Contamination and Radiation Effects on Nonlinear Crystals for Space Laser Systems

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Abstract:
Space Lasers are vital tools for NASA's space missions and military applications. Although, lasers are highly reliable on the ground, several past space laser missions proved to be short-lived and unreliable. In this communication, we are shedding more light on the contamination and radiation issues, which are the most common causes for optical damages and laser failures in space. At first, we will present results based on the study of liquids and subsequently correlate these results to the particulates of the laser system environment. We present a model explaining how the laser beam traps contaminants against the optical surfaces and cause optical damages and the role of gravity in the process. We also report the results of the second harmonic generation efficiency for nonlinear optical crystals irradiated with high-energy beams of protons. In addition, we are proposing to employ the technique of adsorption to minimize the presence of adsorbing molecules present in the laser compartment.

Introduction

Laser induced damage of optical components is a common problem for all high power lasers and becomes even more prominent with space flight lasers. NASA uses space lasers to advance our understanding of space research for space-based altimetry, high spatial resolution profiling of aerosol properties, vegetation canopy height and density, wind speed, cloud parameters, gain and loss of ice packs, improving our prediction capability of the climates, and natural hazards, etc.

The space environment puts extreme requirements on building space flight lasers. These lasers are expected to meet performance requirement and maintain their alignment irregardless of the intense vibrations and the accelerated gravity (15x) that they are subjected to during the lift off. The lasers must also withstand extreme thermal cycles along with pressure changes. Our knowledge of the ultra high vacuum space environment on optical materials and thin film coatings is still very limited. Changes that might take place in the composition of the surfaces and the out-gassing of the adhesives with time under ultra high vacuum in space environment can cause dramatic changes and our knowledge of these effects is still very confined. The cost of launching a piece of hardware into space is very expensive and increases with instrument weight. This puts a great deal of emphasis on miniaturization. In addition, the use of power to run the system in space and energy dissipation is limited by the capture of the solar energy by the solar panels. This puts another constraint on the use of power and the efficiency of the system. A typical space flight laser operates in the millijoules to one joule per pulse regime with pulse durations in the nanosecond range, and beam diameters of a few millimeters. These specifications mean that these lasers produce power densities in the regime of 100 to 500 million watts per square centimeter. Science specifications require that spaceflight lasers emit millions to billions of shots. This lifetime is much longer than that of any typical system on the ground.

A ground-base laser would also be serviced periodically whereas this is not possible with a spaceflight laser.

These many constraints along with the harsh space environment make building space flight lasers a challenge. Unfortunately, several of NASA's space Light Detection And Ranging (LIDAR) laser systems have encountered some sort of difficulty and some missions have been cancelled due to cost growth. The LIDAR In-space Technology Experiment (LITE) on the space shuttle was highly successful, but had laser pulse energy degradation in both of its lasers; a worrisome result for long lifetime missions. Other missions have dealt with the challenges of having few replacement parts due to lack of vendors. This caused higher than desired risk. The difficulty and cost of space qualified lasers and other LIDAR components have often been greater than estimated. Mars Orbiter Laser Altimeter (MOLA), which is considered to be a great success, was lunched in November 1996 and stopped sending data in June 30, 2001 after 216 power (on/off) cycles, firing 671 million times, and 640 million measurements of the Martian surface and atmosphere. The Geoscience Laser Altimeter System (GLAS) was designed to measure ice-sheet topography and associated temporal changes, as well as cloud and atmospheric properties. It contains three identical lasers to prolong the lifetime of the mission. It was launched on 13 January 2003. To our surprise, Laser 1 operated for only 36 days on-orbit before it suffered an excessive power degradation and catastrophic failure due to an unexpected failure mechanism in a pump diode array. Laser 2 lasted for only a month and at the timing of writing this paper, laser 3 has been running at 60% of its energy for the last 31-days.

Based on this brief history, and after three decades of research, the technical community concluded that the optical damage in laser systems is mostly a contamination driven problem. Trace levels of contaminants can lead to laser induced optical damage, which occur at a relatively low power density. Therefore, contamination induced optical damage is considered to be a significant factor in reducing laser reliability and lifetime. In this paper, we will try to shed light on the different mechanisms contributing to the contamination issues and propose a solution to reduce their effects. We will also irradiated a few nonlinear crystals with a high energy beam of protons and examined the effect of irradiation on the second harmonic generation efficiency of these crystals.

