# Qualifying Exam

Peter Langston

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# Papers to be Discussed

1) Ming Zhou, et al., “Effect of multiple wavelengths combination on laser-induced damage in multilayer mirrors,” Optics Express, Vol. 17, No. 22, 26 Oct. 2009

2) Marco Jupe, et al., “Calculations and experimental demonstration of multi-photon absorption governing fs laser-induced damage in titania,” Optics Express, Vol. 17, No. 15, 20 July 2009

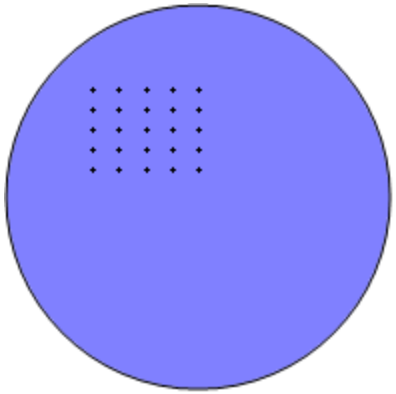
# Outline

- Laser damage
  - Long Pulse (CW)
  - Short Pulse (reduced thermal effects)
- First Paper
- Second Paper
- Open To Questions

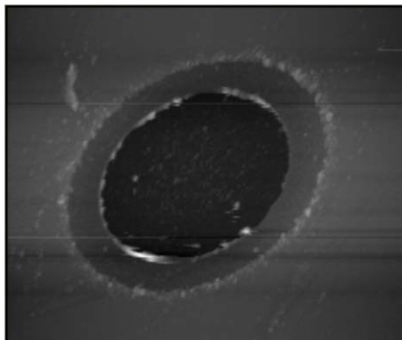
# Laser Damage

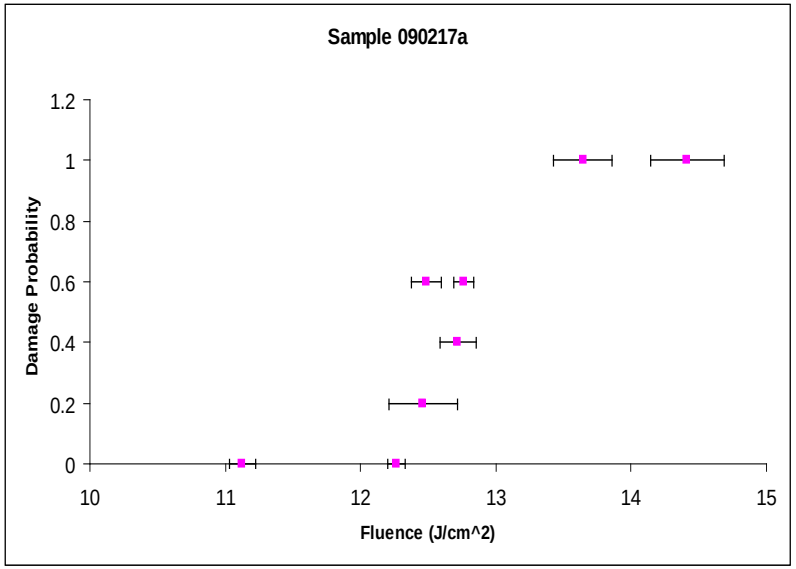
- Long Pulse (CW) >20ps
  - Thermally driven
  - Melt or thermal explosion (UV)
- Short Pulse (reduced thermal effects) <10ps
  - Multiphoton ionization
  - Plasma formation leading to thermal explosion
- LIDT vs. Damage Probability
  - Damage probability is usually expressed as a curve
  - LIDT is a single point on the probability curve

# 1-on-1 Laser Damage Testing



Test optic damage pattern






Typical damage probability curve

# First Paper

- Ming Zhou, et al., “Effect of multiple wavelengths combination on laser-induced damage in multilayer mirrors,” Optics Express, Vol. 17, No. 22, 26 Oct. 2009
  - Thermal model

# Motivation

- To understand the damage mechanism in optical coatings when multiple wavelengths (1064nm, 354nm) are illuminating the surface at the same time.
  - Two general ideas
    - Equivalent damage effect
    - IR may not contribute much to damage growth

# Experimental Setup

Q switched YAG

12ns at 1064nm

8ns at 354nm

Polarizing attenuator

Spot size at $1/e^2$

Fundamental 552μm x 685μm

Third harmonic 241μm x 424μm

Delay for third harmonic

HeNe scatter probe

No information on beam spatial beam profile

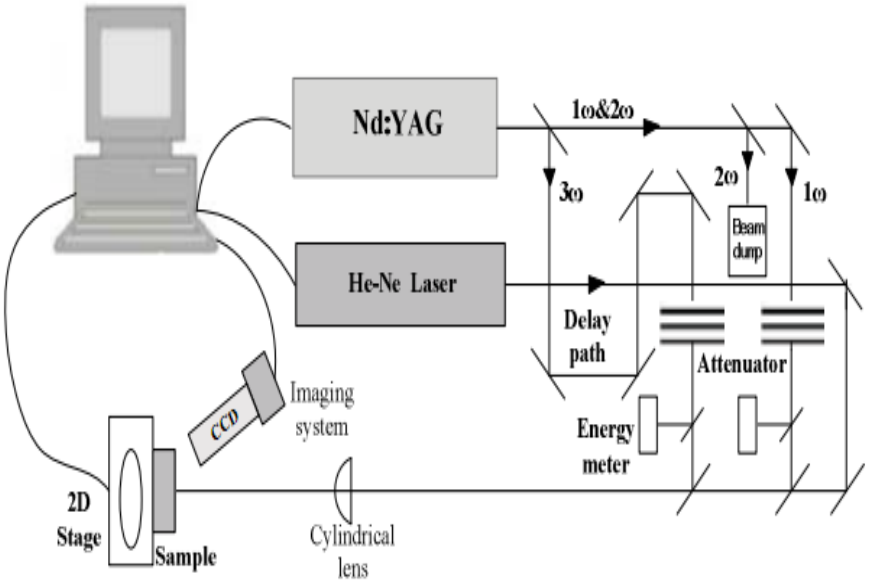



Fig. 1. The laser damage test setup for two wavelengths combined irradiation

# Samples

- E-beam evaporated
- Substrate was K9 (Chinese equivalent to BK7)
- Grown as beam splitters
  - 95.6% transmission at 1064nm
  - 99.9% reflection at 354nm
- All layers were quarter wave at 351nm
- Film structure was $(HL)^{13}3.3H2A1.6L$
  - Where H ($HfO_2$), A ($Al_2O_3$) and L ($SiO_2$)

# LIDT Testing Results

LIDT Fundamental only ~9J/cm2

LIDT Third Harmonic only ~7J/cm2

Fundamental fluence for testing:

S1 = 1.9J/cm2

S2 = 4.0J/cm2

S3 = 5.4J/cm2

S4 = 6.5J/cm2

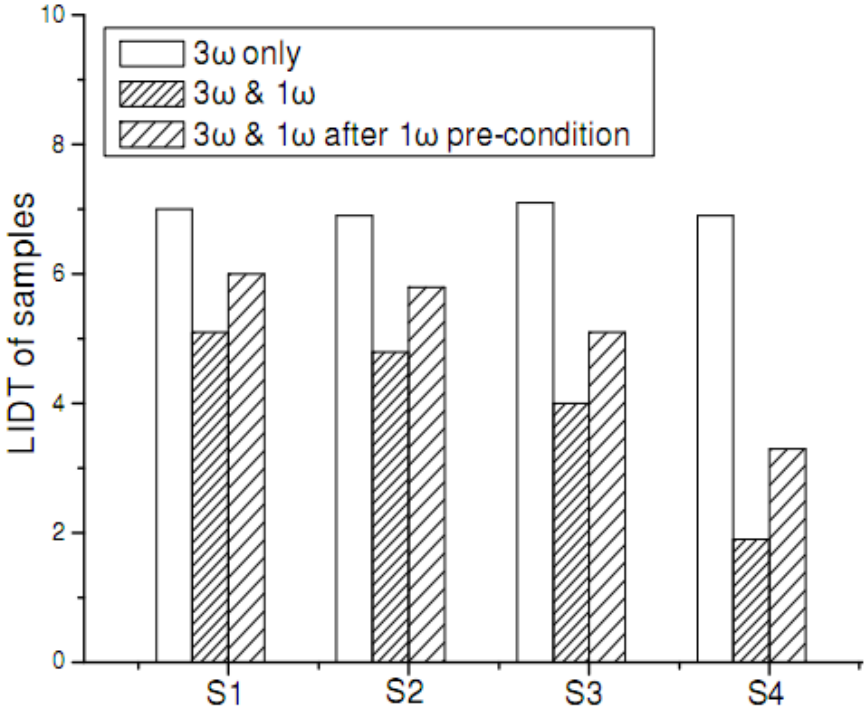
LIDT after conditioning at 1064nm:

S1 = 5.3J/cm2

S2 = 4.8J/cm2

S3 = 4.0J/cm2

S4 = 1.9J/cm2




Assumption: LIDT numbers plotted above for 3ω & 1ω is reporting only the 3ω fluence and not the 1ω fluence.

