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Winter College on Optics and Photonics
7 - 25 February 2000

1218-26

"Measurements of Nonlinearities in Silicon Nanoclusters & Telecommunication Fibers" - I

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Measurement of Nonlinearities in Silicon Nanoclusters and Telecommunication Fibers I

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Nonlinear Optics

$P = \chi^{(1)} \bullet E$ {linear optics; $n = (1 + 4\pi \chi^{(1)})^{1/2} = \varepsilon^{1/2}$}

$+ \chi^{(2)} \bullet EE$ {second harmonic generation,
sum, difference frequency generation,
Pockels effect, ⋯}

$+ \chi^{(3)} \bullet EEE$ {third harmonic generation,
nonlinear index of refraction,
Raman scattering,
two photon absorption, ⋯}

$+ \cdots$

Optical Nonlinearities

- Optical nonlinearities limit the amount of power transmitted through a fiber and can cause noise and crosstalk in lightwave systems. A long-haul transmission system (10,000-km) would require 250 erbium-doped fiber amplifiers (EDFAs) for a typical repeater spacing of 40-km. EDFAs are typically 15-25 meters in length. A technique to measure n_2 ($n = n_0 + n_2 I$) in ***short lengths (< 25-m) of fiber*** would be extremely useful in predicting system performance.
- Silicon nanoclusters, with enhanced third order nonlinearities ($\chi^{(3)}$ (esu)) may prove to be a new and novel nonlinear element for all-optical switching. Due to the extremely small nonlinearity of silica fiber ($\chi^{(3)}$ (esu) $\approx 9 \times 10^{-14}$), a fiber nonlinear optical loop mirror (NOLM) has to be on the order of 10-km long to achieve the necessary π-phase shift for high-speed all optical switching. Nonlinearities as large as ($\chi^{(3)}$ (esu) $\approx 10^{-4}$) have been measured in silicon nanoclusters.

Nonlinear Optical Properties of Silicon Nanoclusters Made by Laser Ablation And Ion Implantation

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Introduction

- Previous experiments with ns pulses indicated large nonlinearity in laser ablated Si nanoclusters (NC) $\chi \sim 10^{-13}$ esu silica fiber
  (S.Vijayalakshmi, and H. Grebel)
  $\lambda = 532\text{nm}: \chi_{Re}^{(3)} \sim 10^{-4}$esu; $\lambda = 355$ nm: $\chi_{Re}^{(3)} \sim 10^{-5}$esu
- Current investigations of ultrashort pulses (λ = 375nm, 532 nm, 790 nm) are consistent with ns results.
- Large nonlinearity may be due to a quantum size effect arising from close-packed nanocrystallites of silicon within micron-sized droplets--a "cooperative" phenomena
- Si ion implantation → uniform size distribution
  → nonlinear response dominated by nonlinear absorption ($\chi^{(3)}_{Im}$)
- Potential as an optical switch for Si based integrated optoelectronic systems.

Laser Ablated Si NC

- Pulses from a KrF excimer laser (λ=248 nm, τ_p =8 ns, P=3W) are focused unto a Si wafer (<100>, n-type, 10^{16} cm^{-3})
- Clusters from silicon are deposited unto a quartz substrate 3 cm away.
- Resultant film made of micron-size droplets (2-3 μm)
- Within the droplets, close-packed Si NC with a size distribution: 3-50 nm
- Due to directional nature of plume, samples possessed three regions with varying film thickness: 100nm, 200nm and 400nm