Contamination

Laser damage due to contamination is a complex issue due to the complexity of the contamination causes. It was found that the optical damages in laser systems caused by contamination are due in most part to nonlinear effects, which are more prominent with high-energy lasers. Although ground systems are also sensitive to contamination, they are typically less sensitive than spaceflight systems. In other words, the absence of gravity in space may play a significant role in enhancing the effect of optical damage caused by particulate contaminants. On the ground, for example, contaminant particles tend to scatter light, however, they will eventually settle on the floor of the laser compartment due to gravity. On the other hand, the behavior of these contaminants in space is totally different. The contaminants in space will have a longer residence in the suspended state and move aimlessly in the laser compartment. These contaminants eventually get attracted to the laser beam by the electrostrictive pressure and driven to the optical surfaces, where they absorb light, burn out and turn black, causing laser diffraction based optical intensification, which potentially result in laser focusing, filamentation, and end up producing an optical damage. This induced laser damage varies significantly with wavelength, pulse duration, beam profile, mode structure (spatial and temporal), and peak power.

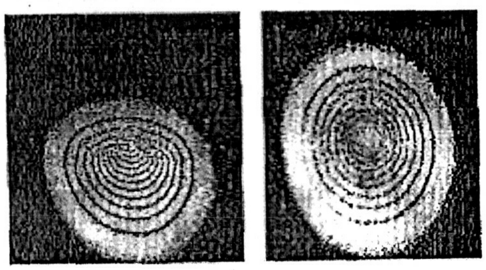
Common damaging contaminants are often metallic particulates or molecular contaminants. Metallic particulates are well known to interact efficiently with radiation. This can result in vaporization and subsequent plasma formation as they absorb energy. If these particulates are contaminating on an optical surface, their absorbed energy might produce vaporization or ionization of the thin film coating on the optical surfaces and eventually system failure. Carbon and inorganic oxide particulates can also induce heating centers on the surface of the optics that result in permanent thermal stresses and fractures. Examples of detrimental molecular contaminants are silicones and aromatic hydrocarbons such as toluene. Although both of these materials are essentially transparent from the near infrared through the visible wavelength range, they are known to damage laser optics in one-micron lasers. This leads to the conclusion that laser-contamination interaction does not follow linear optical behavior.

How do contaminants cause optical damages?

A few relevant experiments were performed on liquids and the results were subsequently correlated to the particulates in a laser system compartment. The experiments demonstrated clearly the concept of optical trapping of molecules by the laser beam. The high-intensity radiation field of the laser at the focus[1] tends to attract molecules towards the region of highest intensity (beam axis), push and trap them against any optical surface (such as a lens, a mirror or a crystal) and form a cluster. The condensation of the cluster takes the opposite shape of the light intensity distribution. For example, if the beam is a Gaussian, the cluster takes the form of a converging lens that tends to focus the beam sharply on the optics causing optical damage.

It was also confirmed through these experiments on liquids that the phenomenon of cluster formation takes place only in absorptive molecules, even if a little absorption is present. Consequently, heat will be generated that will cause thermal stress and refractive index modulation in the cluster on the optical surface that will eventually contribute to optical damage through thermal stress and fracture.

In the first experiment, a cw Ar^+ laser Gaussian beam at 514.5 nm and power level of a few tens of milliwatts, was sent horizontally and focused within a 10-mm path-length of a rectangular cuvette containing an absorptive organic laser dye solution or a Chinese green tea solution in ethanol. A set of perfect circular fringes (Figure1b) appears on a far-field screen, which quickly turns to a set of half-circles (figure 1a). These fringes are due to spatial self-phase modulation[2]. If the cuvette is moved along the beam path such that the beam is focused at the entrance side of the cuvette, the beam fans downward as shown in Figure 2 over an angular range of (0-90^{o}) downward from its axis. It was also found that the fanning was quasi-periodic every nearly 10 to 20 seconds depending on the beam energy, the organic material, and the solution concentration.



(A) (B)

Figure 1. (A) the interference fringes due to spatial self-phase modulation after one second. (B) the initial pattern.

By sending the beam vertically through the cuvette, it was found that the gravity has a role in the shape of the fringes and the fanning. The circular fringes remain circular and the fanning is symmetric with respect to the beam axis. To verify that the Ar^+ laser beam is inducing refractive-index variations, a He-Ne laser beam at low power (~3mw) is sent as a probe, co-propagating with the Ar^+ laser beam at 514.5 nm. Both beams are focused

Figure 2. The fanning of the Laser beam.

