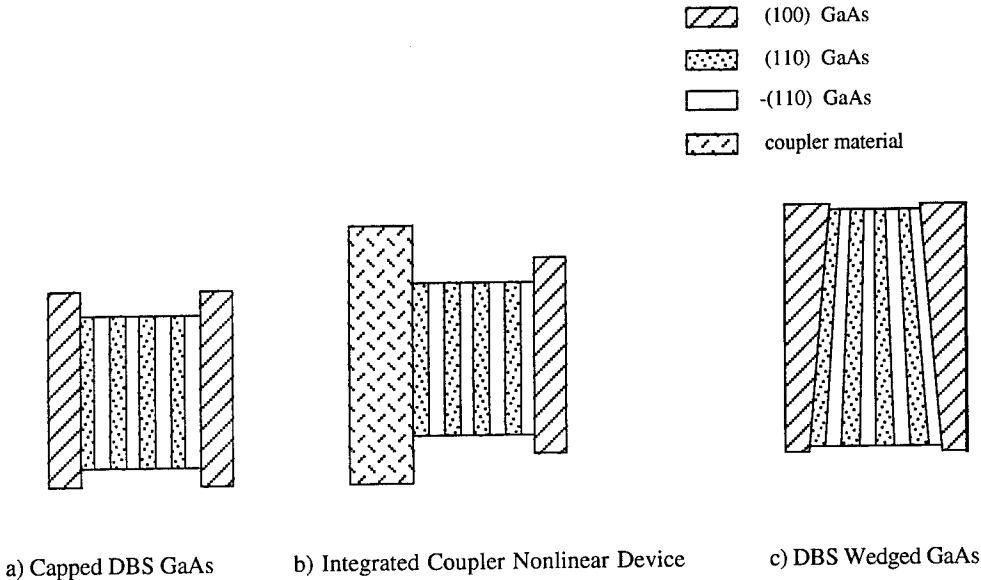


Fig. 8. Variations on DBS structure in GaAs

incident beam remains normal to the crystal at all times. Such structures would be particularly useful for optical parametric oscillators and mixers. We have fabricated both the basic structure and the capped DBS structure.

The DBS structure has a number of benefits compared to birefringently phasematched nonlinear crystals. DBS GaAs has very large acceptance bandwidths due to noncritical phasematching and high index of refraction with low dispersion¹⁸. For a 1-cm-long DBS GaAs structure for SHG at 10.6- μm (~94 layers), the FWHM wavelength acceptance is 0.5- μm , and the temperature acceptance is 270°C. The external angular acceptance is 64° FWHM. Since GaAs is not birefringent, there is no birefringent walk-off at any incidence angle, and QPM produces no phase-velocity walkoff at normal incidence.⁵

4. Nonlinear Devices: Predictions

It is useful to compare the theoretical projections of potential DBS materials such as GaAs and ZnSe to those of established birefringent nonlinear crystals such as ZnGeP₂ and AgGaSe₂. These comparisons are presented give indications of material performance under similar conditions. If we disregard absorption, which is appropriate for pulsed operation, we obtain figures of merit that are the same order of magnitude for all the materials considered. However, absorption and thermal properties must be considered for high-repetition-rate and continuous operation. In these cases, the appropriate figures of merit indicate that the DBS materials are noticeably better than the birefringent crystals. ZnGeP₂ and AgGaSe₂ are both type I phasematched. The “e” and “o” signify the polarizations of the waves in the birefringent crystals, extraordinary and ordinary respectively. Figures of merit for different applications are given in table II.

Calculated singly resonant OPO (SRO) thresholds provide another comparison of materials. Table 4. presents the values that were used for the different material parameters. The threshold for the DBS GaAs SRO was calculated assuming both first and third order QPM; first order QPM was assumed for the threshold calculation for ZnSe. Threshold pump pulse energy was calculated using the analysis of Brosnan and Byer²⁸, with threshold defined as the level of pumping at which the signal output reaches 100 μJ . Both birefringent crystals, ZnGeP₂ and AgGaSe₂, were type I phasematched. Pump absorption was included. An output coupler of $R = 80\%$ was assumed.