Ion Implanted Si NC

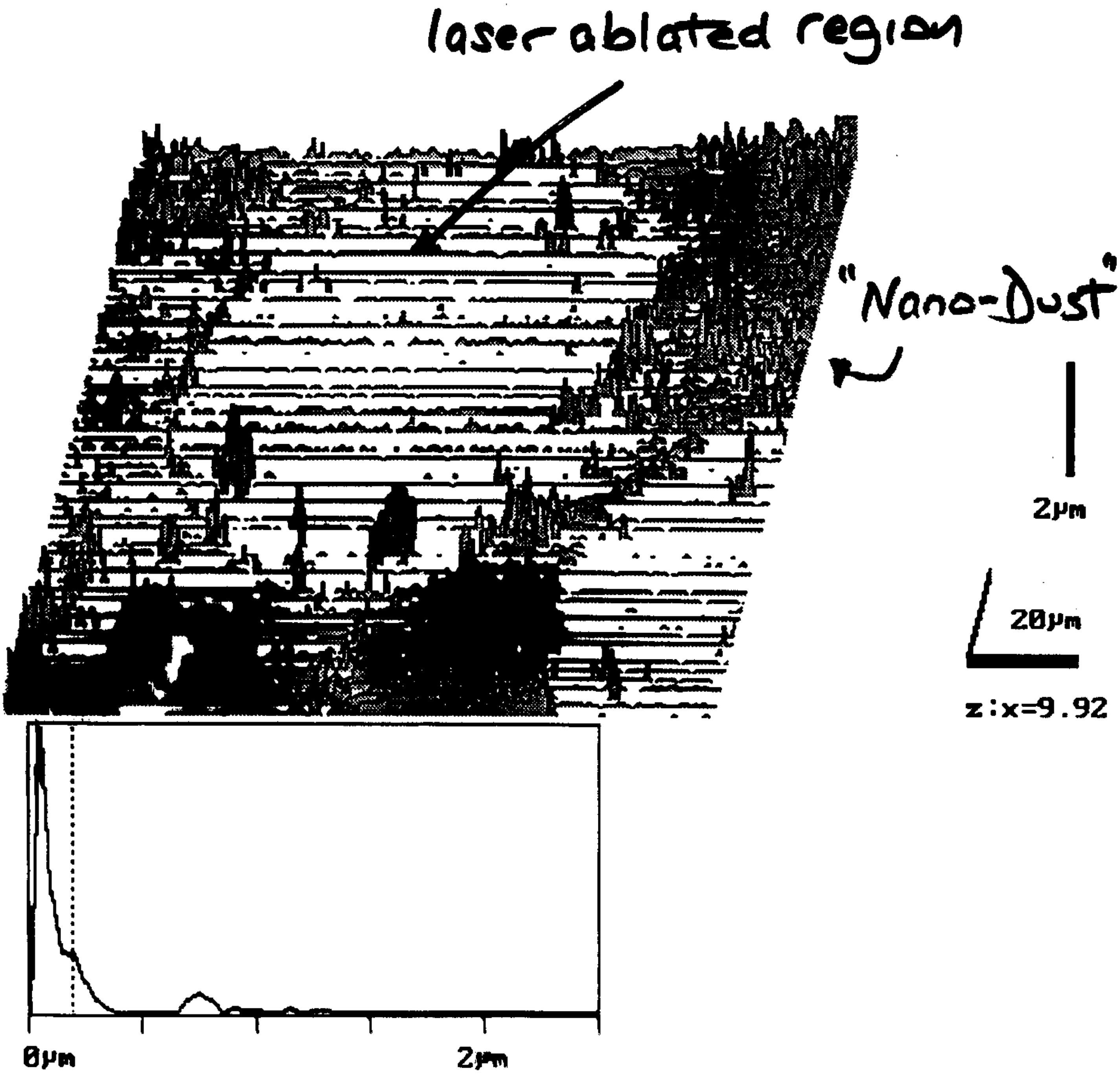
- Si NC formed by Si (400keV) implantated into fused silica followed by annealing (1100° C) in flowing Ar + 4% H_2
- Implantation dose of $6x10^{17}$ cm^{-2} with a peak excess Si concentration of $2x10^{22}$ cm^{-3}
- Cluster mean diameter: 5-6 nm
- Film thickness: 300 nm