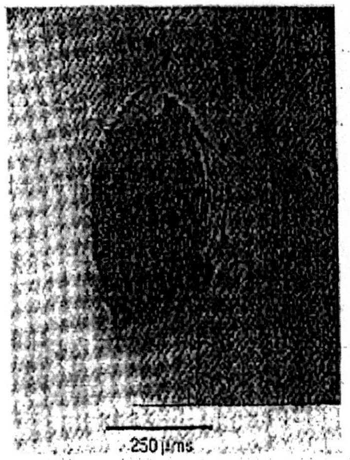


Figure 3. The burned cluster.

within the sample. When the argon laser beam is blocked, the He-Ne laser beam passes through the solution unaffected, when the argon laser is allowed through, an induced half circular spatial self-phase modulation pattern of the He-Ne laser beam can be seen on the screen through a 632.8 nm narrow band filter. Translating the cuvette, such that the focus is on the entrance side of the cuvette, results in the fanning of the Ar^+ laser and the He-Ne laser beams. If the argon laser power is increased much beyond the threshold for fanning, a random and permanent light scattering with undefined direction from the focal point becomes permanent. When the cell is removed away from the laser and examined under the microscope, a black spot ~250 μm in size is apparent on the cell wall at the beam focus (Figure 3).

How can these results be explained?

Ashkin[3] presumed that the radiation pressure from tunable lasers selectively accelerates, traps or decelerates the atoms or molecules of gases at specific resonances. For instance, an Ar^+ laser beam at a power of 1W, and a wavelength (λ)= 514.5 nm, focused on a lossless dielectric sphere of radius $r=\lambda$ and of density = 1 gm/cc induces an acceleration = 1.2×10^8cm/sec^2, which is ~ 10^5 times the acceleration of gravity. When the light induces a temperature gradient, the gravitational force will induce a convection force and the particles move. These thermal forces are usually orders of magnitude larger than radiation pressure. In the absence of gravity in space, the heating energy is localized and the optical trapping force is greater than the convection force.

The trapping[3] of the particles by the field distribution of a Gaussian beam can be understood as shown in Figure 4. If the particle is of a refractive index that is greater than its environment and considering a pair of beams A and C at equal spacing from the sphere axis B, the stronger beam A undergoes Fresnel reflection and refraction at the input and output surfaces as shown. The resulting radiation pressure forces due to reflection are F_{ri} and F_{ro}, which are along the bisectors of the input and the reflected beams at the input and the output surfaces of the sphere. The pressure forces due to the refracting beams are F_{di} and F_{do} at the input and output surfaces, which are along the bisectors of the input and output refracted beams. The resultant of these forces gives acceleration along the +z-axis and a transverse force that will make the spheres to be drawn towards the axis of the beam. If the spheres are of lower refractive index less than the environment, like the case of air bubbles in water, they will be pushed away from the beam axis.

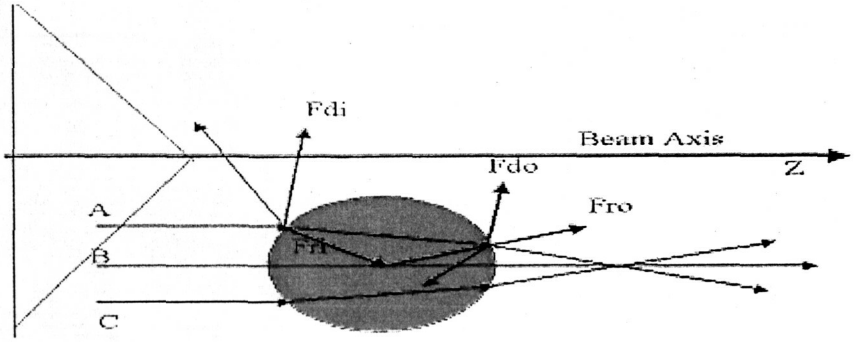


Figure 4. A dielectric sphere of the beam axis. The forces due to (A) move the sphere toward the beam axis and along +z.

Furthermore, It is known[4] that a dielectric sphere in an electric field E behaves as a dipole with polarization P given by

$$P=[(n_a^2 - n_b^2)/(n_a^2 + 2n_b^2)]\, r^3 E = \alpha E$$

Where n_b is the refractive index of the surrounding medium, n_a is the refractive index of the sphere, r is the radius of the sphere, and α is the polarizability of the sphere suspended in the medium. In an optical field gradient, we have a force on the sphere, F_{grad}, given by

$$F_{grad} = (P.\nabla)E = (1/2)\alpha\nabla(E_o^2)$$

Where E_o is the rms electric field in the light wave and the factor (1/2) is a time average over a cycle of light oscillation. For the case of $n_a > n_b$ the force moves the spheres towards the high field region (condensation) while for $n_a < n_b$ the spheres will be subjected to a force that moves them away from the high field region (rarefaction). In the condensed region, the average refractive index will be increased (positive nonlinear Kerr effect) and it will be decreased in the rarefaction region.