In Fig. 9 we compare the calculated threshold pulse energies for degenerate SRO's of DBS GaAs, DBS ZnSe, ZnGeP₂ and AgGaSe₂. The SRO's are pumped by 10-ns 2- μm pulses. Thresholds of all crystals are within an order of magnitude. As the length of the crystal increases, the absorption becomes more important.

TABLE 3. SECOND HARMONIC GENERATION OF 10.6- μm RADIATION IN A 1-cm CRYSTAL

crystal	transmission range (μm)	d (pm/V)	α (cm^{-1})	K_{therm} (W/mK)	$\text{dn/dT} \cdot 10^6$ (K^{-1})	$\text{d}\Delta\text{n/dT} \cdot 10^6$ (K^{-1})	n_{ω}	$n_{2\omega}$	ρ (degrees)
GaAs	0.9-12	83	0.01	52	149	1.6	3.271	3.296	-
DBS									
ZnSe	0.5-14	50	0.0005	18	64	0.7	2.403	2.429	-
DBS									
ZnGeP ₂	0.7-12	70	1	35	150 (n_o) 170 (n_e)	20	3.125 (e)	3.125 (o)	-
AgGaSe ₂	0.7-12	33	0.01	1	70 (n_o) 40 (n_e)	30	2.592 (o)	2.592 (e)	0.68

crystal	θ_{pm} (degrees)	d_{eff} (pm/V)	$\Delta\lambda$ (μm)	$\Delta\Theta_{\text{ext}}$ (degrees)	ΔT (K)	FOM _{cw}	FOM _p	FOM _{od}	FOM _{hp}
GaAs	-	61	0.48	64	270	6500	2.5	160	3100
DBS									
ZnSe	-	37	0.2	38	<500	2400000	2.4	2400	33000
DBS									
ZnGeP ₂	90 @ T=92°C	70	0.06	41	16	1	3.8	$n_e=1.0$	2.4
AgGaSe ₂	55	27	0.12	2	16	2600	1	$n_o=6.9$	1

d - nonlinear coefficient of the crystal
 α - absorption coefficient of the crystal
 K_{therm} - thermal conductivity
 d_{eff} - effective nonlinear coefficient
 $\text{d}\Delta\text{n/dT} - \Delta\text{n} = n_2 - n_1$
 n_{ω} - index of refraction at 10.6 μm
 $n_{2\omega}$ - index of refraction at 5.3 μm

ρ - walkoff angle
 Θ_{pm} - phasematching angle
 dn/dT - derivative of index w/r to temperature
 $\Delta\lambda$ - the acceptance bandwidth
 $\Delta\Theta_{\text{ext}}$ - the external angular acceptance
 ΔT - the acceptance bandwidth for temperatures

NB. for Figures of Merit. They have all been normalized to the lowest value.

FOM_{cw} -Figure of Merit for a continuous laser:
FOM_p -Figure of Merit for a pulsed laser:
FOM_{od} -Figure of Merit for optical distortion:
FOM_{hp} -Figure of Merit for high average power:

$$\begin{aligned} \text{FOM}_{\text{cw}} &= d_{\text{eff}}^2/n^3\alpha^2 \\ \text{FOM}_{\text{p}} &= d_{\text{eff}}^2/n^3 \\ \text{FOM}_{\text{od}} &= K_{\text{therm}}/\alpha\text{dn/dT} \\ \text{FOM}_{\text{hp}} &= d_{\text{eff}}^2K_{\text{therm}}/\alpha n^2\text{d}\Delta\text{n/dT} \end{aligned}$$

