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DEVELOPMENT OF AN OPTIMIZED SECOND HARMONIC GENERATOR CELL

Michael Bass

Raytheon Company
Waltham, Massachusetts

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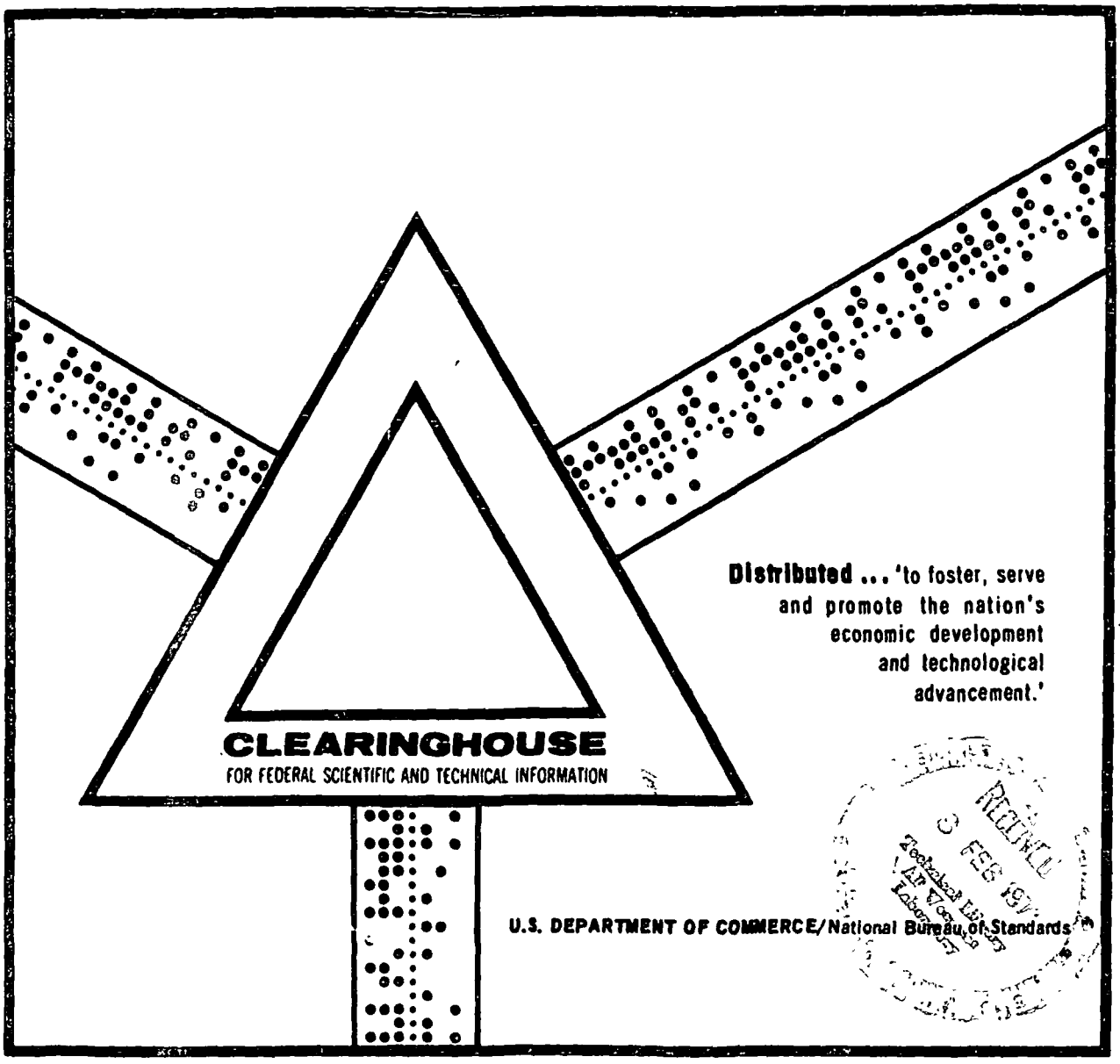
April 1970



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S-1220

DEVELOPMENT OF AN OPTIMIZED SECOND HARMONIC GENERATOR CELL

by Michael Bass

Final Scientific Report
June 12, 1969 to March 12, 1970

April 1970

Prepared under Contract No. NAS 12-2155

RAYTHEON COMPANY, RESEARCH DIVISION
Waltham, Massachusetts

Prepared for

Electronics Research Center
NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

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LIST OF ILLUSTRATIONS

DEVELOPMENT OF AN OPTIMIZED SECOND HARMONIC GENERATOR CELL

by Michael Bass

SUMMARY

An optimized frequency doubler to convert 1.06μ light into light having 0.53μ wavelength has been developed. The nonlinear crystal which is the heart of this device is "hot" $LiNbO_3$. This material was selected because of its large nonlinear coefficient, the fact that it can be noncritically phase matched for frequency doubling along its crystalline a axis and that it was available in large samples of high optical quality.

The crystal was mounted in an oven which could maintain its temperature stably from 159 to 200°C. At the proper temperature for a-axis phase-matching, a 1.06μ beam in the TEM_{00} mode, having a power density of 44 MW/cm was converted to light to 0.53μ with 9.2 percent efficiency. The beam at 0.53μ retained the TEM_{00} mode configuration and divergence.

CHOICE OF THE NONLINEAR MATERIAL

Availability

In order to design and construct a frequency doubler having the desired properties, a nonlinear material had to be chosen from those which were readily available at the beginning of this work. Those materials were KDP, $LiNbO_3$, lithium rich $LiNbO_3$ ("hot" $LiNbO_3$) and $Ba_2NaNb_5O_{15}$. $LiIO_3$ had been reported in the literature but could not be delivered in useful sizes within 9 months.[1] The properties of the available materials which finally led to the selection of "hot" $LiNbO_3$ for the frequency doubler are discussed in detail in the following sections.