Ion implanted Si sample provided by

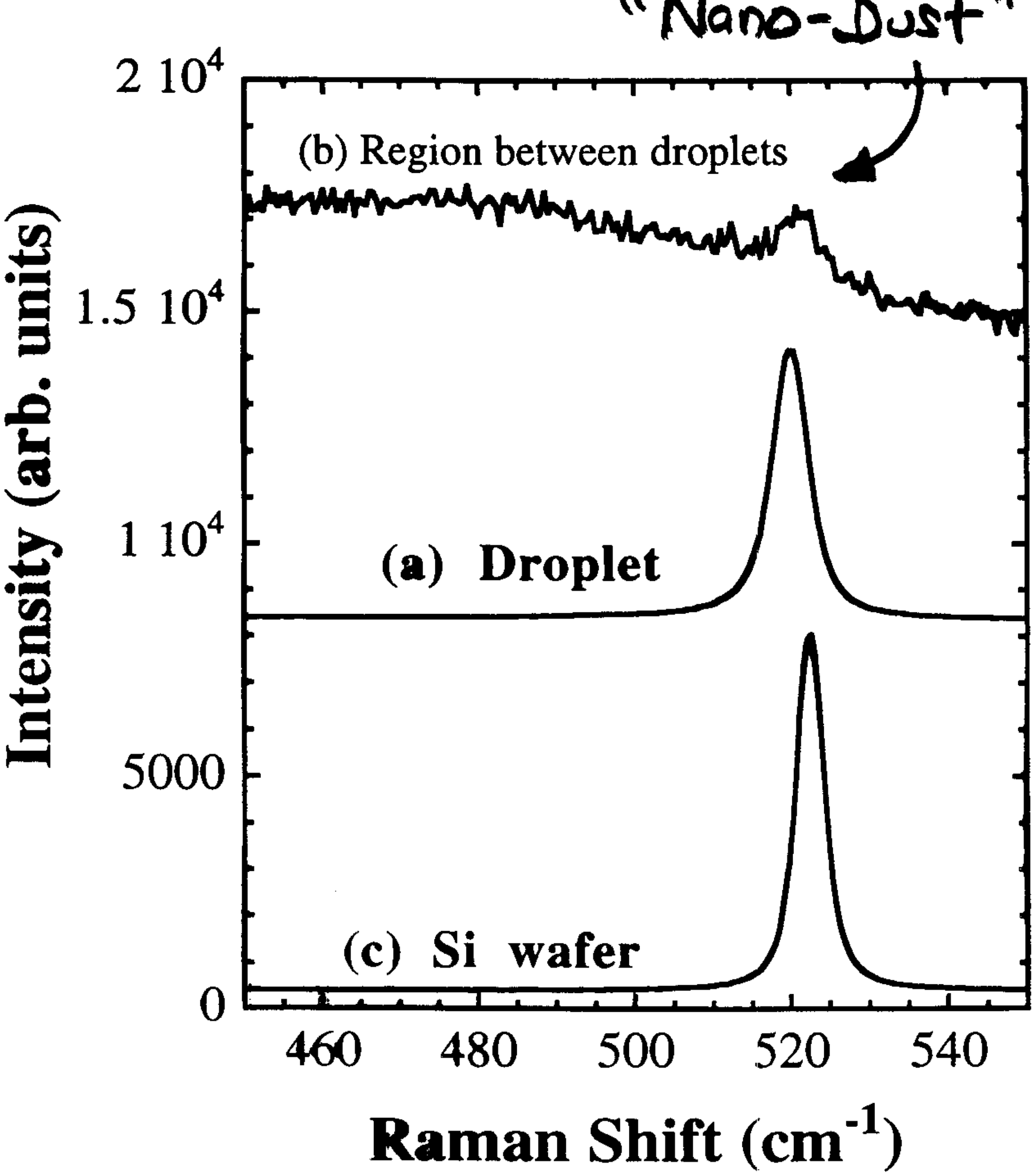
C.W. White

Solid State Division, Oak Ridge National Laboratory

Laser Scanning Microscopy



Micro-Raman Spectra - Laser Ablated



Nonlinear Optical Materials

$$n = n_0 + n_2 I(r,t) = n_0 + \Delta n(r,t)$$

$$\alpha = \alpha_0 + \beta I(r,t) = \alpha_0 + \Delta\alpha(r,t)$$

$$n_2 \sim \chi^{(3)}_{Re} \qquad \beta \sim \chi^{(3)}_{Im}$$

$$\Delta\phi(t) = \frac{2\pi}{\lambda}\Delta n(t)\, L_{eff} \;;\; L_{eff} = \frac{1-e^{-\alpha L}}{\alpha}$$

$$I(r) = I_0 e^{-2r^2/r_0^2}$$

Gaussian spatial profile

Rayleigh range $z_0 = \frac{\pi r_0^2}{\lambda} = \frac{\pi \omega_0^2}{\lambda}$

$\omega_0 \equiv$ beam waist

$$f^{-1} = L \left.\frac{\partial^2 \Delta n}{\partial r^2}\right|_{r=0}$$

$$f = \frac{r_0^2 c\, \epsilon_0 n_0}{n_2 I_0 L}$$

Nonlinear lens of focal length f

The thin nonlinear material ($L << z_0$) acts similar to thin concave ($n_2 > 0$) and convex ($n_2 < 0$) lenses for paraxial rays.