In addition to the light radiation pressure, the electromagnetic field can exert a force on the particles through the mean square electric field. Usually, in inhomogeneous high frequency field, this force is many times larger than the force of the light radiation pressure[5]. The transverse inhomogeneity of a strong electromagnetic beam can exert a strong effect on the electrons and atoms of a medium. Thus, if the frequency exceeds the natural frequency of the electron oscillations, then the electrons or atoms will be forced out of the beam field. At sub-resonance frequencies, the particles will be pulled in. The force will be especially large at resonance. It is noted that this effect can also create either a rarefaction or compression in the beam at the focus of the radiation. The average force acting on the electrons of an atom from the external field is

$$F_{av} = (e^2/m)\{\Sigma\, (\omega_{ok}^2 - \omega^2)/[(\omega_{ok}^2 - \omega^2)^2 + \gamma_k^2]\}\, \nabla E_{av}^2$$

Where, ω_{ok} and γ_k are the resonance frequency and the damping of the k^{th} electron, respectively. If the radiation frequency ω exceeds the natural coupling frequency of the external electrons in the atom, then an average force will act on these electrons and force them out of the field, while if ω is less than ω_o, the F_{av} will attract the molecules toward the region of high intensity field. From Boltzmann formula, we estimate the gradient of the density of the neutral molecules on the axis of the beam as

$$\rho(0)/\rho(\infty) \cong \exp[-U/kT] = \exp\{(\nu e^2 E_o^2)/[m(\omega_o^2 - \omega^2) kT]\}$$

Where ν is the number of electrons in the outer shells of the atoms. It is easy to see that the medium becomes rarefied at the center when $\omega > \omega_o$ and condensed when $\omega < \omega_o$.

Moreover, the intense light at the focus exerts a strong attractive force on the particulates and the molecules and as a result, an increase in density of the matrial ($\Delta\rho$) and in its dielectric constant ($\Delta\varepsilon$) will take place. The electostrictive pressure induced by the field is[6]

$$P = -\rho(\partial\varepsilon/\partial\rho)(E^2/8\pi)$$

The negative sign indicates that the pressure is reduced in regions of high field strength. Consequently, the molecules tend to move towards the axis of the beam or the focus.

How the solution results can be correlated with contaminants in vacuum?

From the discussion above, it is obvious that the condensation at the region of high field intensity is independent of the medium and not restricted to liquids. Consequently, atoms, molecules, and particulates of gases[6, 7] and solids are also subjected to be accelerated, decelerated, and attracted towards the fields of high intensity, where, they will be forced along the beam and be trapped against the surfaces of the optical components. Molecular condensation at the surface will lead to micro-destruction, emission of hypersonic and shock waves, and to a local heating of the medium, which will eventually lead to optical damages.

Why the absence of gravity in space plays a major factor in enhancing the contamination issue of damaging optics?

As can be seen from the above experiments on solution, the circular pattern of spatial phase modulation quickly turns to a pattern of half circles due to gravity. This can be explained as follows: the non-uniformity of the field distribution induces a non-uniform thermal heating and consequently a density distribution, which is stable in the lower half below the beam axis and unstable in the upper half due to convection caused by the gravitational force on the ground. This means that gravity on the ground has a role in the formation of the spatial self-phase modulation and the formation of the interference pattern.

In addition, it was noticed that fanning is quasi periodic at certain level of the laser power. This periodicity is due to the presence of two forces that counter each other, the optical trapping force and the convection force. The material condensation at the focus is proportional to the light intensity distribution of the half circular pattern caused by the spatial phase modulation, where the outer ring is the most intense and the center point is the least intense. This distribution of the material condensation behaves as a set of prisms of different diffractive power, which cause the fanning in the lower half of the cuvette. The formation of these set of clusters at the focus cause thermal hating to build up to the level when a convection force overcomes the trapping force and forces the cluster to be removed from the focus and the fanning of the light disappears. This process repeats itself. The wash out of the cluster by convection is a gravity effect, which is absent in space. Consequently, the cluster formation by optical trapping is unopposed and its damaging effect becomes more prominent in space.

Furthermore, as it was mentioned above the gravitational force on the ground tends to settle down the contaminants on the floor of the laser system and automatically eliminate them from the way of the laser beam. On the other hand, these contaminants in space fly aimlessly in the laser compartment. With time, the laser beam traps many of them and form clusters that eventually cause optical damage.