Nonlinearities

If the selection of the nonlinear material depended solely on the magnitude of the coefficients for optical second harmonic generation, then $Ba_2NaNb_5O_{15}$ would have been chosen. This material has a useful nonlinear coefficient 3 times that of either $LiNbO_3$ or "hot" $LiNbO_3$ and ~30 times that of KDP. However, high quality pieces of $Ba_2NaNb_5O_{15}$ large enough to be useful as a frequency doubler could not be obtained during the course of this work.

[1] M. Gsänger, Technische Hochschule Munchen, Physik Department

On the other hand, since it is possible to trade off nonlinear coefficient for crystal length and maintain the efficiency of the doubler a "hot" $LiNbO_3$ crystal ~3 times longer than one of $Ba_2NaNb_5O_{15}$ could be used as an equally effective a frequency doubler.

Optical Quality

For the present work, an optimized frequency doubler was defined as one which produced as much second harmonic light as possible without distorting the beam profile or increasing its divergence. Thus one principal factor in selecting the nonlinear crystal for this device was the optical quality of available large samples. An acceptable crystal was one which was free of scatter centers, striations, and strain, and poled so that a single ferroelectric domain was present.

Available crystals of KDP or both forms of $LiNbO_3$ satisfied these conditions but no $Ba_2NaNb_5O_{15}$ crystal obtained during the present work was acceptable. These crystals were either imperfectly poled or possessed striations. Therefore, since KDP was ruled out on the basis of small nonlinear coefficients and $Ba_2NaNb_5O_{15}$ on the basis of an available optical quality the choice was restricted to one or the other forms of $LiNbO_3$.

Damage to the Nonlinear Crystal

Volume damage

Most nonlinear materials suffer surface damage due to laser irradiation before volume damage is noted. However, $LiNbO_3$ is susceptible to a form of optical "damage" induced by low-intensity visible light. This damage is associated with imperfections, possibly oxygen vacancies, existing in the crystal. The visible photons have enough energy to break the binding energy which traps electrons at the vacant lattice site. The electrons are then free to move away, leaving behind a local electric field distribution in the crystal. Because of the high electro-optic effect of this material, this local electric field causes refractive index gradients, which diffuse the light beam and seriously upset the index-match conditions necessary for efficient second-harmonic generation.

Annealing the crystal at a temperature somewhat higher than room temperature increases the mobility of electrons in the crystal enough to permit neutralization of the charged vacancy; thereby removing the refractive index gradients. The "damage" can thus be reversed. Operating the second-harmonic generator at somewhat elevated temperatures, one may hope to reach a point where damage is annealed out as fast as it is introduced. Since the ability to index-match along a crystal axis depends upon holding the nonlinear crystal at a specific temperature, we are not free to increase the temperature in order to adquately anneal the "damage." Fortunately, the lithium-rich form of $LiNbO_3$ "hot" $LiNbO_3$, has an index-matching temperature as much as 50° to 140°C higher than that characteristic of

ordinary stoichiometric $LiNbO_3$. This material then can be used as a frequency doubler which will not distort the beam because of induced index of refraction gradients.

Surface damage

In order to determine the maximum incident optical flux which the doubler could withstand, we examined surface damage in $LiNbO_3$ and for comparison, in $Ba_2NaNb_5O_{15}$. Particular emphasis was placed on finding the relationship between the number of shots, N, of 1.06μ radiation required to damage each material and the value of the incident optical power density, P. For a multimode laser this relationship, in the range of P values between no damage at all (i.e. where $N = \infty$) and damage in one shot is found to be closely given by a power law,

$$N \propto P^{-m} , \qquad (1)$$

where $m \simeq 1$ for both materials.

Figure 1 shows the relationship measured for $LiNbO_3$[1] between the number of laser pulses, N, at a certain power density, P, required to damage the surface and that power density. Figure 2 shows the same relationship for $Ba_2NaNb_5O_{15}$. These results were obtained by focusing the multimode 1.06μ Q-switched Nd:YAG laser beam with a 10 cm focal-length lens to a spot of ~0.45 mm diam. on the polished surface of the ~0.5 cm thick crystal. This focus was chosen so that any "hot spots" in the laser beam cross-section might be superimposed and impinge on only one area of the crystal. By varying the pulse energy and duration, the power density could then be selected. A pulse repetition rate of 1 pps was used.

The crystal was said to have just damaged when an incandescent spark was seen by the experimenter. After the spark was detected, a visual inspection of the once clean crystal surface revealed a tiny pit where the laser beam had been focused. The surface of each crystal was studied twice by each of two independent observers: In each case, the surface was carefully finished to $\lambda/4$ in the Raytheon optical shop for one experiment and for the other experiment it was roughly polished in the laboratory. Similar results were obtained in each case by each observer.

Figures 1 and 2 show that, for power densities greater than P_1, a single laser pulse is sufficient to damage the material. Of course, the higher power densities only a fractional pulse would be necessary since only P_1 is required to cause damage. Therefore, if the plots were extended to fractional shots, the relation between N and P would be