# LIDT Analysis

- Effect of two λ LIDT testing
  - Need to deconvolve 1ω from 3ω effects in test results
- Define the contribution function $R_{1\omega}$

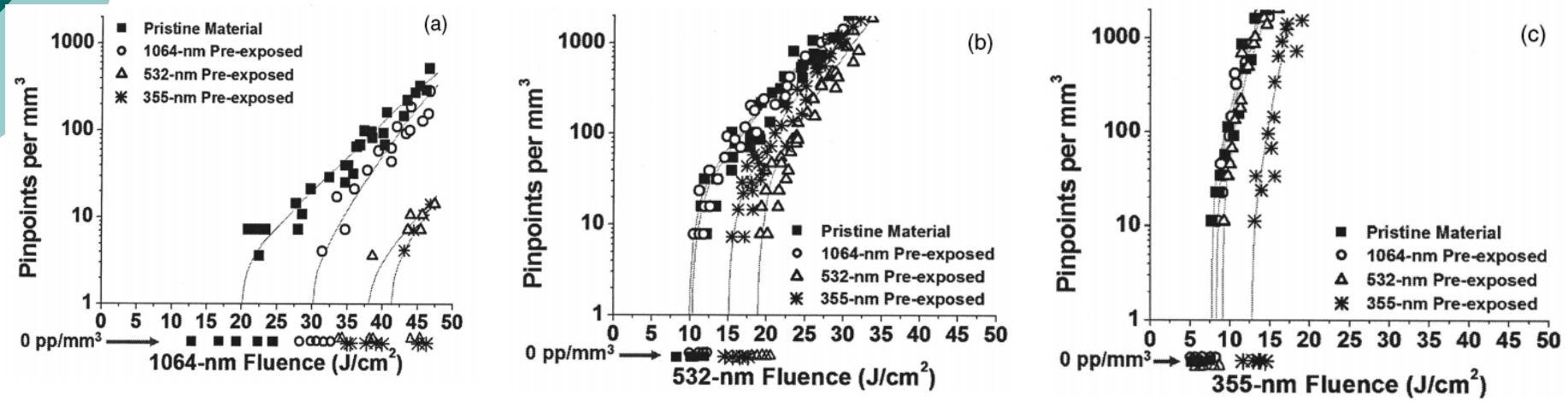
$$R_{1\omega} = \frac{\Phi_{3\omega} - \Phi_{3\omega+1\omega}}{\Phi_{1\omega}}$$

  - Where $\Phi_{3\omega+1\omega}$ is the LIDT for two λ combined irradiation
  - $\Phi_{3\omega}$ is the LIDT for 3ω only and $\Phi_{1\omega}$ is the 1ω fluence in combined LIDT fluence.

    $R_{1\omega}$ = 0.7, 0.4, 0.5, 0.9

# Laser Conditioning (Defects)

- Laser conditioning, rapid heating of high absorbing sites can result in ejection of contaminant from the top few layers. If done at the proper fluence no plasma formation occurs.

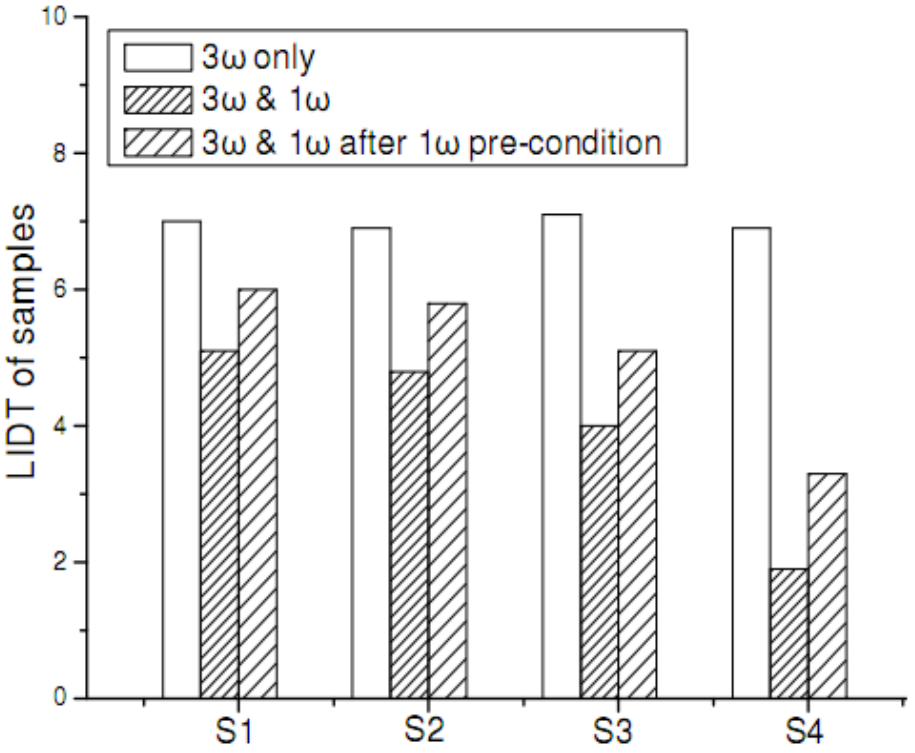



- Laser conditioning at 1ω can significantly improve the LIDT of the sample tested with 1ω but will have little effect on the LIDT when tested at 3ω.

P. DeMange, "Multiwavelength investigation of laser-damage performance in potassium dihydrogen phosphate after laser annealing," Optics Letters, Vol. 30, No. 3, Feb. 1, 2005

# Effect of Laser Conditioning

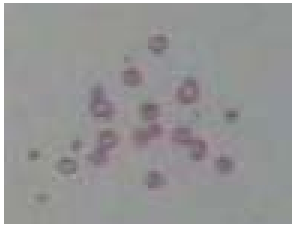
- Laser conditioning will address the defects that are most susceptible to light at 1ω.
- In all cases the damage threshold was improved after laser conditioning.



# Damage Initiation

- **Perfect crystal (no defects)**
  - Multiphoton Ionization (MPI)

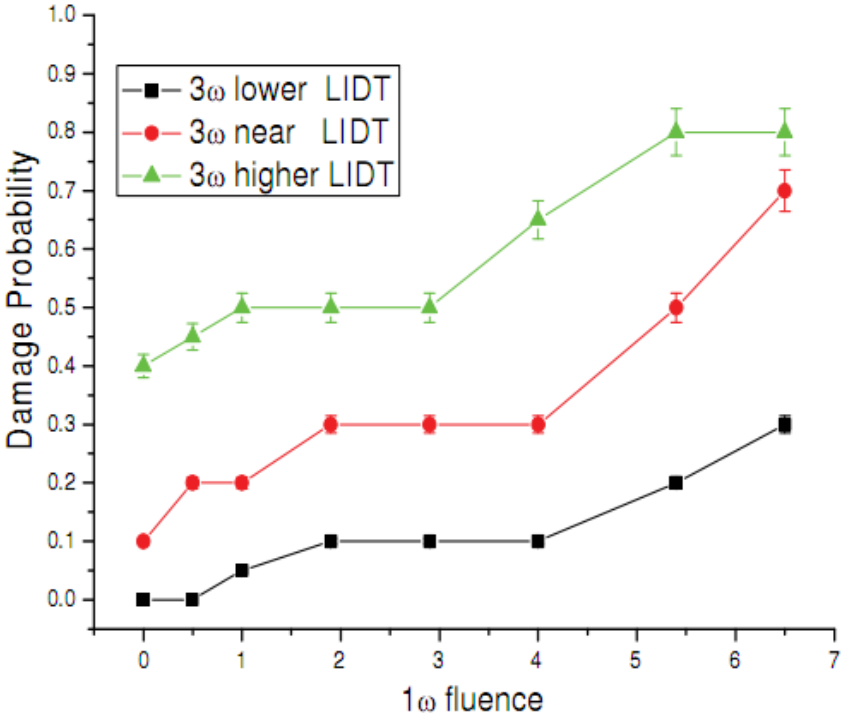
    band gap picture
- **Defects in coating**
  - Direct absorption (UV explosion)
  - Concentration enhanced (MPI)
- **Damage behavior**
  - Perfect crystal (no defects)
    - MPI at the point of highest field
  - Defect in coating
    - Localized damage
    - Probabilistic



# Probability Curves

**Three probability curves were generated:**

- Lower LIDT 3ω 5.5J/cm$^2$
- Near LIDT 3ω 7.3J/cm$^2$
  - 10% damage prob.
- Higher LIDT 3ω 8.0J/cm$^2$
  - 40% damage prob.




- The most significant growth occurs above 4J/cm$^2$
- This rapid increase could support the claim that 1ω plays an increasing role in the damage process

* Lost In Translation?