TABLE III. VALUES USED TO CALCULATE THE SRO THRESHOLDS

crystal	α_p @ 2 μm (cm^{-1})	α_{si} @ 4 μm (cm^{-1})	n_p (2 μm)	n_{si} (4 μm)	θ_{pm} (deg.)	L_c (μm)	Λ (μm)	d_{eff} (pm/V)	ρ (deg.)
GaAs	0.01	0.01	3.341	3.304	-	26.9	53.7	61	-
DBS ZnSe	0.002	0.0015	2.435	2.421	-	71.8	143.5	20	-
DBS ZnGeP ₂	0.3	0.02	3.153	3.153	56	-	-	64	0.65
AgGaSe ₂	0.10	0.01	2.619 (o) (e)	2.619 (e) (o)	52	-	-	26	0.64

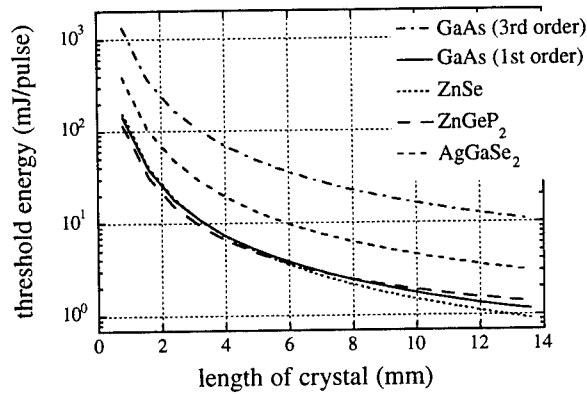


Fig. 9. Calculated threshold pulse energies for 2- μm pumped 10-ns pulsed SRO's at degeneracy. The cavity length was 15 mm.

It is important to recall that damage thresholds and average power capabilities will be better for the DBS materials. This will allow superior OPO performance at higher levels of pumping. It should be noted that we have used bulk absorption for GaAs and ZnSe. This assumes no scattering or absorption due to the diffusion bonding. We have not yet shown this experimentally.

5. Nonlinear Devices: Experimental

The preliminary nonlinear testing of DBS GaAs used SHG of CO₂ laser radiation. The dispersion equation¹⁸ predicts a coherence length, L_c , of 106 μm for doubling 10.6 μm radiation in GaAs. Available {110} wafers were 435 μm thick ($\pm 5 \mu\text{m}$). While this thickness was adequate for initial testing of the diffusion bonding technique, it was not an odd multiple of L_c and, therefore, not optimal for SHG at 10.6 μm .

Five samples, with 2, 3, 5, 7, and 9 layers were characterized, and compared to a single wafer. Linearly polarized 10.6- μm radiation from a grating-tuned, 200-ns, Q-switched CO_2 laser was incident normal to the DBS GaAs sample with approximately 2-MW/cm² peak intensity. Second harmonic output power at 5.3 μm was measured with an InSb detector. The SHG power for normal incidence of the fundamental was measured as a function of the angle Φ between the polarization of the fundamental and the $\langle 110 \rangle$ direction. The expected $(1 + 3\sin^2\Phi)\cos^2\Phi$ dependence of the output power was observed. For the remainder of the measurements, the fundamental polarization was fixed along a $\langle 111 \rangle$ direction.

Figure 10 shows the relative SHG power for diffusion bonded stacks of 2, 3, 5, 7, and 9 layers, as well as for a single plate. The measured thickness of the wafers was used to calculate the total phase mismatch as a function of the number of layers, N , and the output power was compared to the expected $\sin^2(\Delta kNL/4)$ dependence, where $\Delta k \equiv k_1 - 2k_2 - K_m$. Here k_2 and k_1 are the wave vectors for the fundamental and second harmonic frequencies, respectively. The period of the modulation, Λ , is twice the wafer thickness. The agreement with the expected dependence on the number of layers demonstrates that the DBS GaAs acts as a monolithic structure with a modulated nonlinear coefficient, providing a phase coherent interaction.