Experimental Results

1. More contaminants enhance the damage process:

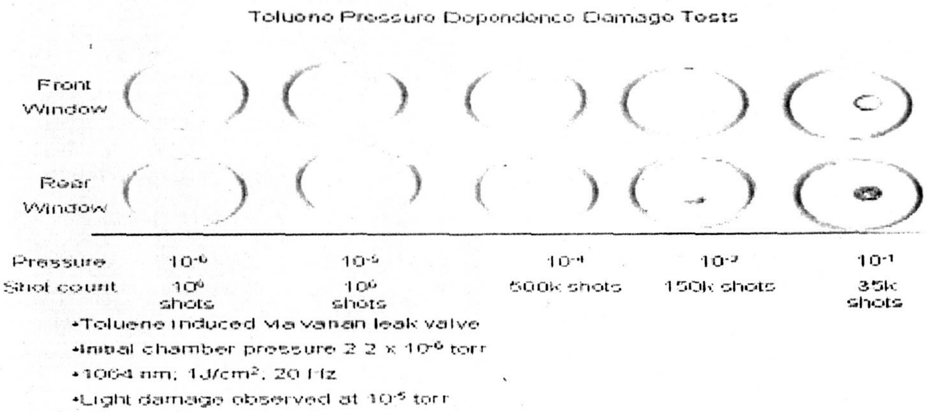


Figure 5. Damage tests caused by Toluene at different pressures

The data above reflects the damage induced on the optical window by increasing the Toluene concentration, using a Nd:YAG laser at 1064 nm of energy density 1J/cm^2 (130 mJ/pulse with 0.13 cm^2 spot) at 20 Hz. No trace of damage was seen after a million laser shots on any of the optical windows at pressures of 10^{-6} and 10^{-5} Torr of toluene. While after only 35,000 laser shots and 10^{-1} Torr, the damage was very obvious on both the front and the rear windows.

2. Contamination caused damage below the inherent laser damage threshold:

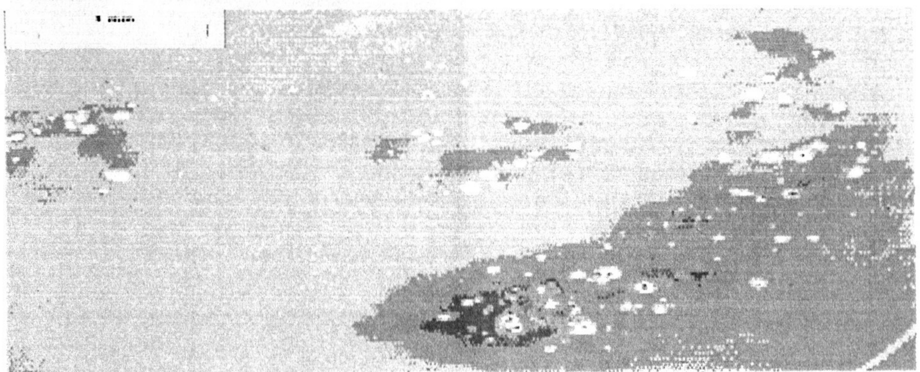


Figure 6. Damage on BBO crystal at 1MW/cm^2

The damage shown above on the surface of a BBO crystal took place at an energy density of only ~1 MW/cm^2 , which is much less than the damage threshold of 2.6GW/cm^2 for the crystal. This damage is attributed to the presence of contamination and the associated changes in the optical properties that necessarily occur upon the deposition of the contaminant. Additionally, changes of the laser induced damage threshold on the order of one thousand fold are virtually unheard of in the absence of contamination.

3. Optical damage caused by the nonlinear self-focusing:

A diode pumped solid state laser with a face pumped 22 bounce total internal reflectance Nd:YAG slab laser (Figure 7) was found to have been damaged at the last few bounces of the high reflection coated surface(Figure 8).

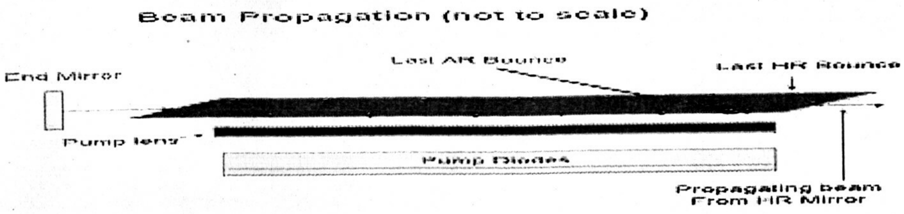


Figure 7. The diode pumped Nd:YAG slab.

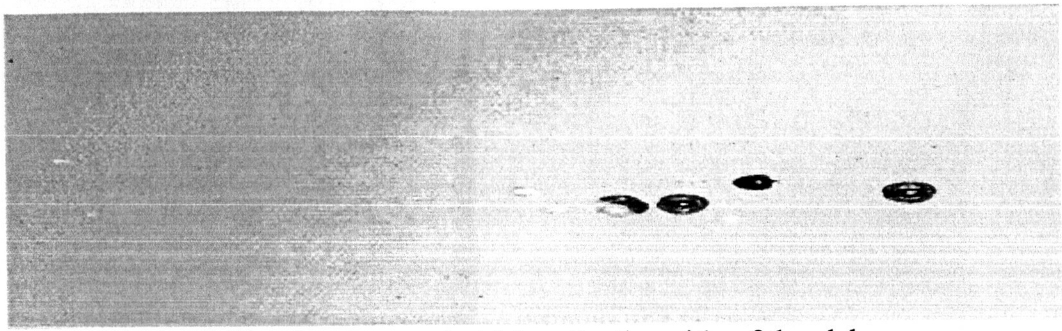


Figure 8. The damage on the high reflection side of the slab

Additionally, damage also occurred on the antireflection side (Figure 9).