# Damage Morphology

**All damage sites exhibit signs of small defect initiation.**

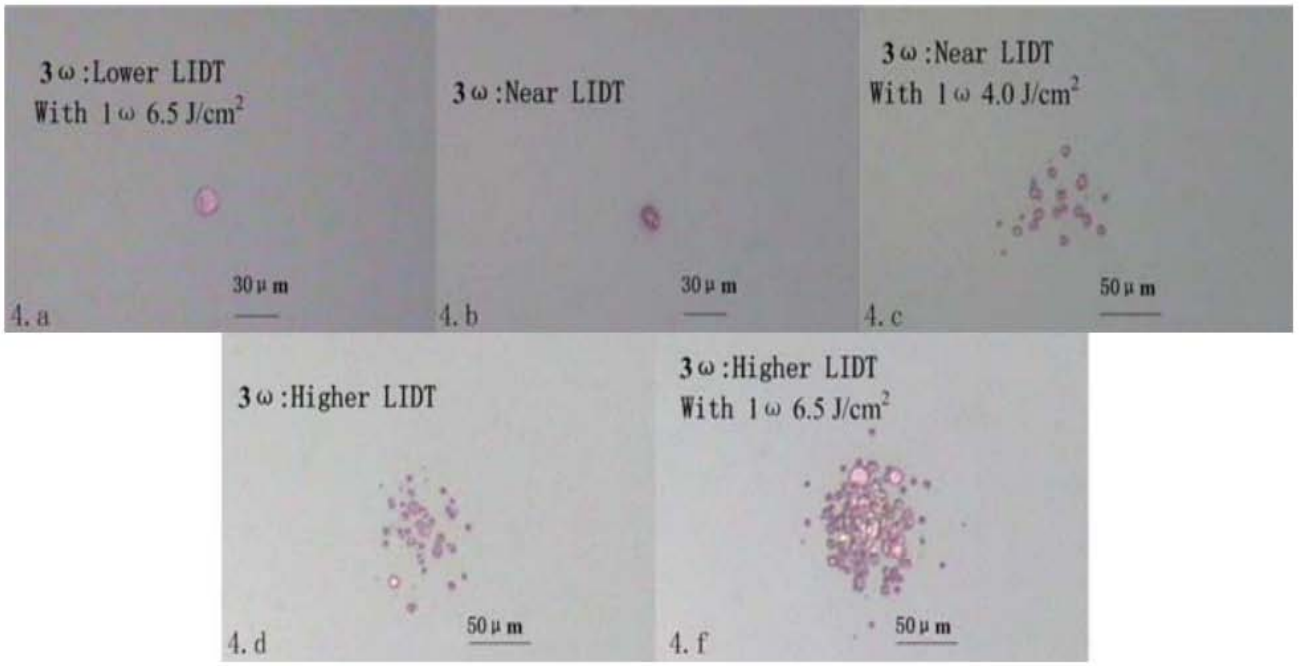



Fig. 4. The surface morphology of samples which were irradiated by different fluence combined of 1ω and 3ω

# Small Absorbing Particle Model

- Working Assumptions
  - Small absorbing defects are the initiators of damage
  - Only one defect type is the cause (size, absorption cross section)

Power absorbed by defect: $Q_i = \frac{3\sigma_i I}{4\pi a^3}$

where $\sigma$ is the absorption cross section

From paper ⇒ $\sigma_i = (1-R)\frac{16\pi^2 a^2 k_i}{\lambda_i}\int_0^{\pi/2} \sin\varphi(\cos\varphi)^3 d\varphi$

From Geometric Optics [2] ⇒ $\sigma_i = (1-R)\frac{16\pi^2 a^3 k_i}{\lambda_i n}\int_0^{\pi/2} \cos\varphi\sqrt{n^2-\sin^2\varphi}\sin\varphi d\varphi$

[1]. "Laser damage threshold for dielectric coatings as determined by inclusions," David Milam and R. A. Bradbury and Michael Bass, Applied Physics Letters, Vol. 23, No. 12, 15 Dec. 1973

[2] Bohren, et al. "Absorption and scattering of light by small particles," John Wiley & Sons Inc. 1983

# Temperature Evolution in Defect

**The temperature evolution of the particle and the surrounding film is given by the heat equation.**

- For the defect: $$c\rho\frac{\partial T_p}{\partial t} = K_0\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial T_p}{\partial r}\right) + Q$$

- For the film: $$c\rho\frac{\partial T_f}{\partial t} = K_0\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial T_f}{\partial r}\right)$$

where c is the specific heat, $\rho$ is the density, $K_0$ is the thermal conductivity of the respective defect/film and Q is total energy absorbed:

$$Q = Q_{1\omega}R_{1\omega} + Q_{3\omega}$$

With boundary conditions: $T_p = T_f = 0$ at $t = 0$

$$T_p = T_f \qquad C_p\frac{\partial T_p}{\partial r} = C_f\frac{\partial T_f}{\partial r} \quad \text{at} \quad r = a$$

# Inclusion Size and its Sensitivity to Damage

Temp of inclusion is max when: $\frac{a^2}{4D} = 0.81\tau$

This corresponds to a optimal sized defect for absorption: [2] $2a = 3.6\sqrt{D\tau}$

"In the case of Hafnium inclusions in hafnia… the critical radius is about 150nm at 1064nm and 100nm at 355nm." [3]

Defect is assumed to be hafnium inclusion in hafnia ~150nm radius.

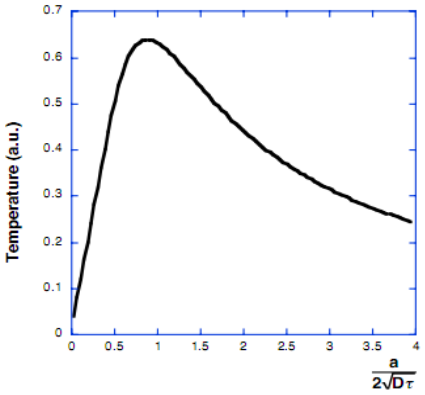



Fig.3: Inclusion temperature induced by a rectangular laser pulse for absorbers of different sizes. Particle size is normalized by the thermal length √Dτ.

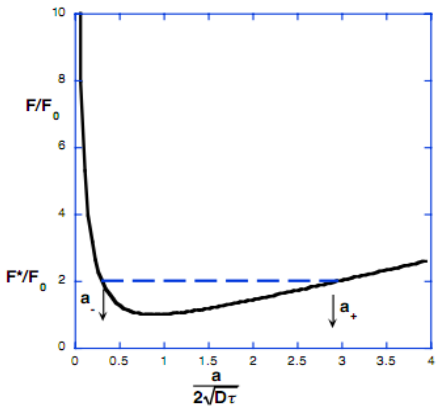



Fig.4: Normalized fluence damage threshold F/F0 vs. normalized particle size. At fluence F*, particles with sizes $a_- < a < a_+$ are above their threshold. The resulting damage density is found by integrating the precursor density n(a) between sizes $a_-$ and $a_+$.

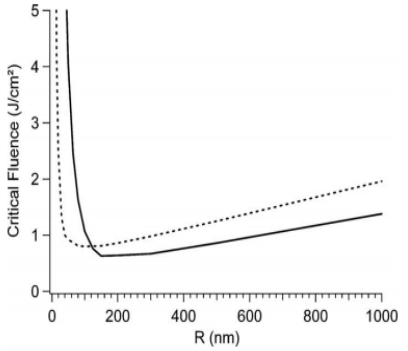



FIG. 2. Critical fluence calculated for a hafnium inclusion in a hafnia matrix. Irradiation is at 1064 or 355 nm with a pulse duration of 12 ns at $1/e$ (Gaussian temporal shape).


[3] Laurent Gallais, "Investigation of nanodefect properties in optical coatings by coupling measured and simulated laser damage statistics," J. Applied Physics, 104, 2008.

[2] M. D. Feit, "Implications of nanoabsorber initiators for damve probability curves, pulselength scaling and laser conditioning," SPIE Vol. 5273, 2003

# Numerical Simulation of Defect Temperature (Damage Initiation)

- Damage occurs when the inclusion reaches a critical temperature $T_c$. $T_c$ is the temp where hafnium begins to fuse at 2300K
- $T_c$ is only reached with 3ω ~7.3J/cm² or when 3ω +1ω near LIDT or higher
- This is consistent with earlier damage data

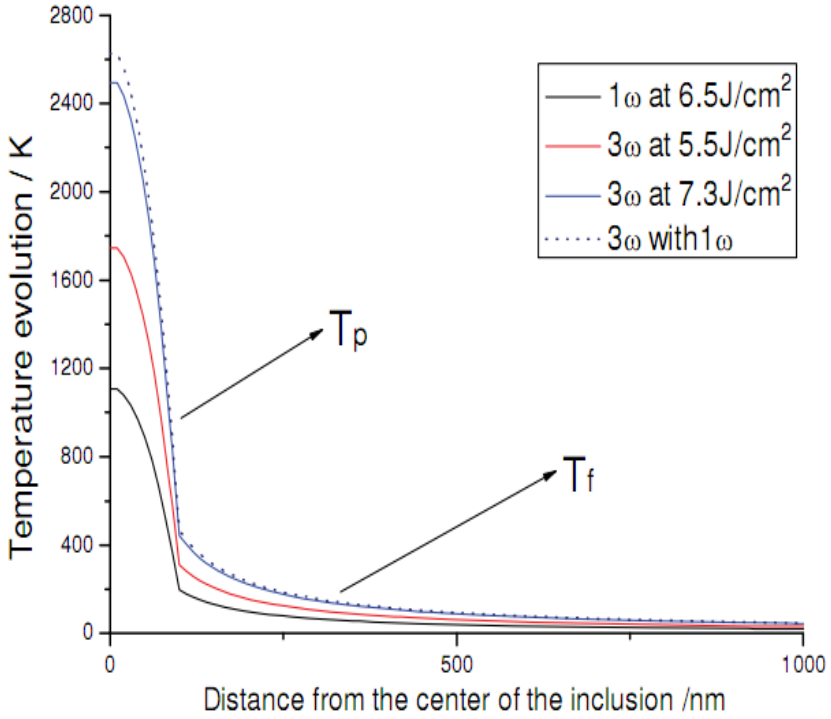



Fig. 5. The temperature rise as a function of the distance from the center of defect, where $k_{1\omega}$ = 3.1, $k_{3\omega}$ = 2.58, $a$ = 150 nm, $D_p = 9.9 \times 10^{-6}$ m²/s, $D_f = 6.8 \times 10^{-7}$ m²/s, $C_p$ = 18.4 W/m·k, $C_f$ = 1.67 W/m·k, $\tau_{1\omega}$ = 12ns, $\tau_{3\omega}$ = 8ns [22]

# Not all Defects are the Same

- There will be a defect size distribution. Those defects that cannot reach $T_c$ under $3\omega$ only may become high absorbers in the presence of $1\omega$.