$$N \propto P^{-1} \qquad \text{for } P \geq P_1$$
$$N \leq 1 \qquad (2)$$

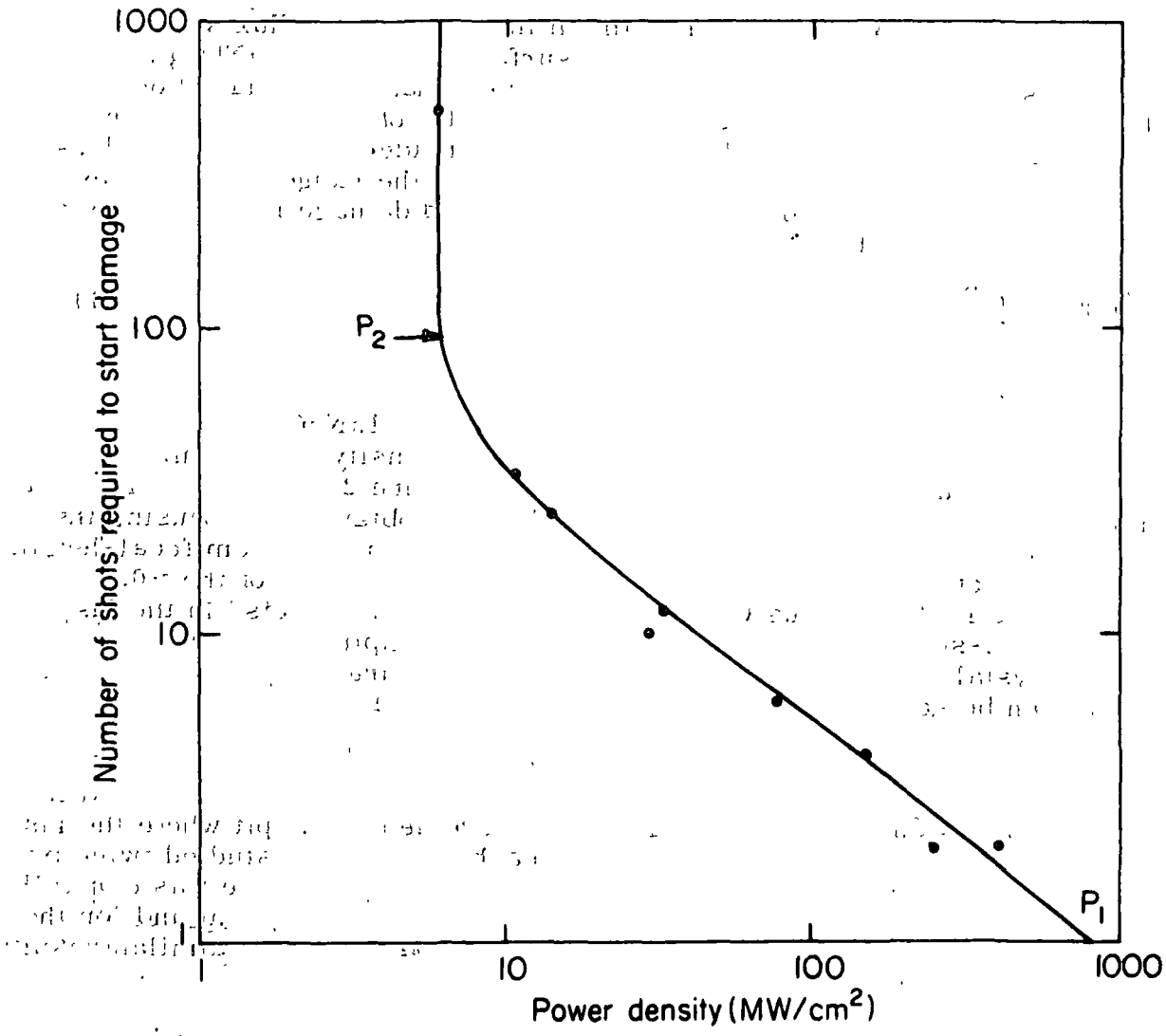


Fig. 1 The Experimentally Determined Relationship between the Number of Laser Shots Required to Start Surface Damage and the Multimode Laser Power Density on the Surface of $LiNbO_3$