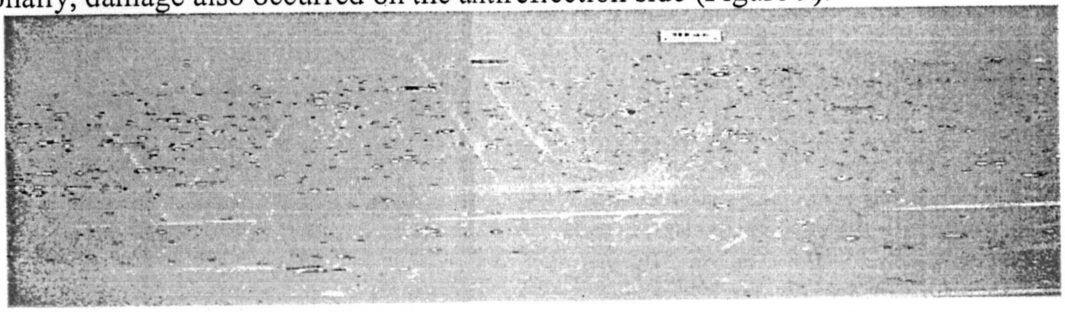


Figure9, A 100X magnification of the damage sites on the antireflection surface of the slab

The laser was subsequently cleaned and reassembled with a replacement laser slab. After alignment and readjustment, the slab was inspected and found to have identical damage profile on the antireflection pump face.

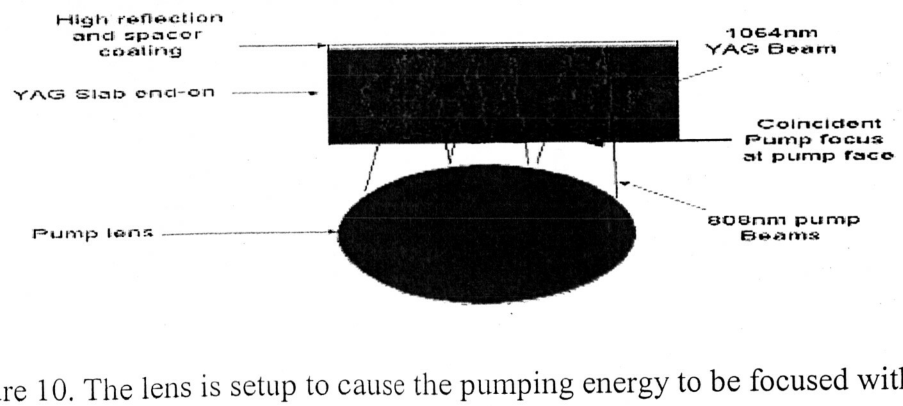


Figure 10. The lens is setup to cause the pumping energy to be focused within the slab.

It was eventually determined that the pump lens was misplaced resulting in the convergence of the pump light at the pump face after traversing the gain medium. This resulted in a factor of three enhancement of the pump intensity over the designed value of ~3 kw/cm^2, which was too low to directly cause damage. It was determined that as the pump focal point was moved away from the pump face, the damage rate fell from 200 damage sites per ten million shots to zero damage points per ten million shots. The many damage sites on the AR coating were attributed to the self-focusing and filamentation, which led to delamination of the AR coating as shown in Figure 11.

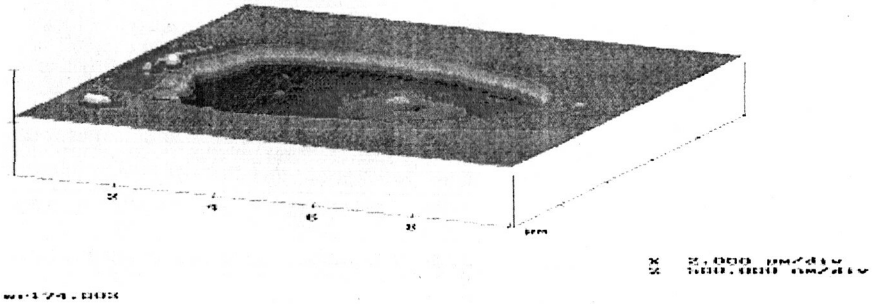


Figure11. Scanning probe micrograph of the antireflection coated surface, coating blister hole.