- Each defect has it's own threshold fluence (T).
  The probability that at least one defect is present in the area 'A' irradiated with a fluence above T is:
  $$P(F) = 1 - e^{(-N(F))}$$
  where N(F) = number of defects in the laser spot that will damage if fluence F is seen.

- If irradiated by multiple $\lambda$: $N(F) = N_{1\omega}(F) + N_{3\omega}(F)$ where $N_{3\omega}$ is the number of sensitive defects in the beam at $3\omega$.
  Generically:
  $$N_i(F) = \int_0^F g_i(T) S(F) dT$$
  where $\iota = 1\omega, 3\omega,$ $g_i(T)$ is the defect density, S(F) is a weighting factor

# Not all Defects are the Same (continued)

- S(F) is the portion of the laser spot where the fluence is above the threshold T.
  For a Gaussian Beam: $S(F) = (\frac{\pi\mu^2}{2}) \ln(\frac{F}{T})$
- $g_i(T)$ is the surface density of defects that will damage at fluences between T and T + dT

  Assuming a Gaussian probability distribution of defects:

$$g(T) = \frac{2d}{\Delta T\sqrt{2\pi}} e^{\left[-\frac{1}{2}\left(\frac{T-T_0}{\Delta T/2}\right)^2\right]}$$

  where the ensemble function g(T) depends on three parameters:
  - $T_0$ the threshold mean fluence
  - $\Delta T$ the threshold standard deviation (full width $1/e^2$)
  - And 'd' the defect density:

$$\int_0^\infty g(T)dT = d$$

# Theoretical Comparison of Probabilities

**Theoretical probability compared with measured probability**

Defect densities:

$d_{3\omega} = 5.5 \times 10^5$ mm$^{-2}$

$d_{1\omega} = 1.6 \times 10^5$ mm$^{-2}$

$d_{1\omega'} = 8.2 \times 10^4$ mm$^{-2}$

From Plot:

- If defects susceptible to 1ω are conditioned out, LIDT will be increased
- Assuming comparable test parameters (paper does not clarify) the theoretical fit seems to follow measured data

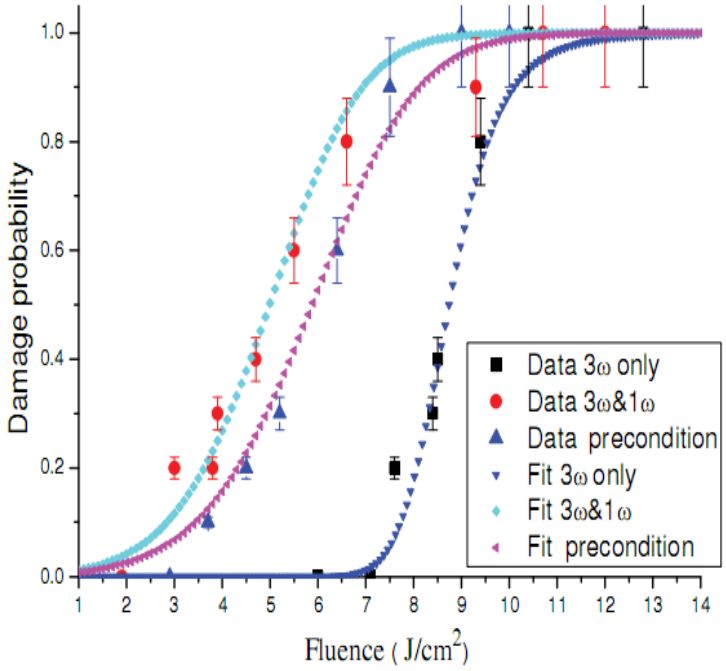



Fig. 6. Numerical analysis for the irradiations of 3ω only, 3ω with 1ω, and 3ω with 1ω after 1ω pre-condition, respectively, where $T_0 = 9.0 J/cm^2$, $\Delta T = 3.0 J/cm^2$, $\mu_{3\omega} = 380 \mu m$

# Conclusions

- Their conclusion is that reducing the number of sensitive defects in the coating is a good way to increase the LIDT of a coating.
- This paper was a good initial investigation into the effect of multiple frequency damage processes. It identified possible topics for additional study. 3ω defects removal.
- They draw some conclusions about the effect of the fundamental that seems too general given the relatively small effect demonstrated in the data. I would prefer to have seen more emphasis on the demonstration of the nonlinear effect the fundamental seems to have. This could have been done by simply adding more data points to the initial damage testing and accounting for kinetic energy being imparted to conduction band electrons.
- They gave no indication as to how they arrived at the defect densities for the last damage probability curves.
- I did not like the way they presented the LIDT of the samples tested under multiple wavelengths. This was very confusing.
- They also made statements in the paper that seem to contradict the flow of the rest of the paper.
- They pretty much ignore the effect of avalanche ionization.
- Too many unknowns in the paper to follow their investigative process with confidence.
- If you're willing to ignore avalanche effects, the paper seemed to argue their point well.

# Second Paper

- Marco Jupe, et al., “Calculations and experimental demonstration of multi-photon absorption governing fs laser-induced damage in titania,” Optics Express, Vol. 17, No. 15, 20 July 2009
  - Rate Equation Model

# Motivation

- As femtosecond lasers have become more common place their uses have greatly increased which, as always, increases the demand for them and for greater performance.

- Optical elements in the laser see:
  - Higher pulse energy
  - Shorter pulse widths (lots of λ's)
  - Longer lifetime

# Assumptions

- LIDT in the fs regime is dominated by Multiphoton Ionization (MPI)
- The electronic structure of the material is the LIDT limiting factor
- Characteristic damage behavior should be observable that will support these claims
  - Defect free band structures are quantized
  - Photon energy is also quantized

# Damage Process

- Damage to optics from fs lasers is assumed to be dominated by electronic damage
- Electrons are ionized leading to a critical electron density
- This electron density is calculated from the plasma frequency of the target's free electrons

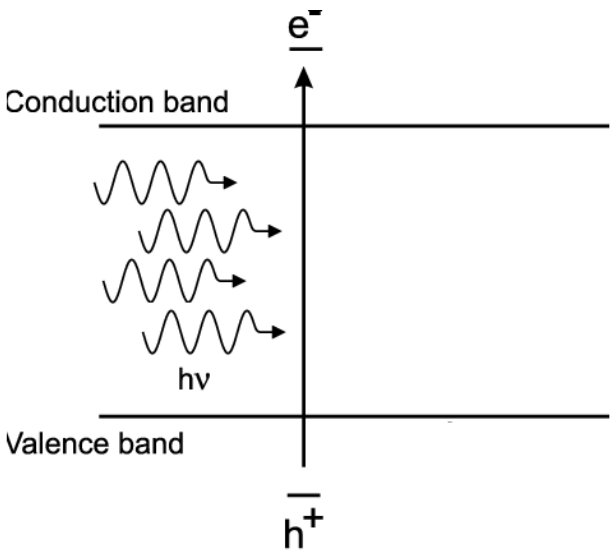
$$\omega_p = \sqrt{\frac{Nq^2}{\varepsilon_0 m}}$$

where N is the number density of free electrons, q is the charge (electron charge), $\varepsilon_0$ is the permittivity of free space and m is the mass of an electron.

# Damage Modeling

A rate equation model is used to describe the number of electrons in the conduction band.

$$\frac{\partial \rho(t)}{\partial t} = W_{PI}(I(t)) + W_{AV}(I(t), \rho(t)) - W_{rel}(\rho(t), t)$$




where the change in the electron density with time is: $\frac{\partial \rho(t)}{\partial t}$

The photo ionization rate is: $W_{PI}(I(t))$

The avalanche ionization rate is: $W_{AV}(I(t), \rho(t))$

$W_{rel}(\rho(t), t)$ is all relaxation process rate

# Keldysh Theory

**Keldysh theory expresses the ionization probability as a function of the frequency of the electric field oscillations.**

$$W_{PI}(I(t)) = \frac{2\omega_0}{9\pi}\left(\frac{\omega_0 m}{\hbar\sqrt{\Gamma}}\right)^{\frac{3}{2}} Q(\gamma, x) e^{\left(-\pi\frac{K\left(\sqrt{\Gamma}-E\sqrt{\Gamma}\right)}{E\sqrt{\xi}}\right)} ]x+1[$$

$$\gamma = \frac{\omega_0}{e}\sqrt{\frac{mU_i c\varepsilon_0 n_0}{2I}}$$

$$\Gamma = \frac{\gamma^2}{\gamma^2+1}$$

$$Q(\gamma, x) = \sqrt{\frac{\pi}{2K\sqrt{\xi}}}\sum_{n=0}^{\infty}\{e^{\left(-n\pi\frac{K\left(\sqrt{\Gamma}-E\sqrt{\Gamma}\right)}{E\sqrt{\xi}}\right)}\Phi\sqrt{\eta(n+2\mu)}$$

$$x = \frac{2U_i}{\pi\hbar\omega_0\sqrt{\Gamma}}E\sqrt{\zeta} \qquad \mu = ]x+1[-x \qquad \eta = \frac{\pi^2}{2K\sqrt{\zeta}E\sqrt{\zeta}} \qquad \zeta = \frac{1}{\gamma^2+1}$$

$$\Phi = \int_0^z e^{(y^2-z^2)}dy \qquad K(k) = \int_0^{\pi/2}\frac{1}{\sqrt{1-k^2\sin^2\phi}}d\phi \qquad E(k) = \int_0^{\pi/2}\sqrt{1-k^2\sin^2\phi}\,d\phi$$

An analytical solution to this expression is not available so the Keldysh ionization rate is determined numerically

# Limiting Cases $\gamma = \frac{\omega_0}{e}\sqrt{\frac{mU_i c\varepsilon_0 n_0}{2I}}$

$$\gamma << 1$$

- Slowly varying (low frequency) very strong electric field

  same as the probability of a static charge tunneling through a potential barrier
- In this case the band gap is deformed by the extremely high field strength acting on the sample during the pulse
- Tunneling probability is dictated by the instantaneous value of the field intensity and predicts a continuous variation of the ionization probability with photon energy

$$\gamma >> 1$$

- High frequency, high field
- The electron doesn't have enough time to tunnel through the barrier in less than one oscillation of the optical field
- This is the case for MPI where an electron jumps the band gap due to instantaneous absorption of multiple photons

# Rationale & Selected Material

- If MPI is the dominant process there should be an abrupt change in the ionization probability/rate when the process switches from n- to n+1 photon absorption.
- Measurable with the appropriate selection of photon and band gap energies.