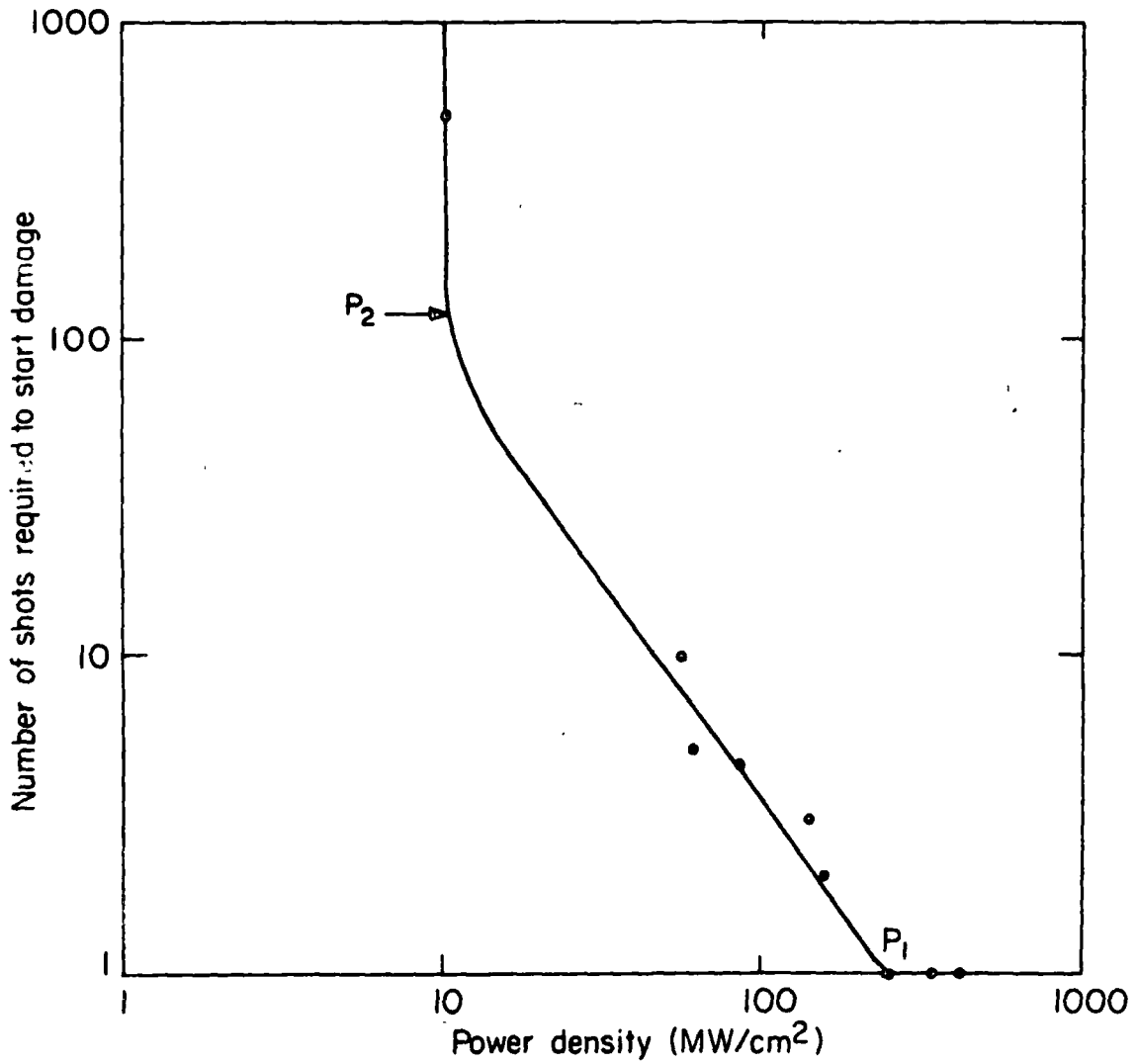


Fig. 2 The Experimentally Determined Relationship between the Number of Laser Shots Required to Start Surface Damage and the Multimode Laser Power Density on the Surface of $Ba_2NaNb_5O_{15}$

When the power density is less than P_2 a great many pulses would be required to detect any surface damage; thus, for $P \leq P_2$, $N \to \infty$.

For most of the range $P_2 < P < P_1$, Figs. 1 and 2 show that

$$N \propto P^{-m}$$

where

$m \simeq 0.8$ for $LiNbO_3$

and

$m \simeq 1.3$ for $BaNaNb_5O_{15}$,

is a good approximation. At both the high and low limits of this range of power densities, some deviation from a simple power law is observed. In the high power limit the relation must become that given in Eq. (1) and for low powers it must asymptotically become $N \to \infty$ for $P \lesssim P_2$.

We see from the preceding results that the occurrence of surface damage in $LiNbO_3$ and $Ba_2NaNb_5O_{15}$ has a well defined threshold; in terms of incident power density the threshold for surface damage is P_2. The damage process at power densities above P_2 appears to involve a localized change on the surface. It might be that microscopic damage spots occur which then grow with successive pulses until they become visible. However, the change in the surface, whatever it may be, accumulates until the last pulse of light strikes the area and causes a breakdown (we see a spark) and damage becomes visible to the dye.

The results given above indicate that for long-lived systems (i.e., systems withstanding many laser pulses), using $LiNbO_3$ or $Ba_2NaNb_5O_{15}$, the multimode power density on the crystal surface should, to be safe, not exceed $\sim$5 MW/cm^2. However, $LiNbO_3$ and "hot" $LiNbO_3$ were able to withstand 1.06μ pulses with power density $\sim$ 50 MW/cm^2 when the beam is restricted to the TEM_{00} mode. This apparent contradiction is resolved when one considers the filamentary nature of the multimode laser beam. When a filament is focused onto the crystal surface the power density in its image can be $\gtrsim$ 100 MW/cm^2 even when the averaged power density (the values used in Figs. 1 and 2) is only 10 MW/cm^2.

The damage pattern shown in Fig. 3 gives evidence that the focused filaments may be responsible for the damage. This figure shows laser-irradiation-induced damage on the surface of a $Ba_2NaNb_5O_{15}$ crystal as seen through a microscope with 80X magnification. This damage was caused in one shot by focusing the multimode output of a Q-switched Nd:YAG laser onto the surface. The relevant experimental parameters were: focal length of the lens 10 cm, beam diameter on the surface $\sim$ 0.4 mm, laser energy $\sim$ 7mJ and pulse duration $\sim$ 30 nsec. The crystal had not been

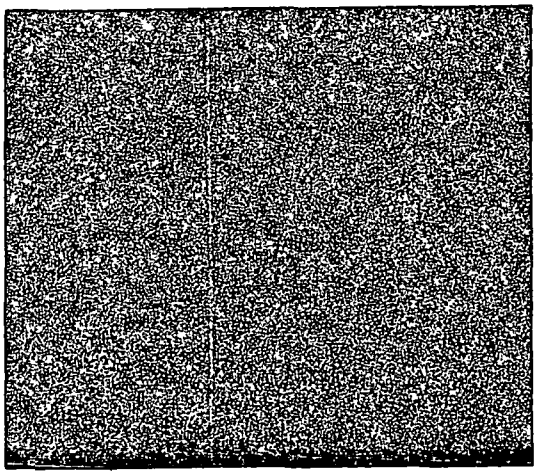


FIGURE 3

detwinned or poled and had been polished in our laboratory. The area irradiated is the darker region in the photo, but damage pits appear in only a few well defined places where various filaments may have been focused.

These studies have placed the following 1.06μ power density limits on "hot" $LiNbO_3$ for continuous use as a frequency doubler:

(1) TEM_{00} mode $\leq$ 50 MW/cm^2

(2) Multimode (filamentary) $\leq$ 5 MW/cm^2 (average)

Optical Absorption of "hot" $LiNbO_3$

We measured the visible and near infrared transmission spectra of "hot $LiNbO_3$," which are shown in Fig. 4. The sample was 1.5 cm long and not anti-reflection coated. Note the nonzero absorption at 0.53μ. In addition, we found indirect evidence of a very small but nevertheless very important absorption at 1.06μ in this material. We placed a crystal of "hot" $LiNbO_3$ in an oven at a temperature of 163°C or ~5°C below its a axis index-matching temperature. Then we doubled a long pulse (~100 mJ in ~ 100 μsec) 10 pps Nd:YAG laser and monitored the second harmonic power generated as a function of time. The average 1.06μ power was ~ 1W. The curve in Fig. 5, a plot of second harmonic power versus time, shows the "sin At/At" dependence which would be expected if the crystal was heated up along the beam from its initial temperature to one above the index matching temperature. The total temperature change was about 7-8°C and the maximum average harmonic power was less than 5mW near the peak of the curve. When the crystal cooled, the same experiment was performed when the laser was Q-switched. The 1.06μ average power was less than 0.1W and the maximum average second harmonic power was ~ 20 mW. No appreciable heating was observed in this case. We therefore conclude that absorption at 1.06μ does occur in "hot $LiNbO_3$" and may limit its usefulness in high average power applications.