Closer examination of the damage sites revealed that the damaged points are on the order of fifty nanometers in diameter as shown in figure 11. This in effect proved the presence of filamentation within the laser slab due to hot spots induced by self-focusing.

5. Contamination damage by induced spatial self-phase modulation:

Figure 12 below[8] show the damage cause by 1,2,3-trichlorobenzene in nitrogen due to spatial self-phase modulation pattern formation.

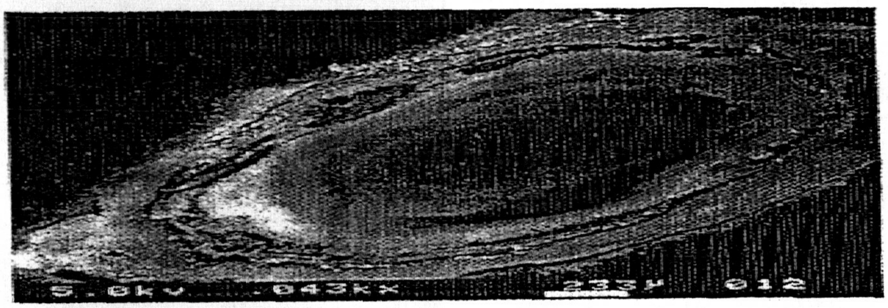
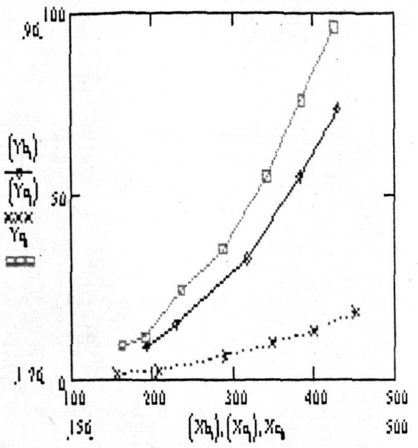


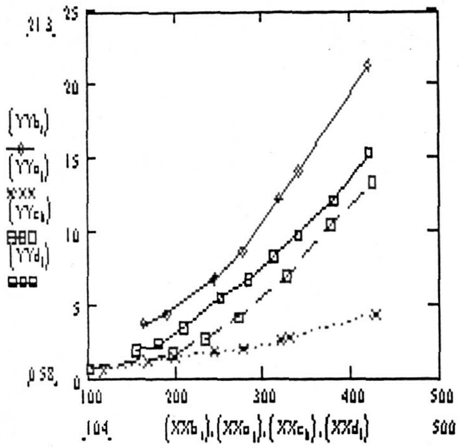
Figure12. 43x SEM photo of damage due to 1,3,5-trichlorobenzene in nitrogen

Radiation

A test setup was built to examine the effect of proton irradiation on the second harmonic generation (SHG) efficiency of BBO and KTP nonlinear crystals. The SHG efficiencies for both crystals were measured before and after irradiation at different levels of input power of the fundamental from a Nd:YAG laser at 10 Hz, 8ns pulse duration, and 6mm beam diameter. The proton irradiation was performed at Indiana University Cyclotron Facility. The crystals were irradiated for 1000s with a beam of protons, of energy 200MeV and flux of $1x10^{12}$ protons/cm^2. Due to the residual radiation caused by proton activation, post-testing was delayed by almost a month to allow the radioactivity of the crystals and holders to decay. The data in figure 13 show the SHG efficiency for each of the crystals. The SHG data were taken every other week at different power levels of the 1064nm, ranging between 100-450 mW, i.e. power density range between 5.3 and 15.9 Mw/cm, without focusing.



(A)



(B)

Figure 13. (A) the SHG measurements of the KTP crystal as a function of the input power at later days from the time of irradiation. (B) the SHG measurements of BBO at later days. X-axis represents the input IR in mW and Y-axis is the SHG in mW.

The first experiment after irradiation showed that the efficiency of conversion to SHG for both of the crystals went down by more than 5-times. By repeating the measurements every two weeks, it was found that both crystals were going through self annealing that is caused by the laser and recovered themselves totally to their initial conditions after nearly a month from receiving them. The annealing of the crystals by the laser is due to the heating induced by the F-centers' absorption. It is worth noting here that the healing of an irradiated KTP crystal, that was irradiated under the same circumstances of the above two crystals, took place instantaneously using a different laser at ~9-times the power density of the laser used in the above experiment.

Proposed Methodology for minimizing the role of contaminants in damaging a space laser[9].