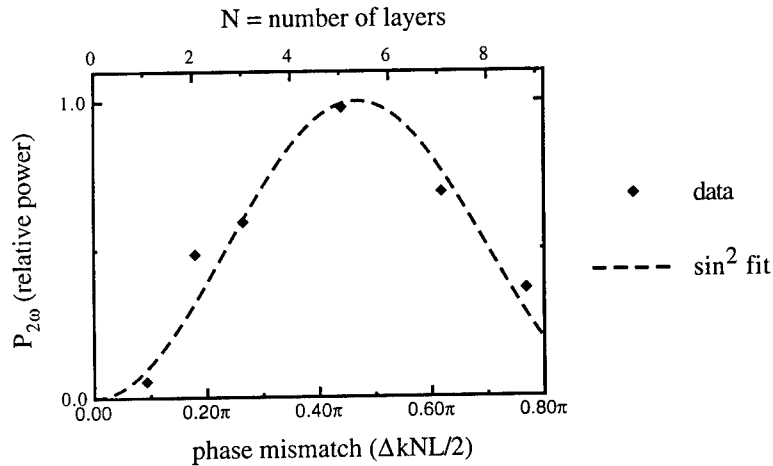


Fig. 10. Second harmonic output power, $P_{2\omega}$, of a CO_2 laser as a function of phase mismatch and number of layers in the diffusion-bonded quasi-phasematched GaAs structures.

C. Damage Testing

Many potential applications of these DBS GaAs structures involve frequency conversion at high average and high peak power; therefore, some preliminary damage measurements were performed. We exposed the both bulk and DBS GaAs to increasingly higher intensities for up to 5 minutes, then examined the samples for damage under a microscope. A 26-W cw CO₂ laser beam was focused to a $\sim 60\text{-}\mu\text{m}$ $1/e^2$ radius spot, producing an intensity of $\sim 500\text{ KW/cm}^2$. No damage was detected, whereas AgGaSe₂ damages at $5\text{-}60\text{ KW/cm}^2$ with a cw CO₂ laser¹. A 1-kHz pulsed CO₂ laser with 60 to 500- μs pulse width was focused to a peak intensity of 30 MW/cm^2 . No damage was seen in the bulk GaAs. We were unable to get accurate data from the DBS GaAs at this power level because the degraded outer surfaces developed damage characteristic of thermal runaway; however, the samples were not noticeably damaged with a single pulse, compared to AgGaSe₂, which damages at $\sim 10\text{ MW/cm}^2$ for 80-180 ns pulsed operation². Previous work in this lab has found GaAs wafers to damage at $\sim 300\text{ MW/cm}^2$ for a 200ns pulse.

D. Conclusions

Our preliminary investigations of DBS GaAs have included numerical modeling, process development, SHG, and measurement of optical damage characteristics. These results indicate the feasibility of fabricating DBS structures. We chose GaAs for the initial work, and were able to fuse wafers to form monolithic crystals with bond strengths comparable to the bulk material. We measured SHG nonlinear conversion which followed theoretical predictions. We also determined that GaAs damage thresholds are at least 500 kW/cm^2 for cw and 30 MW/cm^2 for peak power at $10.6\text{ }\mu\text{m}$. These measurements and analyses have demonstrated the potential of high average power OPO operation with this material.

Before we can fabricate a working DBS OPO, we need to expand our work in certain areas: a) we are studying the bonding process. Understanding the mechanisms will permit us to improve the bonds and transfer the technology to other materials. b) we have developed a polishing facility which will allow us to polish the wafers to precise thicknesses and to repolish the ends after processing. c) we need to increase the numbers of layers to improve conversion efficiency. d) we also need to measure damage thresholds at different wavelengths, and with higher power lasers before we can accurately predict maximum operating intensities for the DBS devices.

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II. RESEARCH RESULTS

There exists a need for high average power coherent sources in the mid infrared (IR). Nonlinear frequency conversion can provide these sources using existing lasers; however, currently available IR nonlinear crystals can not handle the high powers. The goal of this work is to develop synthetic microstructured semiconductors for high power IR frequency conversion.