These results mean that the initial temperature of the oven must be chosen so that the sample heats just to its index matching temperature at the laser's 1.06μ power level. If that power level varies the harmonic output can fall drastically. It may be necessary to monitor the crystal temperature to guide a fast response oven in order to maintain efficient doubling for long periods.

OVEN CHARACTERISTICS

The oven in which the "hot" lithium niobate crystal was mounted was

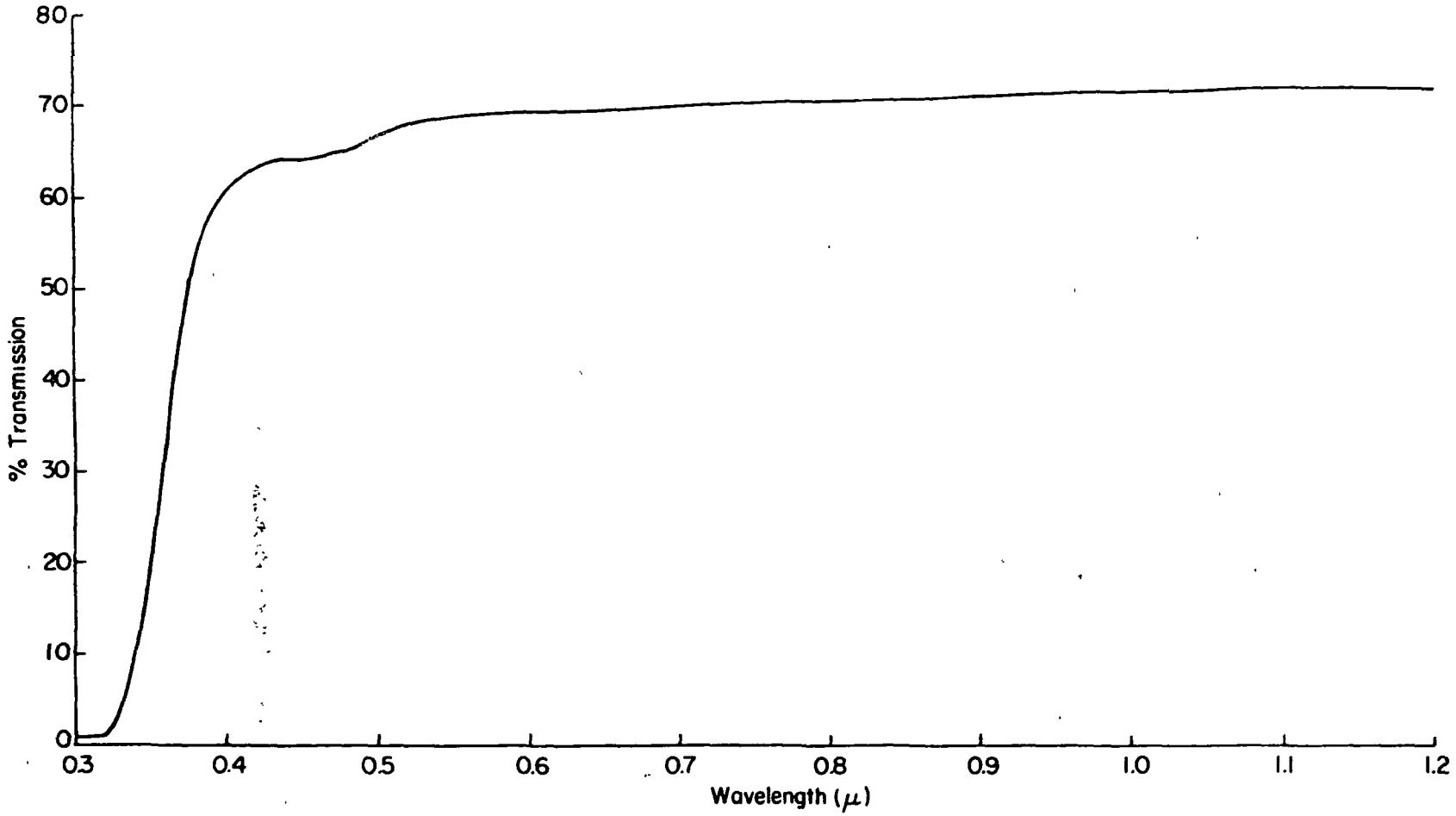


Fig. 4 Transmission vs Wavelength of "Hot" $LiNbO_3$. The sample faces were not coated and so the high index of refraction (~ 2) caused the ~ 30 percent Fresnel reflection observed. The sample was 1.5 cm thick

manufactured by Oven Industries. * The oven itself and the control electronics come in two separate units. The performance specifications are:

Cavity temperature:	adjustable, 150°C to 200°C
Setting tolerance:	± 0.5°C
Cavity temperature stability:	± 0.01°C at room ambient, with mounted windows**
Internal cavity size:	0.875" and 0.875" and 1.000"
Controller:	AC proportional
Input voltage:	115V AC, 60 Hz
Max. Power output:	175 watts to resistive load

The dial setting of the control unit for optimum second harmonic generation depends somewhat on the average power level at the fundamental wavelength, due to slight local heating in the lithium niobate crystal. (See II-E) The correct setting varies between 238 and 248 for the particular crystal supplied.

One face of the lithium niobate crystal is mounted against one inside surface of the oven cavity. A 1 cm long section of Viton "O"-ring material of 0.070" diameter is used to apply pressure to the crystal on the opposite face in order to establish thermal contact and to hold the crystal in place. The viton strip is recessed slightly in the wall of the oven cavity.

Two teflon tubes are mounted between the oven cavity and the outside container of the oven to prevent the thermally insulating material from contaminating the crystal surfaces.

---

* Detail drawings and other data relevant to the oven are given in Appendix A.