Preliminary adsorption experiments were performed to examine the adsorption of different trace elements onto different surfaces of various metallic materials. In these experiments, a few metallic sheets were inserted into a PYREX glass desiccator that contains a small concentration of trace gases diluted in pure nitrogen atmosphere (figure 14). The desiccator is outfitted to a multi-path absorption cell, an FT-IR spectrometer to monitor the instantaneous concentration of the adsorbing molecules concentrated in the nitrogen atmosphere in the desiccator. Several metallic test surfaces such as copper, stainless steel, and aluminum were tried. Trace gases of HCl, NO_2, SO_2, and NH_3 were examined at low concentrations of a few hundred parts per million in a pure N_2 gas atmosphere. It was found that the reaction rates for most of the atmospheric trace gases being investigated were very fast. The data below (figure 15) show the spectra of NO_2 at the instantaneous concentration of 30ppm of the trace gases in N_2 at different time intervals. It is now proposed to adopt this adsorption mechanism in future spaceflight lasers to eliminate or minimize the contaminants in the laser compartment. In doing so, a long tape of the adsorbing metal will pass slowly through the optical compartments and be taken up on take-up reel (Figure16).

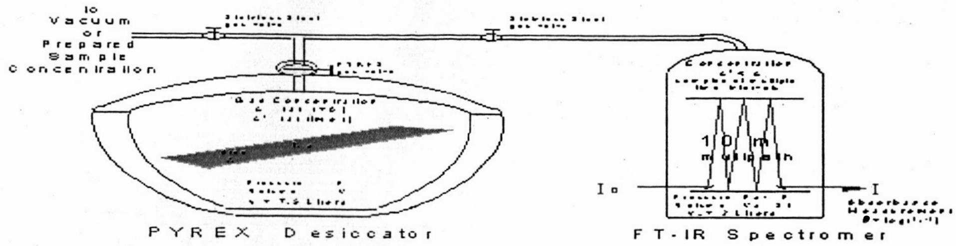


Figure14. Desiccator Adsorption Experimental Arrangement.

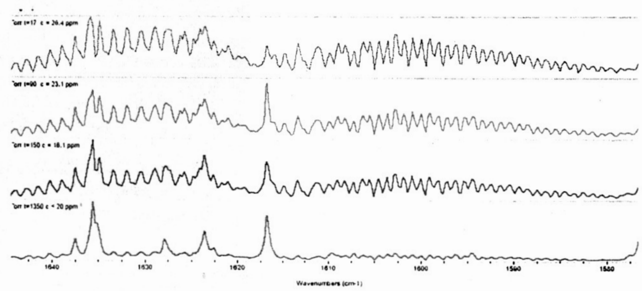


Figure15. Time interval absorbance spetra of NO_2-Aluminum interface for NO_2 @ 30 ppm in N_2 exposed to 1300 cm^2 Aluminum surface.

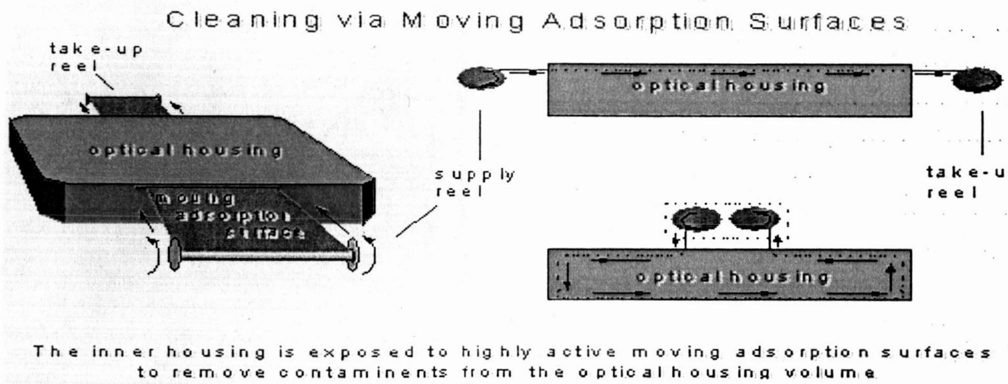


Fig. 16. The proposed system to adsorb the contaminants out of the laser compartment.

Summary and Conclusion

In summary, it was shown that the optical damage, in many cases, is a contamination driven issue that drives a set of nonlinear phenomena such as self-focusing, filamintation, Kerr effect, and spatial self-phase modulation. We have demonstrated that contamination leads to the induction of optical damage even when the laser power density is significantly less than the inherent laser induced optical damage thresholds of the components used. We also explained and demonstrated the trapping forces in the form of radiation pressure and electrostrictive forces induced on the contaminants by the laser beam, which can eventually lead to the formation of clusters on the surfaces of the optical components. We plan to make use of the adsorption mechanism and propose a methodology to minimize or eliminate the contaminants presence in the laser compartment. The effect of irradiating the KTP and BBO crystals with a high-energy beam of protons on their SHG was examined. It was found that these crystals can be annealed by the laser and self-heal themselves with time depending on the power density of the laser.

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