The availability of appropriate crystals has been one of the most limiting factors in nonlinear optics. III-V and II-VI semiconductors are well-characterized and well-developed nonlinear crystals with good thermal properties; however, they can not phasematch interactions by conventional birefringent techniques. Diffusion-bonded-stacked (DBS) structures can use non-birefringent materials to quasi-phasematch interactions, and therefore, have the potential to provide nonlinear optical crystals tailored to specifications. These DBS nonlinear materials can be used to make many devices: harmonic generators, difference and sum-frequency generators, and optical parametric oscillators.

For all of our initial research, we have chosen to use GaAs because of its availability, low cost, and uniform crystal quality. Other semiconductors have different characteristics which might make them more attractive than GaAs for certain applications. Once we understand the process in GaAs we will be able to explore other material systems, opening up the possibility of custom made synthetic crystals for a range of nonlinear interactions.

A. Brief Summary of Work To Date

1992 Progress:

- basic calculations for second harmonic generation with DBS GaAs.
- built special furnace with Roger Route for diffusion bonding.
- developed a cleaning technique for preparing the wafers for bonding.
- successfully diffusion bonded 2 through 7-wafer stacks of random orientation.
- demonstrated enhancement of second-harmonic generation by quasi-phasematching in a two-wafer GaAs stack.

We completed a number of preliminary runs with the special furnace that Roger Route constructed for diffusion bonding. In these preliminary investigations, we were using standard GaAs wafers polished on both sides. The wafers were cleaned and stacked, one on top of the other, alternating the crystal orientation, and heated in a furnace to diffusion bond the stack. We used a 1-kg tungsten weight to provide the necessary pressure. We successfully diffusion bonded 2 through 7-wafer stacks of randomly oriented GaAs. On November 22 we tested the first of our devices. We doubled the output of a 10.6- μm CO₂ laser to 5.3- μm , and measured close to theoretical improvement with the two-layer stack over a single wafer.

1992 Summary:

The successful demonstration of diffusion bonding and quasi-phasematched second-harmonic generation were important milestones for this program. A number of attempts were necessary to determine cleaning and contacting procedures in preparation for bonding, and improvement was still necessary for this critical step of the process. Larger stacks were necessary to study the linear and nonlinear properties. It became evident that it would be necessary to move the GaAs polishing in-house since it was not possible to find small companies that could polish accurately thin wafers of GaAs in small quantities.

1993 Progress:

- improved on the wafer preparation process, both cleaning and contacting.
- built a vacuum chuck for holding the wafers vertically as they were being contacted.
- successfully diffusion bonded wafers of different orientations.
- successfully diffusion bonded 2 through 9-wafer stacks of (110) GaAs appropriately oriented for nonlinear enhancement.
- demonstrated enhancement of second-harmonic generation of 10.6- μm radiation by 5th order quasi-phasematching in the 2 through 9-layer stacks of GaAs.
- presented a post-deadline paper at CLEO'93.
- measured damage thresholds at 10.6- μm .
- submitted a paper to Electronic Letters.
- theory for other semiconductors for SHG's, DFG's, OPO's and SFG's.
- established a polishing facility for GaAs in Ginzton Lab. With in-house polishing support, we can obtain wafers within 2- μm of our specifications.

- built another bonding holder designed to apply increased and more uniform pressure than the original holder.
- studied the role of oxides in diffusion bonding.

We completed the bonding of 2 through 9 layer stacks of alternatively oriented GaAs. We successfully diffusion bonded different wafer orientations, {110} to {100}, as well as identical wafer orientations, {110} to {110} and {100} to {100}. The samples cleaved along the crystal planes leaving the bonded surfaces intact. The exterior surfaces of the stack, which were in contact with graphite spacers, were noticeably degraded. We did not repolish these surfaces, as the resulting uncertainty in the layer thicknesses would have hampered analysis of the SHG results. We measured second-harmonic generation by quasi-phases matching in 2, 3, 5, 7, and 9 layer stacks, and demonstrated that the devices provided phase coherent nonlinear interactions even though the available wafers were not the correct thickness for optimal conversion.