** With no windows to seal the oven we found the temperature to be stable to within ± 0.04°C.

THE OVEN CARRIAGE

The oven is mounted in a frame which permits micrometer-controlled adjustment of the orientation of the crystal in the two mutually orthogonal directions normal to the direction of the incident beam. These movements are designed so that the intersection of the axes of rotation occurs at the center of the doubler crystal. In this manner the movement of the crystal during alignment is minimized. Both movements are spring loaded to prevent backlash and can be locked in position when adjusted.

The "hot" $LiNbO_3$ crystal is oriented so that efficient frequency doubling is possible for horizontally polarized fundamental light. This means that the second harmonic which is generated will be vertically polarized.

The base plate of the mount is fitted with 2, 1/4 - 20 tapped holes to permit its use with optical bench posts.

PERFORMANCE TESTS AND RESULTS

The frequency doubler was tested using a pulsed Q-switched Nd:YAG laser as the source of light at 1.06μ. This laser, when restricted to oscillate in the TEM_{00} mode only, produced a pulse having 7 nsec duration and an energy content of 1 mJ at a prf of 1, 5 or 10 pps. The incident power density was selected by adjusting the beam diameter with extracavity optics.

The pulse energy at 1.06μ was measured using a TRG model 100 Ballistic thermopile having a sensitivity of 197.6 μV/joule. Since the energy at 0.53μ could be so small as to be unmeasurable with this device, an ITT S1 vacuum photodiode (Model F4000) was used to measure both the 1.06μ and 0.53μ energy. We calibrated this photodiode at 1.06μ by comparing its output with the energy measured using the thermopile. The calibration of the photodiode at 0.53μ could then be found by multiplying this result by 1.61, the measured photodiode sensitivity at 0.53μ relative to that at 1.06μ.

The efficiency of optical second harmonic generation is maximum when it occurs in a noncritically phase-matched direction parallel to a crystalline axis. Since the crystal used herein was an a axis crystal of "hot" $LiNbO_3$, we aligned this direction parallel to the Nd:YAG laser beam and raised the crystal temperature until index matching of 0.53 and 1.06μ light occurred along the a axis. This temperature chosen was the one where maximum SHG was detected. When the crystal is below this critical temperature there are many index-matching directions, which are all in a cone centered about the a axis. Since some 1.06μ light is scattered into this cone, we can observe its presence by a bright ring of light at 0.53μ centered about the forward direction. Thus one can determine the proper index-matching temperature by slowing increasing the crystal's temperature until this ring of green

light just disappears. Under these conditions we found:

% conversion of 1.06 into 0.53*	1.06μ power density
0.5	0.6 MW/cm^2
0.7	1.4
9.2	44.0

These results show the dependence of doubling efficiency on the fundamental power density.

Due to the high quality of the doubler crystal, the spatial distribution of the harmonic light remained the same as that of the fundamental.

POTENTIAL SUBSTITUTE NONLINEAR MATERIALS

The nonlinear material used in the present doubler was chosen because of its high nonlinear coefficients and the fact that the optical quality of available samples satisfied the requirements that the doubler not distort the beam or increase its divergence. If $Ba_2NaNb_5O_{15}$ crystals of similar optical quality become available in the future one should consider their use in a frequency doubler. In addition, $LiIO_3$, which has nonlinear coefficients greater than $LiNbO_3$ should be considered for future doublers because its optical and nonlinear properties are insensitive to temperature from 20 to 256°C.** This material, though not having symmetry which permits noncritical phase matching along an axis, can, however, be used to obtain efficient temperature independent doubling and so could be used without an oven.

OPERATING INSTRUCTIONS

1. Align frequency doubler so that the 1.06μ laser beam passes through the center of the crystal and is normal to the polished crystal faces

   a. The 1.06μ beam must be polarized horizontally

   b. The maximum aperture of the frequency doubler is 7 mm

   c. The doubler crystal is coated so that one side transmits 1.06μ and the other side transmits 0.53μ. Thus the 1.06μ beam must always enter through the former side. An arrow on top of the oven indicates the proper entrance and exit directions.

---

*These values are obtained by averaging the results obtained from several different pulses.

**F. R. Nash, J. G. Bergman, G. D. Boyd and E. H. Turner, J. Appl. Phys. 40, 5281 (1969).

d. If the windows are used they must be put on the correct end of the oven. That is the 1.06μ transmitting window on the entrance end and the 0.53μ transmitting window on the exist end. Due to a volatile component within the oven a "scum" may form on the windows. Therefore, unless the added thermal stability is essential, the windows should not be used.

2. Set the oven temperature slightly below the index-matching temperature (a dial setting of ~230 on the oven control unit). Turn on the 1.06μ laser (in the TEM_{00} mode the power density must not exceed ~ 50 MW/cm^2; if the multimode it must be less than 5 MW/cm^2) and on a piece of paper in front of the doubler one will see a bright green spot in the forward direction surrounded by a green ring of light. As the temperature is slowly increased this ring will collapse into the center. The index-matching temperature is reached when the radius of the ring of green light is just zero. If one monitors the harmonic intensity during this tune up, it will be maximum when the index-matching temperature is just reached.

APPENDIX A

DATA AND SPECIFICATIONS FOR OVEN SUPPLIED BY OVEN INDUSTRIES

The material contained in this appendix was supplied to the Research Division by the oven manufacturer. It appears in the same form as received from Oven Industries. The original material was supplied to NASA-ERC.