Z-SCAN

(Mansoor Sheik-Bahae, Eric Van Stryland et al., 1990)

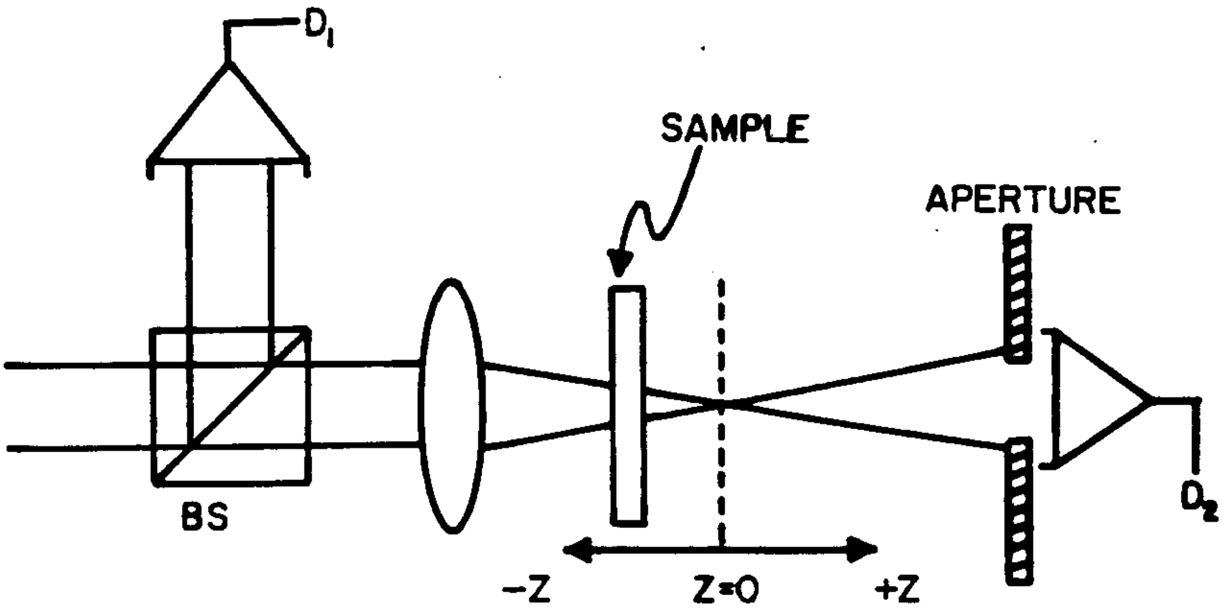


FIGURE 13.8 Experimental setup for the z-scan measurement. The transmission through the thin sample and aperture is determined by the ratio of the energy measured by detector D_2 to that measured by detector D_1, $T = D_2/D_1$.

Z-SCAN

A technique that uses a thin material ($L<<Z_o$) to determine the sign and magnitude of the index change ($\Delta n=n_2 I$) and the magnitude of the nonlinear absorption ($\Delta\alpha=\beta I$). A small aperture is placed on-axis in front of a detector in the far field. For an incident laser beam, the normalized transmittance is measured at different positions along the z direction while the sample is translated symmetrically through the focused beam waist. If $n_2>0$ and the sample is positioned before the focus, the nonlinear medium will focus the beam earlier and to a smaller waist (dotted line Prefocal). Since the waist is made smaller the beam expands more rapidly owing to diffraction; remains collimated over a shorter distance in the near field; and diverges at a larger beam angle in the far field, which reduces the irradiance at detector D_2. When the sample passes into the postfocal position the positive self-lensing of the nonlinear material tends to reduce the beam divergence (dotted line Postfocal), which results in increased irradiance at detector D_2. If $n_2<0$ and the sample is placed in the prefocal region, the beam waist at the focus will be increased and the focus will be closer to the aperture (dashed line Prefocal). As a result, more radiation will pass through the aperture, producing an increased signal on the detector. When the material passes into the postfocal region the negative lensing effect of the material will spread the already diverging rays even more (dashed line Postfocal) so that the irradiance will be significantly decreased at the detector.