Some preliminary optical damage measurements were performed. A 26-W cw CO₂ laser beam was focused to a $\sim 60\text{-}\mu\text{m}$ $1/e^2$ radius spot, producing an intensity of ~ 500 KW/cm². No damage was detected in either the bulk GaAs or the DBS GaAs, whereas AgGaSe₂ damages at 5-60 KW/cm² with a cw CO₂ laser. A 1KHz pulsed CO₂ laser, with a pulse width from 60 to 500- μs , was focused to an intensity of 30-MW/cm². While no damage was seen in bulk GaAs optics, we were unable to get accurate data from the DBS GaAs at this power level because the degraded outer surfaces developed damage characteristic of thermal runaway; however, the samples were not noticeably damaged with a single pulse, compared to AgGaSe₂, which damages at ~ 10 MW/cm² for 80-180 ns pulsed operation.

We purchased a Logitech lapping and polishing machine to permit us to polish the wafers to the correct thicknesses, and to repolish the devices after processing. It was installed in Ginzton Laboratory, and was monitored and approved for GaAs polishing. Leslie Gordon attended a Logitech training course for the lapping and polishing of GaAs, and we worked with representatives of Rodel and Fujimi, manufacturers of polishing pads and solutions, to develop high quality polishing processes for use with the Logitech machine.

We completed computer modeling of various potential DBS devices to help us to predict the best configurations and materials. In March we finished the patent application

for the DBS structure, and filed it with the patent office. A summary of this work was presented at a post deadline paper at CLEO, May 1993.

In August, we submitted a paper to Electronics Letters. We developed a new design for the diffusion bonding holder, and build a prototype in 304 stainless steel. This holder permitted us to apply increased pressure in a repeatable manner, and would be used in our original oven. The holder also incorporated the vacuum chuck for wafer assembly. The holder concept worked well; however, the stainless steel exhibited too much creep at the bonding temperatures (800-850°C).

We began to vary the surface preparation of the wafers in an effort to improve the bonding quality and decrease the loss at the interfaces. This included etching the surfaces with various acids and bases to remove the oxide layers before contacting. While it is generally accepted that the cleaner and the flatter the wafers are the better the contacting and bonding will be, it is not understood what role the oxides play in the bonding process.

1993 Summary:

As this year progressed, it became evident that it was necessary to understand the relative importance of the different parameters: flatness, temperature, pressure and time before upscaling the production to much larger structures i.e. 50-100 layers. We had measured losses of 2% per interface. This was sufficiently low to demonstrate that quasi-phasesmatching could work in DBS materials; however, that loss was not acceptable for devices with a large number of layers. We needed to study the loss mechanisms and find a way to reduce the losses.

1994 Progress:

- demonstrated diffusion bonding in polycrystalline ZnSe.
- presented a paper at SPIE OE/LASE '94.
- developed different wafer cleaning techniques using various acids and bases to etch away the oxide layers before contacting.
- studied the loss/interface as a function of different wafer preparations.
- reduced the loss from 2% to 0.5% per interface.
- extensively studied the voids at the interfaces in an effort to reduce the loss per interface.

- studied void filling or solid phase epitaxial regrowth at the interface as a function of temperature and pressure.
- studied the effects of bonding under high pressure in a hydraulic press.
- built a new furnace where the pressure is controlled from the outside by a spring.
- built an interferometer at 1.064- μm to provide "no contact" thickness measurement of the GaAs wafers, and to examine the bonding quality.
- designed and built a miniature quartz holder/oven for fast cycling times and very uniform temperature and pressure. The pressure is applied from the outside by an air piston, and may be controlled very accurately.

For many applications, ZnSe has superior thermal properties to GaAs; however, single crystal growth of ZnSe is not as developed as that for GaAs. We demonstrated the first reported diffusion bonding in polycrystalline ZnSe, confirming that it is not unreasonable to assume that we will be able to transfer much of what we learn about diffusion bonding in GaAs to the other semiconductors.