[O]I CONTROLLER TEST REPORT

SERIAL NUMBER [illegible] MODEL NUMBER 7079 DATE 12-22-69

I. PHYSICAL INSPECTION

- ☑ COMPONENT DAMAGE
- ☑ SOLDERING
- ☑ BOARD MOUNTING AND POT ALIGNMENT
- ☑ THERMISTOR ASSEMBLY
- ☑ MECHANICAL FIT

ACCEPTED I. [illegible signature]
INSPECTOR

II. OPERATIONAL EVALUATION – SINGLE UNIT

BANDWIDTH ΔR = 160 Ω

INPUT	OUTPUT
117 VAC	114 VDC

LOAD 30 w

	TEMP	RESISTANCE
FULL-ON	Chart [illegible]	[illegible]
FULL-OFF		
NOMINAL SETTING	[illegible]	

AGE: TIME ON ______ TIME OFF ______ TOTAL 16 hr

III. OPERATIONAL EVALUATION – MULTIPLE UNITS

INPUT ______ LOAD ______

SERIAL NUMBER	OUTPUT	TEMP SETTING		TOTAL AGE HRS
Note: Chart Tag to Customer				

ADDITIONAL UNITS LISTED ON BACK

IV. FINAL INSPECTION

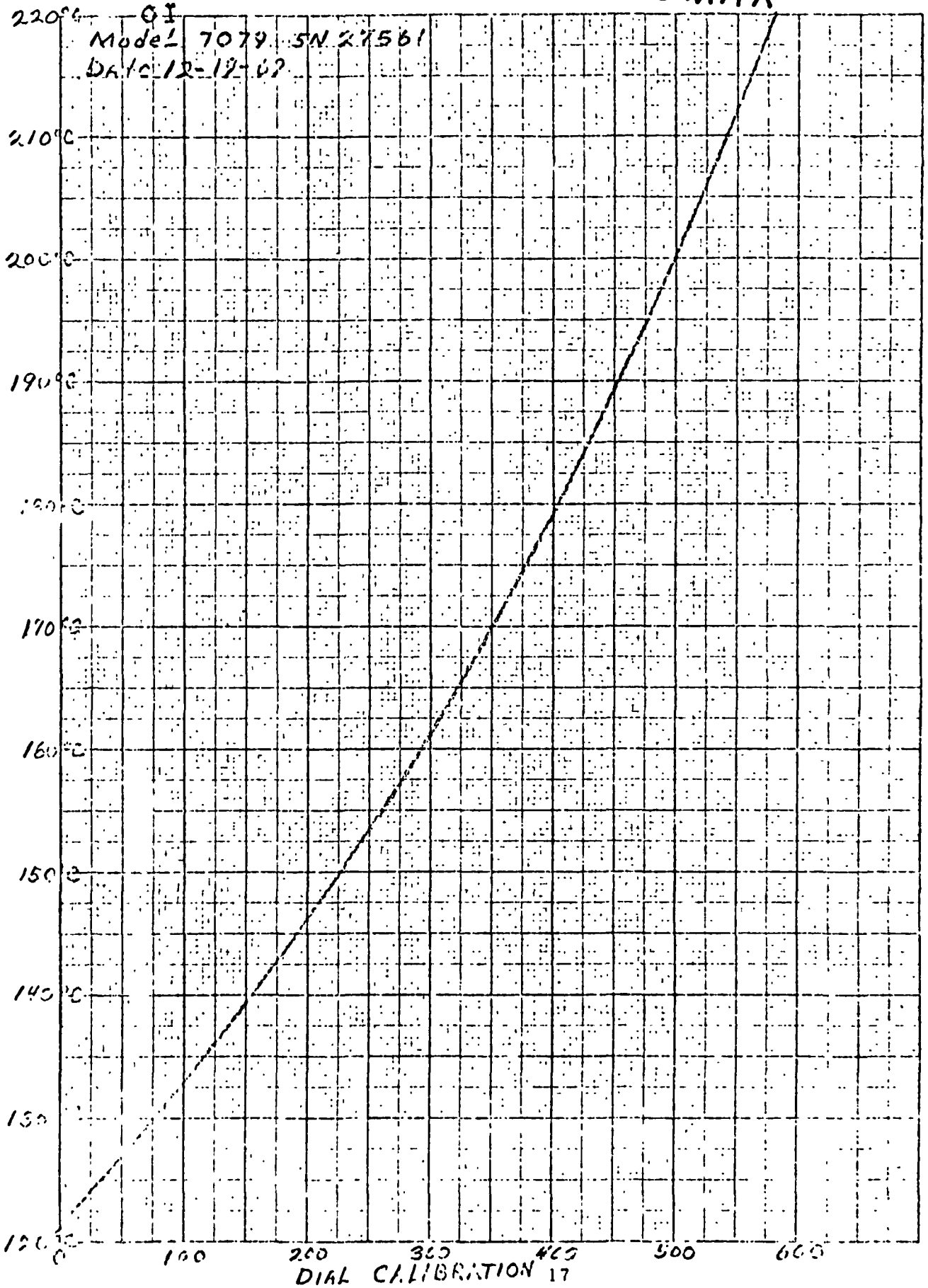
- ☑ UNITS CLEAN
- ☑ THREADS AND SCREWS CLEAR
- ☑ LABEL/MARKING
- ☑ ON UNITS

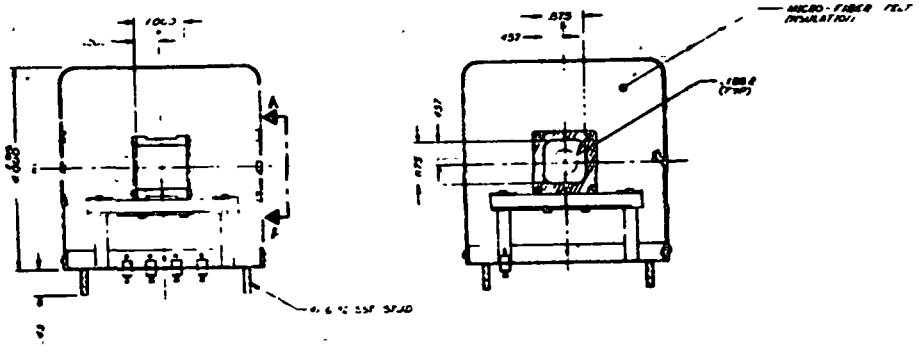
TOTAL NO. OF UNITS COVERED BY THIS REPORT ______

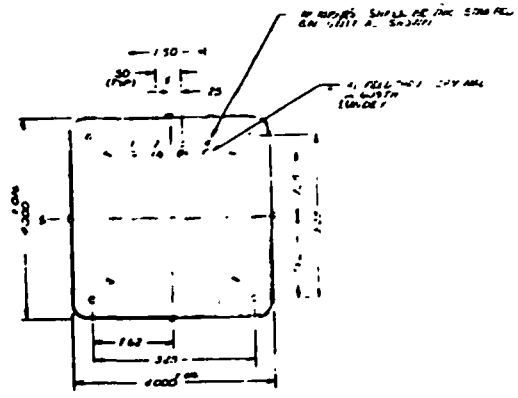
II - III [illegible signature] ACCEPTED TEST