For $TiO_2$

- $\gamma$ ~ 3 at the peak fluence

  mild tendency toward MPI

- LIDT of coatings is usually limited by the high refractive index material

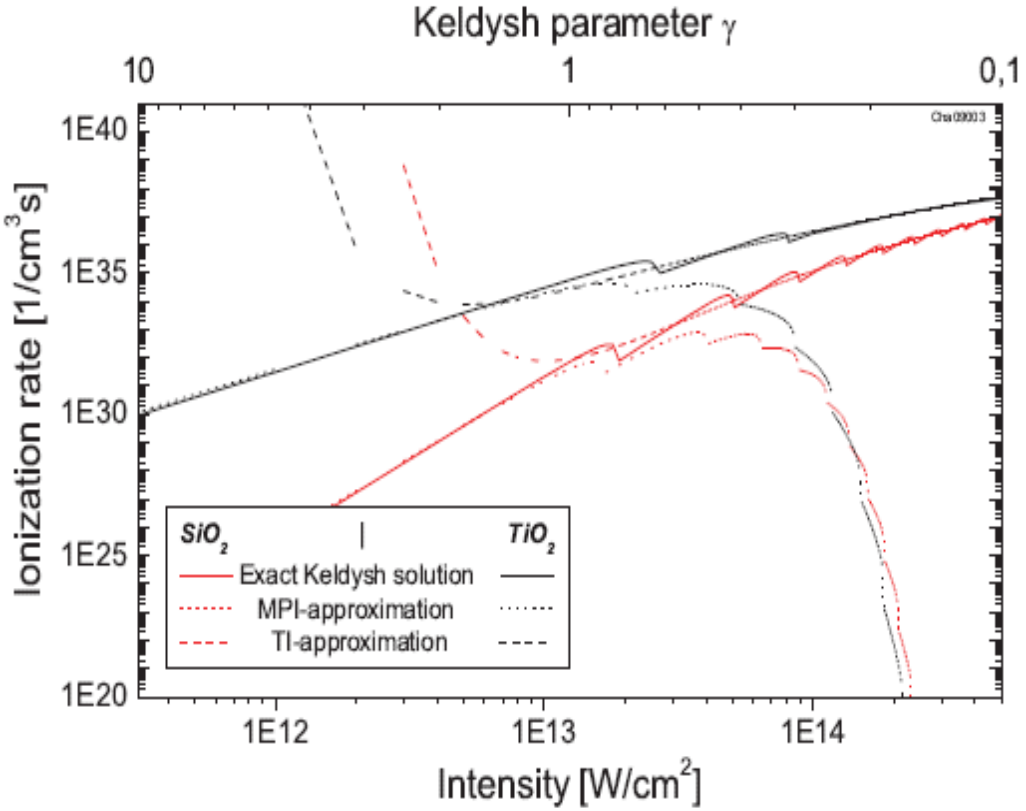
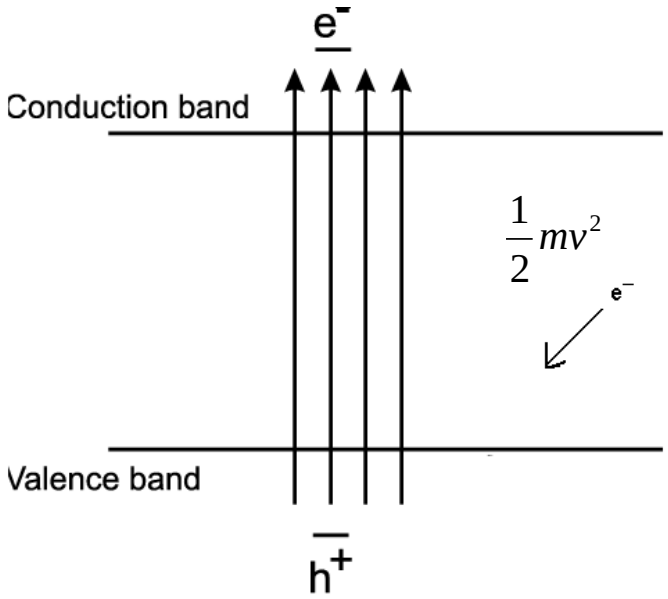



Fig. 1. Ionization rates in fused silica and $TiO_2$ - top scale: $W_{PI}$ [Keldysh parameter] bottom scale: $W_{PI}$ [Intensity] (at $\lambda$=800nm)

# Avalanche Ionization $W_{AV}(I(t),\rho(t))$

- The second major contributor to conduction band electron populations
- Is seen when electrons in the conduction band hit electrons in the valance band. K.E. > $E_g$
- The Drude model of electron interactions is used to calculate:



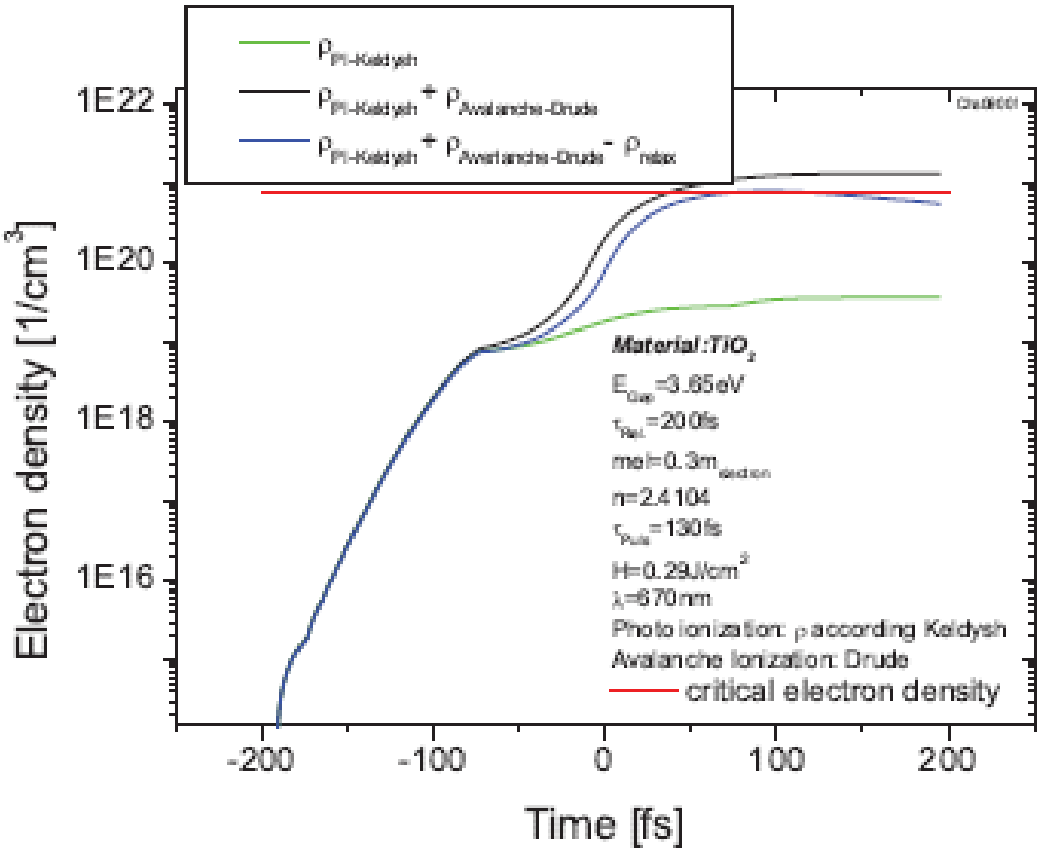

$$W_{AV}(I(t),\rho(t)) = \frac{\sigma}{U_i}\rho(t)I_0$$

$$\sigma = \frac{e^2}{c\varepsilon_0 n_0 m}\frac{\tau_c}{1+\omega^2\tau_c^2}$$

$$\tau_c = \frac{16\pi\varepsilon_0^2\sqrt{m(\frac{U_i}{10})^3}}{\sqrt{2}e^4\rho}$$

# Avalanche Ionization Cont. + Recombination $W_{rel}(\rho(t),t)$

- Before avalanche ionization can become a significant influence in the electron population there needs to already be a significant population of electrons in the conduction band.