We presented a paper at SPIE OE/LASE '94 in January on our latest experimental results and calculations on various different DBS devices, focusing on OPO's.

In an effort to understand the source of the loss in the diffusion bonded samples, we experimented with sample cleaning/preparation and bonding pressure. It is generally accepted that removing the oxide layer before contacting the wafers will improve the bond; however, the most optimal process to accomplish this is not obvious. After cleaning and degreasing the wafers, we have tried various etches of HCl, HF, and NH_4OH . H_2SO_4 and HNO_3 were not used because they were reported to form oxides on GaAs surfaces. Concentrations of very dilute to maximum strength were used. We found the best results with the higher concentrations, especially in HF, but the results were not transparent. Larger stacks will be necessary to measure the difference between the etches. We also compared contacting after etching under liquid: methanol, propanol, and DI water with contacting in air. While it is not evident yet whether one of these liquids is significantly better than the others, we achieved much more uniform, and lower loss bonds when we contacted the surfaces under a liquid compared to air. This technique virtually eliminated particulate matter at the interface.

We evaluate the quality of the bond in four ways: a) IR transmission with low power microscope or IR viewer, b) IR microscope at high power focused on the interface, c)

cleaving the sample and observing the cross-section of the interface under high power magnification, and d) accurate measurements of the transmission at specific wavelengths. Evaluation a) will identify bonds with particulates $>0.2 \mu\text{m}$. None of our samples have this problem anymore since we have been contacting under liquid. With evaluations b) and c), we identify small $\leq 0.2 \mu\text{m}$ "bubbles" at the interface. We believe that these "bubbles" are either voids or gallium pools. We have been varying the temperature and the pressure of the bonding process and been observing the resulting structures at the interface. There appear to be different growth mechanisms at different temperatures studied. Above 900°C , the growth is not as uniform, and strange star-shaped formations develop at the interface. We are studying the growth mechanisms to develop a process that would produce uniform interfaces with no "bubbles". We have currently reduced the loss to $\approx 0.5\%$ per interface for $2\text{-}\mu\text{m}$ radiation. The loss was measured in a $2\text{-}\mu\text{m}$ interferometer at Lightwave Electronics.

One of the major limitations with our original holder is the amount and uniformity of the pressure applied during the bonding process. We have constructed two new ovens to avoid these limitations and also provide the ability to vary the pressure during the bonding process. The first oven applies pressure by means of a spring. The spring can be exchanged for one of higher or lower spring constant. The second oven applies pressure by means of a air piston. Pressure can be regulated from $0\text{-}100 \text{ kg/cm}^2$, and can be adjusted at any time during the bonding. We are just beginning bonding with these new oven/holders.

We have recently completed a $1.064\text{-}\mu\text{m}$ interferometer to provide no contact evaluation of the uniformity of the polished wafers, and uniformity of the bonded interface.

1994 Summary:

Cleanliness and uniformity of pressure seem to be the two most important parameters in the bonding process. Contacting under a clean liquid eliminates large particulates $\geq 0.2 \mu\text{m}$. We have reduced the loss to a uniform $\approx 0.5\%$ per interface for $2\text{-}\mu\text{m}$ radiation. For a more accurate measurement of the loss we will now have to make larger stacks. To further reduce the loss we will need to eliminate the "bubbles" that we have observed at the interface; however, with the current loss we are in a position to fabricate larger stacks, and perform device demonstrations in the new year.

B. Outline of Future Work

The work on diffusion bonding may be divided into two categories: material development and device fabrication and testing. When we first began this project, we fabricated devices to demonstrate the feasibility of the DBS structure. Our initial stacks were very lossy; however, we demonstrated that the diffusion bonding process preserved the homogeneity of the linear optical properties, and created a monolithic structure with a modulated nonlinear coefficient. Before we could manufacture large stacks, we needed to understand the critical parameters in the bonding process. During the past year we have been focused on the material development to reduce the loss at the interface. Now we are in a position to fabricate new devices, but we need to maintain a parallel materials development.