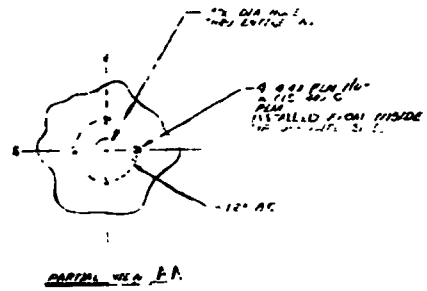
IV. [illegible signature] INSPECTOR

220°C MAX


OI
Model 7079 SN 27561
Date 12-12-07
220°C
210°C
200°C
190°C
170°C
160°C
150°C
140°C
130
0
100
200
300
400
500
600
DIAL CALIBRATION







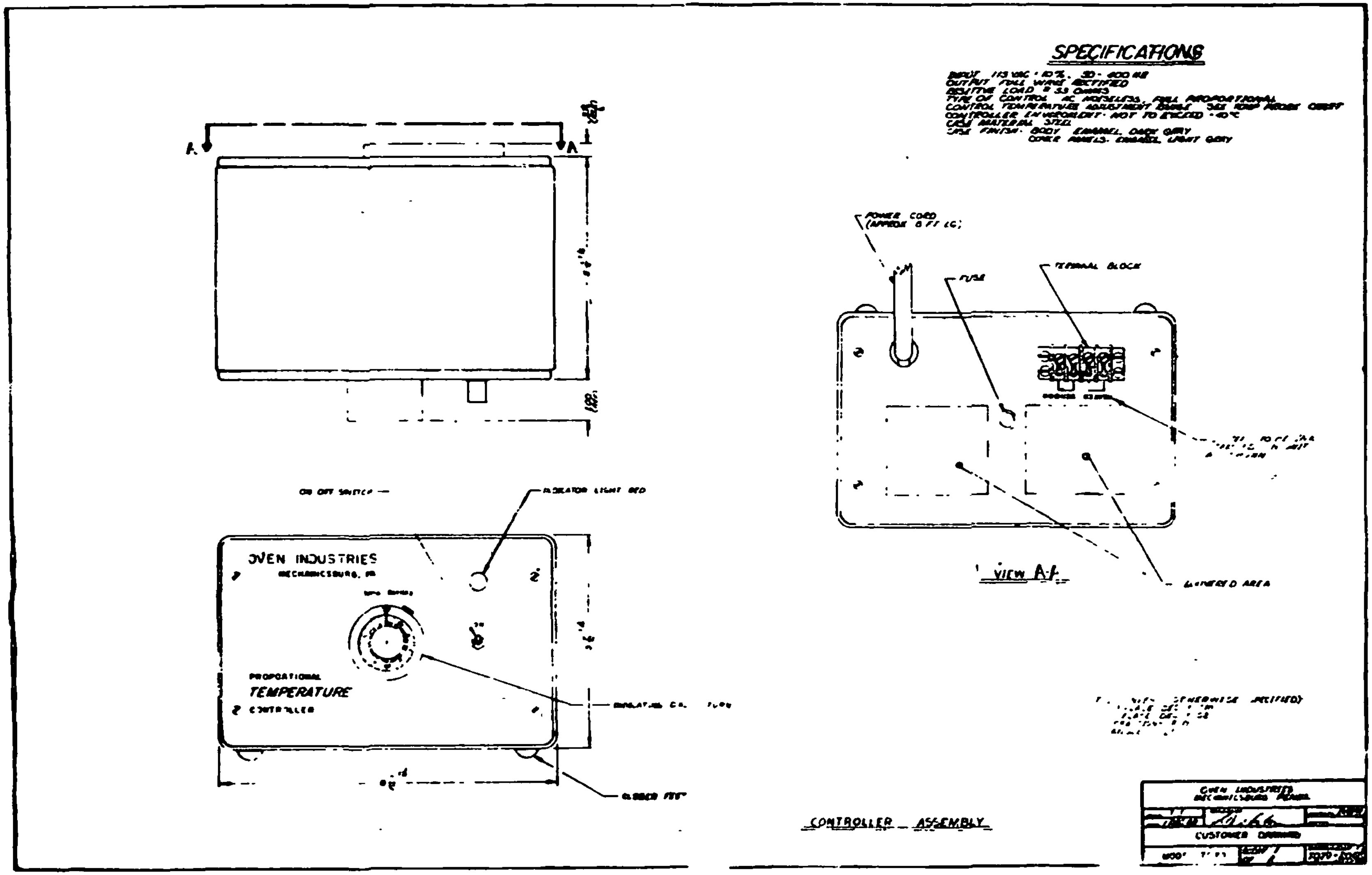
SPECIFICATIONS

CAVITY TEMPERATURE ADJUSTABLE, 150°C - 300°C
SETTING TOLERANCE ± 5°C
TEMPERATURE STABILITY ± 1°C @ ROOM AMBIENT
EXTERNAL CASE MATERIAL ALUMINUM

PIN CONNECTIONS

PIN 1 — SENSOR
2 — SENSOR
3 — HEATER
4 — HEATER

OVEN ASSEMBLY


SPECIFICATIONS
POWER CORD
FUSE
TERMINAL BLOCK
VIEW A-A
OVEN INDUSTRIES
PROPORTIONAL
TEMPERATURE
CONTROLLER
ON OFF SWITCH
INDICATOR LIGHT RED
RUBBER FEET
CONTROLLER ASSEMBLY
CUSTOMER DRAWING