- Recombination term accounts for all depopulation processes
- ~ 130 fs pulses
- Any recombination process that is much greater then 130 fs has very little effect

# Effect of Multiphoton Ionization on LIDT

Because there is a huge difference in the absorption cross section for n+1 opposed to just n it is very reasonable to assume a quantized behavior in damage threshold data.

$$\sigma_{(N)} = \frac{1}{t_{eff}\left(\frac{I_{sat}}{\hbar\omega}\right)^{N}}$$

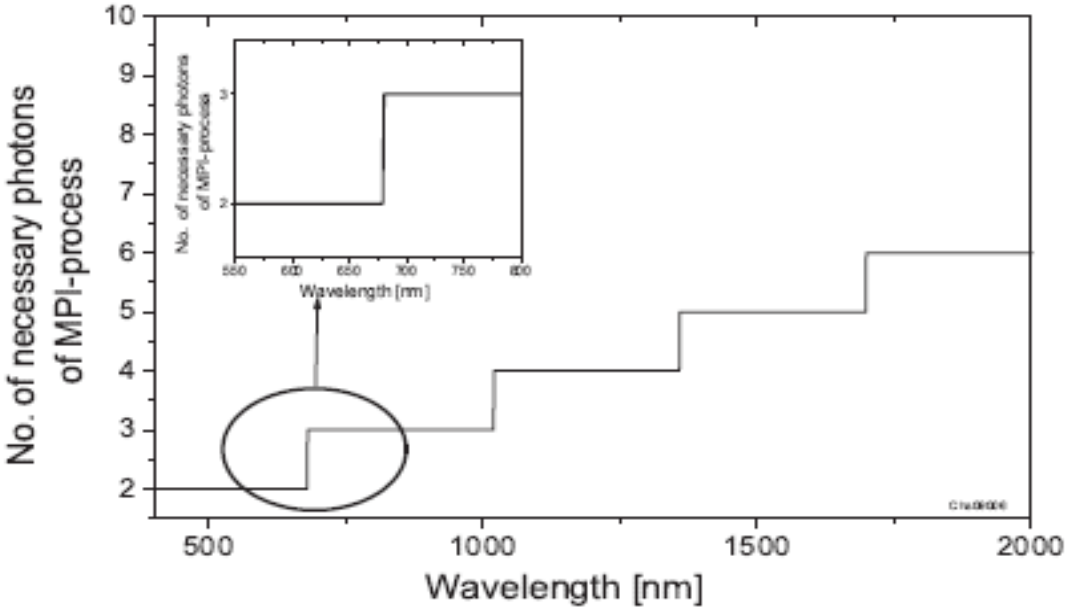



Fig. 3. Ionization steps of the applied material calculated from the total losses

# Calculations

- Ionization rates as functions of power density increase until a transition from 2 to 3 photon process
- Fig #3 shows the calculated transition energy in terms of wavelength for the TiO2 film.

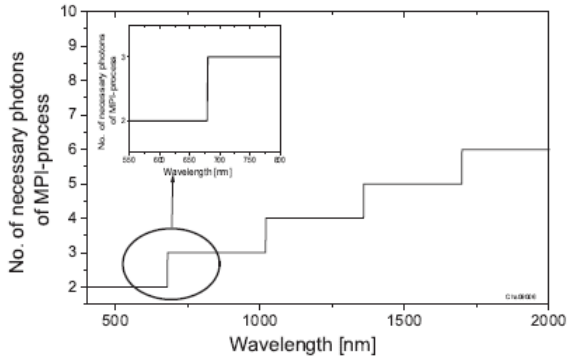



Fig. 3. Ionization steps of the applied material calculated from the total losses

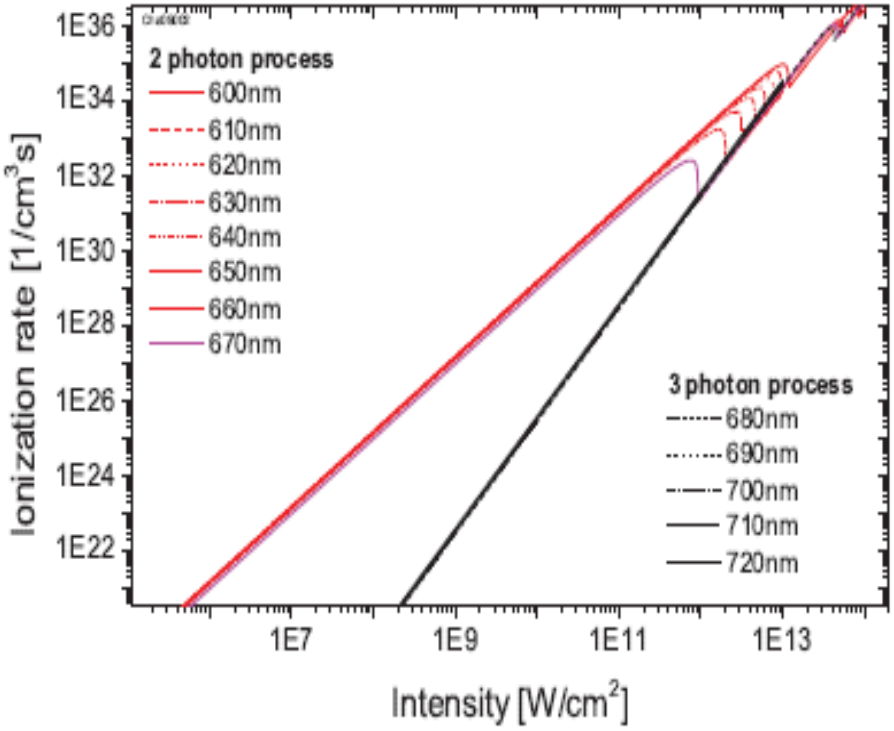



Fig. 4. Calculated ionization rate according to Keldysh's theory. Obviously, the ionization characteristic is changed abruptly from 670nm to 680nm. This behavior indicates the step to the next MPI order.

# Modeled LIDT

- Significant LIDT growth ~680nm (the calculated transition point)

- Periodic behavior

- Resonant and nonresonant excitation of the electronic states and the quantized photon energy
- Applied photon energy differs from the band gap energy decreasing absorption
- Non-resonant condition gives more K.E. to Avalanche ionization effect

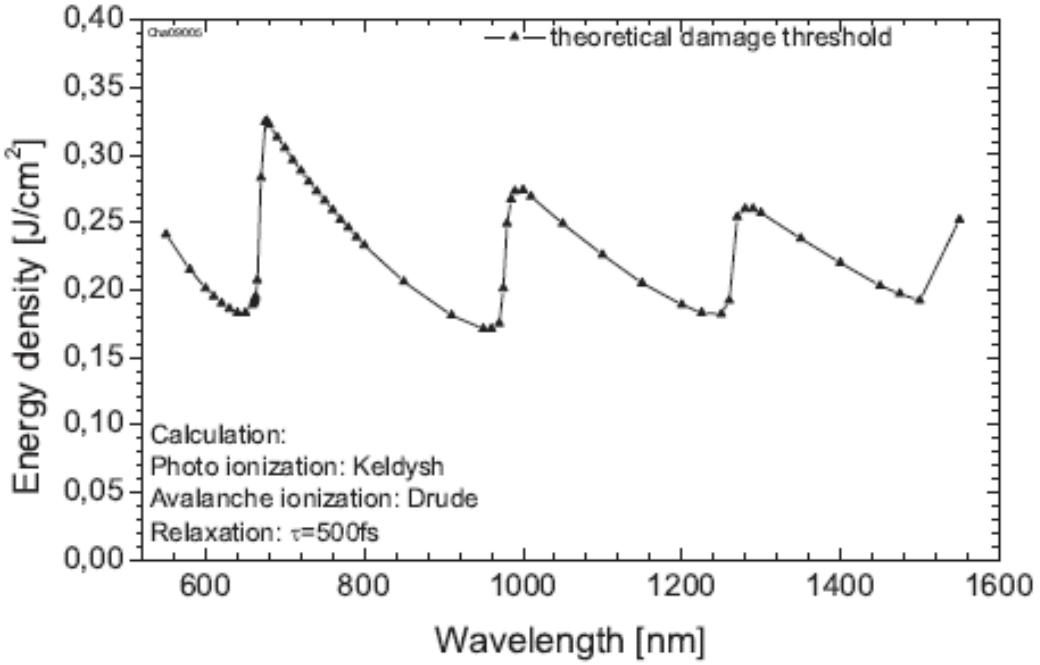



Fig. 5. Calculation of wavelength dependence on LIDT of $TiO_2$ from two up to the six-photon absorption

# Critical Electron Density

- Owing to the plasma frequency's dependence on the frequency of light impinging on the test surface the critical electron density will fall as the wavelength is increased