Stage 1:

- demonstrate high quality bonding in a 20-layer stack.
- demonstrate SHG in a 20-layer stack of GaAs optimized for third order quasi-phasematching of 10.6- μm radiation.
- measure the efficiency, angle acceptance, temperature acceptance and wavelength acceptance of the 20-layer stack.
- measure the damage threshold of diffusion bonded GaAs at 1, 2, and 10 μm .
- improve surface preparation and wafer contacting.
- study GaAs regrowth and mass transport along the interface as a function of temperature, pressure, and atmosphere.
- reduce the loss per bond even further, $>0.1\%$ loss per interface.
- demonstrate high quality bonding in a 50-layer stack.
- demonstrate SHG in the 50-layer stack of GaAs optimized for third order quasi-phasematching of 10.6- μm radiation.
- fabricate a 50-layer stack in GaAs optimized for SHG first order quasi-phasematching of 10.6 μm -radiation .
- demonstrate high efficiency SHG conversion in the 50-layer stack.
- demonstrate 2- μm pumped OPO in the 50-layer stack.
- measure the efficiency, angle acceptance, temperature acceptance and wavelength acceptance of the 50-layer stacks for both SHG and OPO.
- demonstrate diffusion bonding in single-crystal ZnTe and ZnSe.

- study ZnTe and ZnSe regrowth and mass transport along the interface as a function of temperature, pressure, and atmosphere.

Stage 2:

- continue the material study in GaAs: surface preparation and contacting
regrowth and mass transport
- reduce the loss per bond even further, >0.05% loss per interface.
- fabricate a low loss 100-layer stack.
- demonstrate high efficiency SHG conversion in the 100-layer stack.
- demonstrate 2- μm pumped OPO in the 100-layer stack.
- measure the efficiency, angle acceptance, temperature acceptance and wavelength acceptance of the 100-layer stack for both SHG and OPO.
- measure the damage threshold at 1, 2, and 10 μm of lower-loss diffusion bonded GaAs.
- demonstrate a wedge structure in a 20-layer stack.

III. SCIENTIFIC PERSONNEL SUPPORTED BY THIS CONTRACT

Principal Investigators

Professor Robert L. Byer	Applied Physics Department
Robert S. Feigelson	Material Science and Engineering

Other Scientific Faculty and Staff

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Students and Degrees Awarded

Leslie A. Gordon, Applied Physics
Dong Zheng, Applied Physics
Loren Eyers, Applied Physics
Sermon Wu, Materials Science and Engineering

IV. LIST OF ALL PUBLICATIONS SUPPORTED BY ARO CONTRACT DAAL03-92-G-0400

1. L. Gordon, G. L. Woods, R. C. Eckardt, R. K. Route, R. S. Feigelson, M. M. Fejer and R. L. Byer, "Diffusion bonded stacked GaAs for quasi-phase-matched second harmonic generation of a carbon dioxide laser," *Electron. Lett.* **29**, pp. 1942-1944 (1993).
2. L. Gordon, G. L. Woods, R. Route, R. S. Feigelson, R. C. Eckardt, M. M. Fejer and R. L. Byer, "Diffusion bonded GaAs for SHG," postdeadline paper CPD19, Conference on Lasers and Electro-Optics (CLEO), Baltimore, MD, May 1993; in *CLEO, 1993, Postdeadline Papers* (Optical Society of America, Washington, DC, 1993), pp. 38-39.
3. L. Gordon, R. C. Eckardt and R. L. Byer, "Investigations of diffusion-bonded quasi-phase-matched GaAs for infrared parametric oscillation," paper 2145-44, *Optoelectronic for Information and Microwave Systems (SPIE OE/LASE '94)*, Los Angeles, Jan. 22-29, 1994; *Proc. SPIE* **2145**, 2145, pp. 316-326 (1994).