$$\rho_c = \frac{\varepsilon_0 m \omega_0^2}{e^2}$$

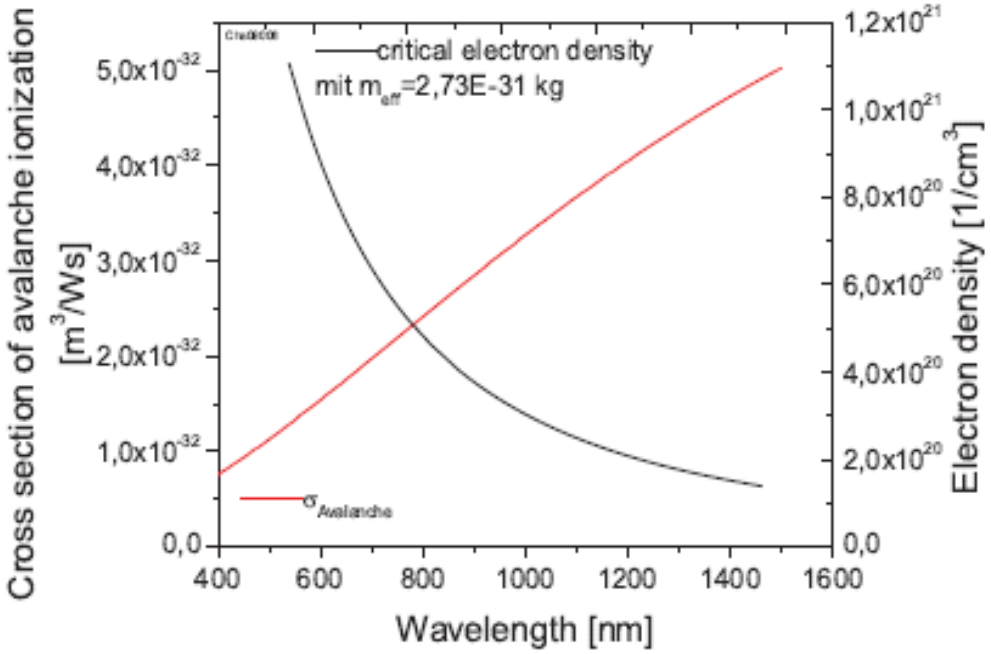
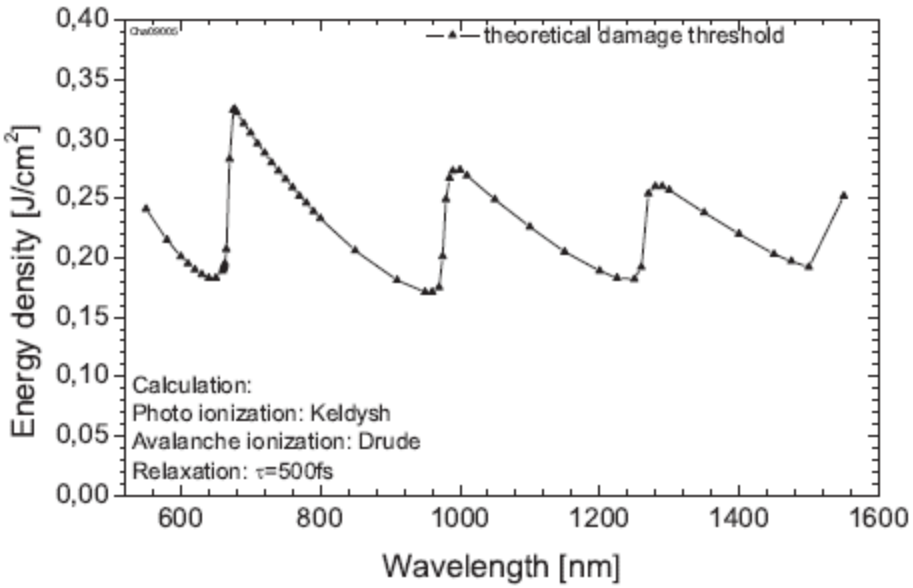
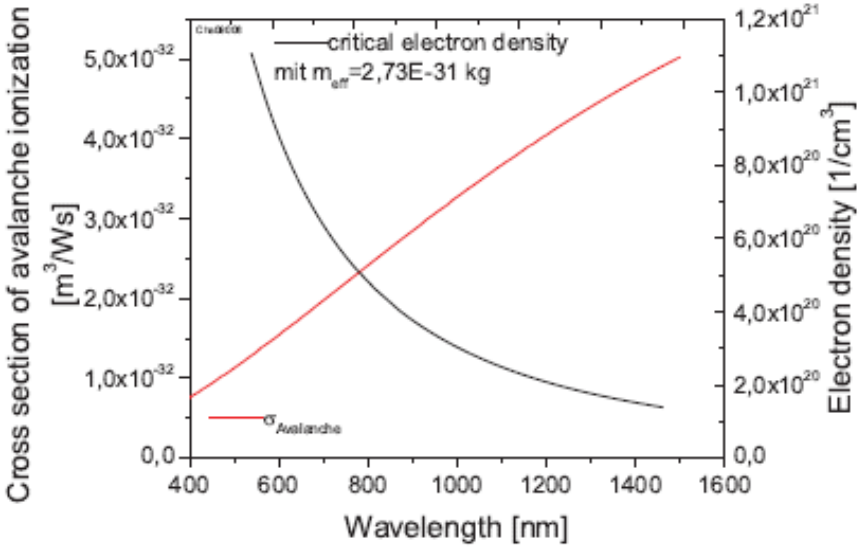



Fig. 6. Wavelength dependence of critical electron density and cross section of avalanche ionization rate

# Predicted LIDT Behavior

- **The paper notes that we might expect to see a significant increase in the laser damage threshold as they test across the wavelength range in the data set**
  - The critical electron density decreases as you go to higher wavelengths
  - The avalanche ionization cross section increases
- **Three separate processes are working in conjunction to keep the laser damage threshold nearly constant**
  - A decreasing critical electron density
  - Increasing avalanche ionization cross section
  - A decreasing photo-ionization rate



# Sample Prep

- TiO2 single layers deposited by ion beam sputtering
- Thickness was 440nm on B270 substrates
- Band gap was found from the second derivative of the transmission measurement.
  - Increases the ability to resolving narrow absorption lines.
- Titania layers have Eg = 3.61eV

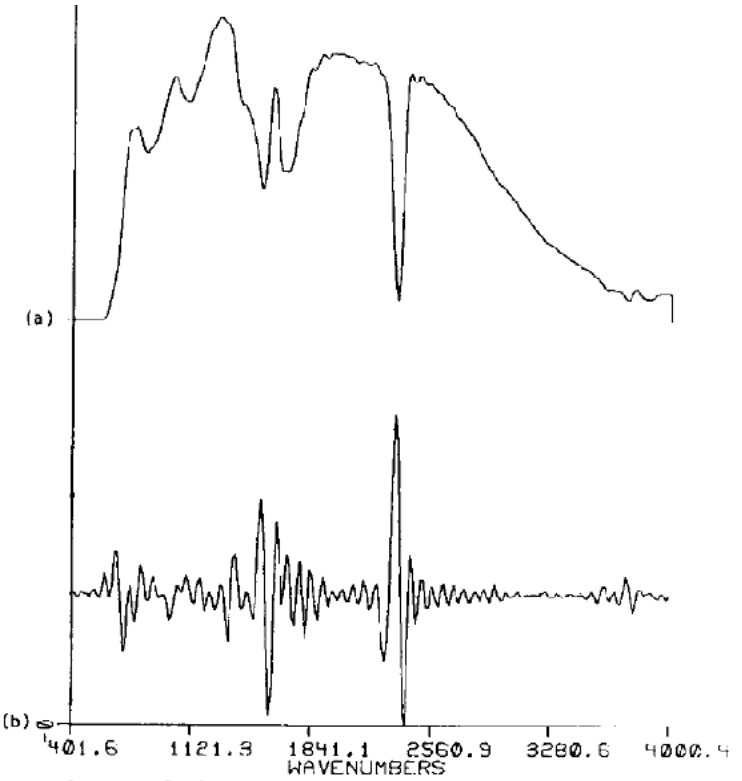
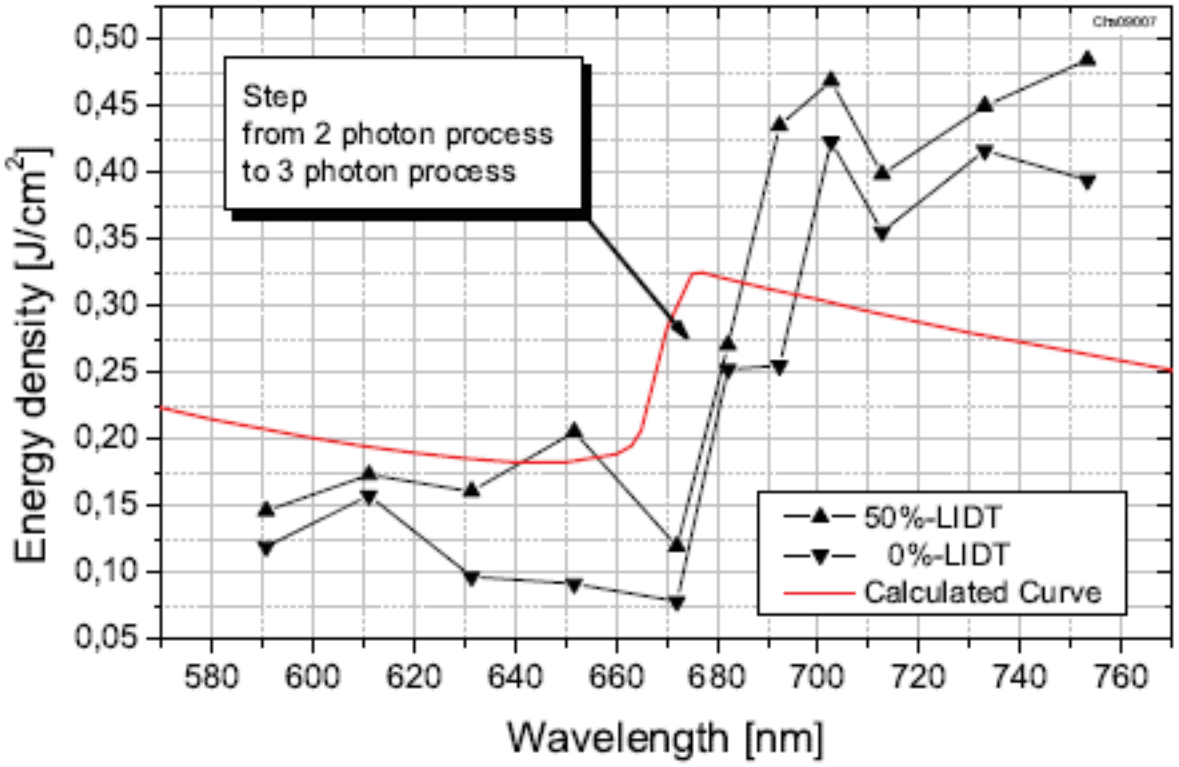



FIG. 1. a, low resolution spectrum of the atmosphere; b, second derivative of a.

# LIDT Testing

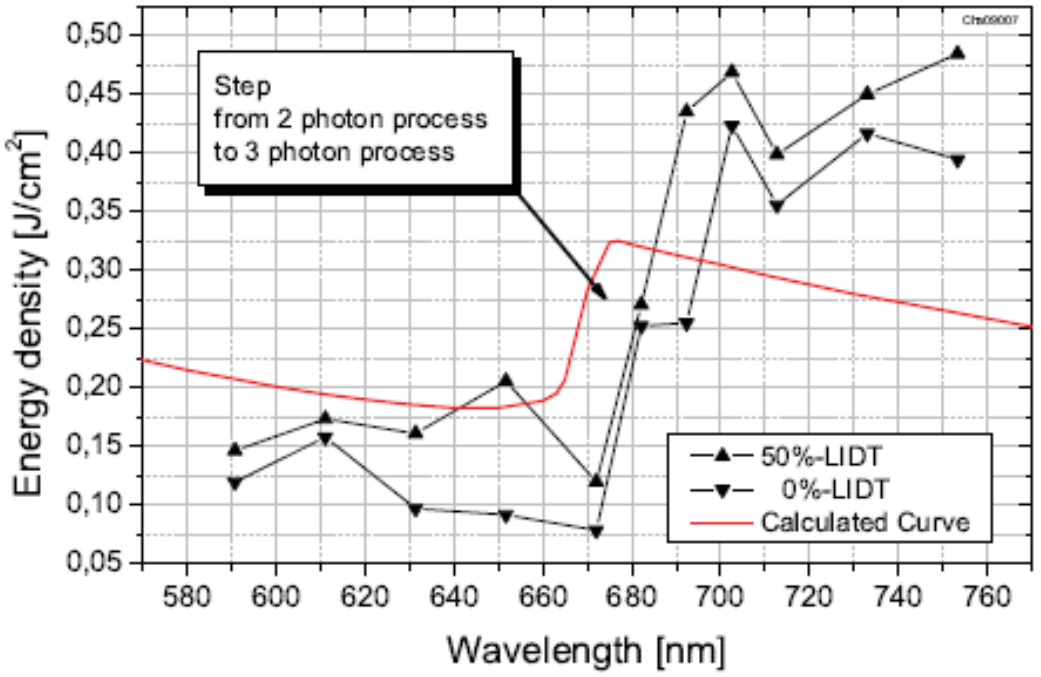
- Two LIDT measurements were made (OPA was used)
  - One set at the 0% LIDT of the film
  - Second set at the 50% LIDT measured both with 1000 pulses per test site

- LIDT values remain relatively constant in the range from 500nm to ~670nm
- A transition is made to higher LIDT at wavelengths much above 680nm
- This correlates quite well with the predicted value for the transition from two to three photon processes



# Calculated vs. Measured LIDT

- There is a noticeable difference in the predicted and measured LIDT enhancement when transitioning from a two to three photon process
- Discrepancy: overestimation of the amount of avalanche ionization



# Conclusions

- The N-on-1 tests seem out of order given the discussion throughout the paper. All of the modeling parameters were aimed at studying the preexisting material characteristics without the effects of incubation - no intermediate states.
- A quantized behavior was predicted and demonstrated for the LIDT when transitioning between a 2 and 3 photon process.
- The numerical model seemed to reasonably predict the materials behavior.
- The paper was pretty good.

# Summary

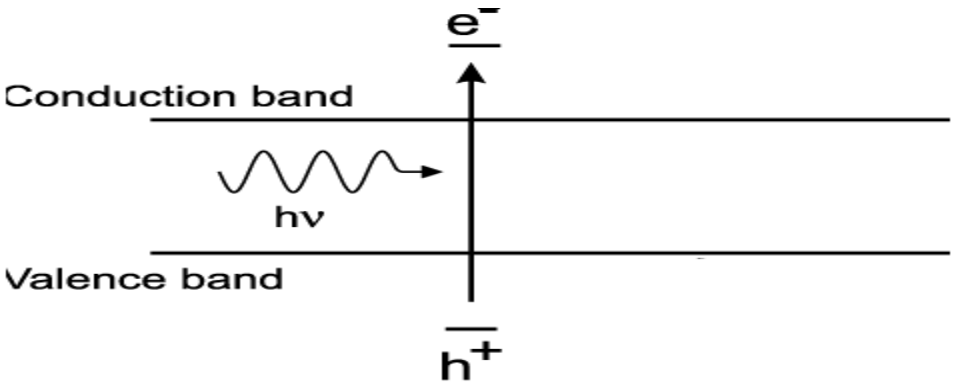
- First Paper:
  - Interesting and useful topic
  - Hard to follow (English was poor)
  - Not thorough in discussing their train of thought
  - Seemed to make general assumptions with little data
  - A few errors
- Second Paper:
  - Somewhat intuitive but good confirmation
  - Good discussion of what was happening throughout
  - Very questionable comparisons made towards the end
  - Some minor errors

# Back-Up Slide: (1)
# 1ω Contribution (Potential Error)

S1, S2, S3, S4

$R_{1\omega}$ = 0.7, 0.4, 0.5, 0.9

Contribution fraction first decreases then increases with higher 1ω contribution. λ = 354nm ~ 3.5eV, λ = 1064nm ~ 1.1eV




| Material | $E_g$ (eV) |
|---|---|
| SiO2 | 8.3 |
| Al2O3 | 6.5 |
| HfO2 | 5.1 |

- The contribution from 1ω in the LIDT of the coating is not linear with the energy increase
  - The more 1ω in the combined fluence the more important role it plays in the damage process
  - Could be explained by avalanche ionization

# Back-Up Slide: Heat Equation (1)

$$\frac{\partial T}{\partial t} = D_0 \Delta T$$

$$boundary\_Conditions$$

$$\alpha I(t) = -4\kappa \frac{\partial T}{\partial r} + \frac{4}{3}\rho a \frac{\partial T}{\partial t}$$

$$solution: \_T = T_0(1 - e^{-\frac{4D\tau}{a^2}})$$

$$T_0 = \frac{\alpha F a}{4\kappa\tau}$$

$$D = \frac{3\kappa}{4\rho C}$$

$D_0$ thermal diffusivity

$\kappa$ thermal conductivity

$\rho$ density

C heat capacity

a defect radius

F laser fluence

$\alpha$ absorptivity

M. D. Feit and A. M. Rubenchik, "Inplications of nanoabsorber initiators for damage probability curves, pulselength scaling and laser conditioning," SPIE Vol. 5273 (2003)

# Back-Up Slide: Gaussian Defect Distribution (1)

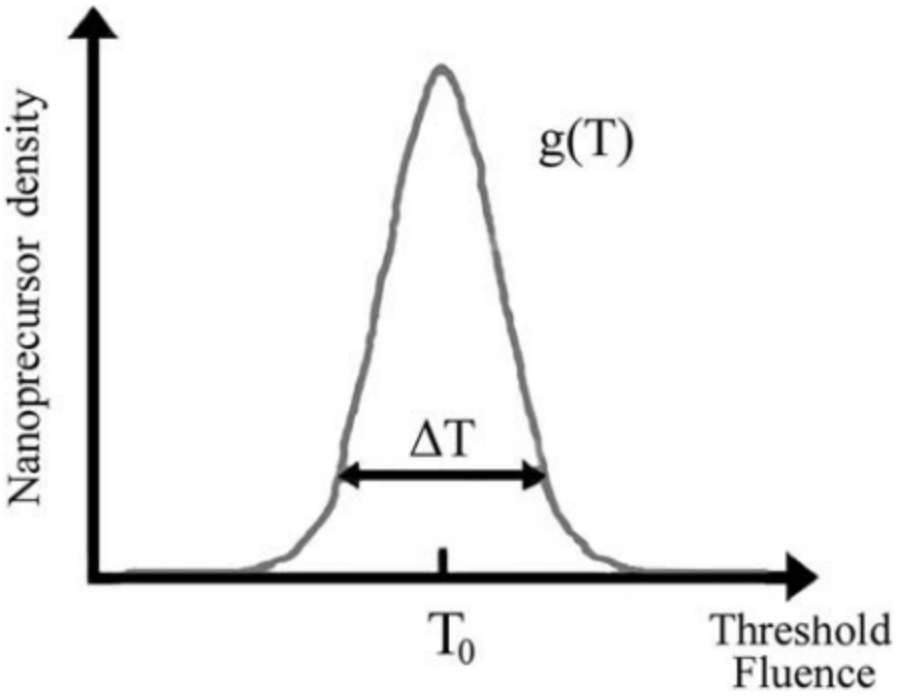



Fig. 1. Gaussian defect distribution.

H. Krol, et al., "Investigation of nanoprecursors threshold distribution in laser-damage testing," Optics Communications, 23 June 2005

# Back-Up Slide: Potential Experiments (2)

Potential Experiments

- A decrease in the ionization rate should be mirrored by the LIDT performance of the material when testing above and below the n and n+1 photon condition.
- Two potential experiments to test this theory:

  1) LIDT tests can be made on a material with an engineered band gap. This can be accomplished for a transition from a two photon process at 800nm to a three photon process by creating a ternary compound composed of TiO2 and SiO2.

  2) The output of a laser can be tuned to vary the photon energy across the transition point for a well defined band structure. (OPA)