Z-SCAN

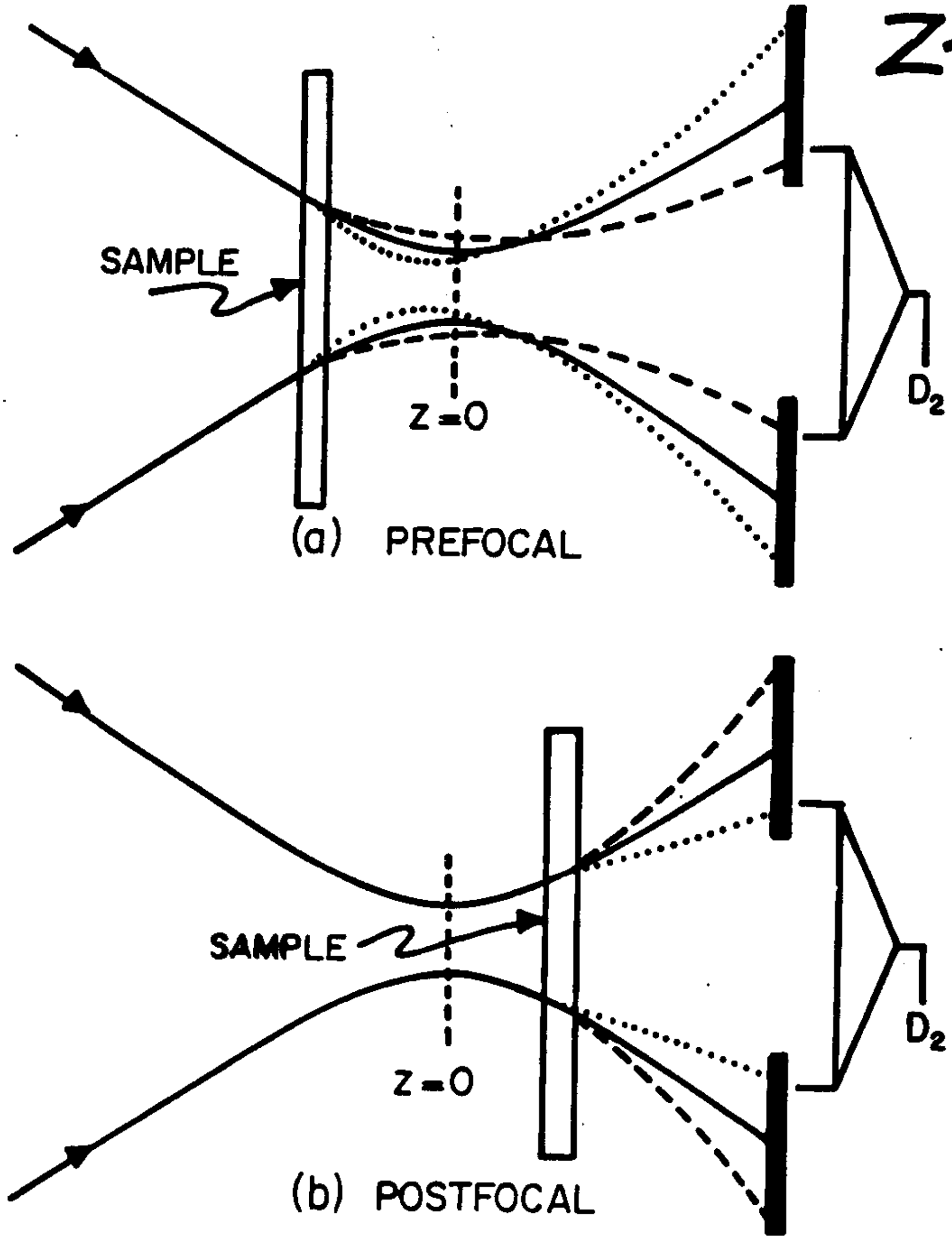


FIGURE 13.9 Nonlinear refraction: (*a*) prefocal ($z < 0$) diffraction, (*b*) postfocal ($z > 0$) diffraction. The solid line in both figures is linear (low-intensity) diffraction. The dotted line shows $n_2 > 0$ and the dashed line corresponds to $n_2 < 0$.

• • • • $n_2 > 0$

– – – – $n_2 < 0$

Z-SCAN (clased)

Time averaged opertured on-axis transmission

$$T = 1 + \frac{4\Delta\phi x}{(x^2+1)(x^2+9)}$$

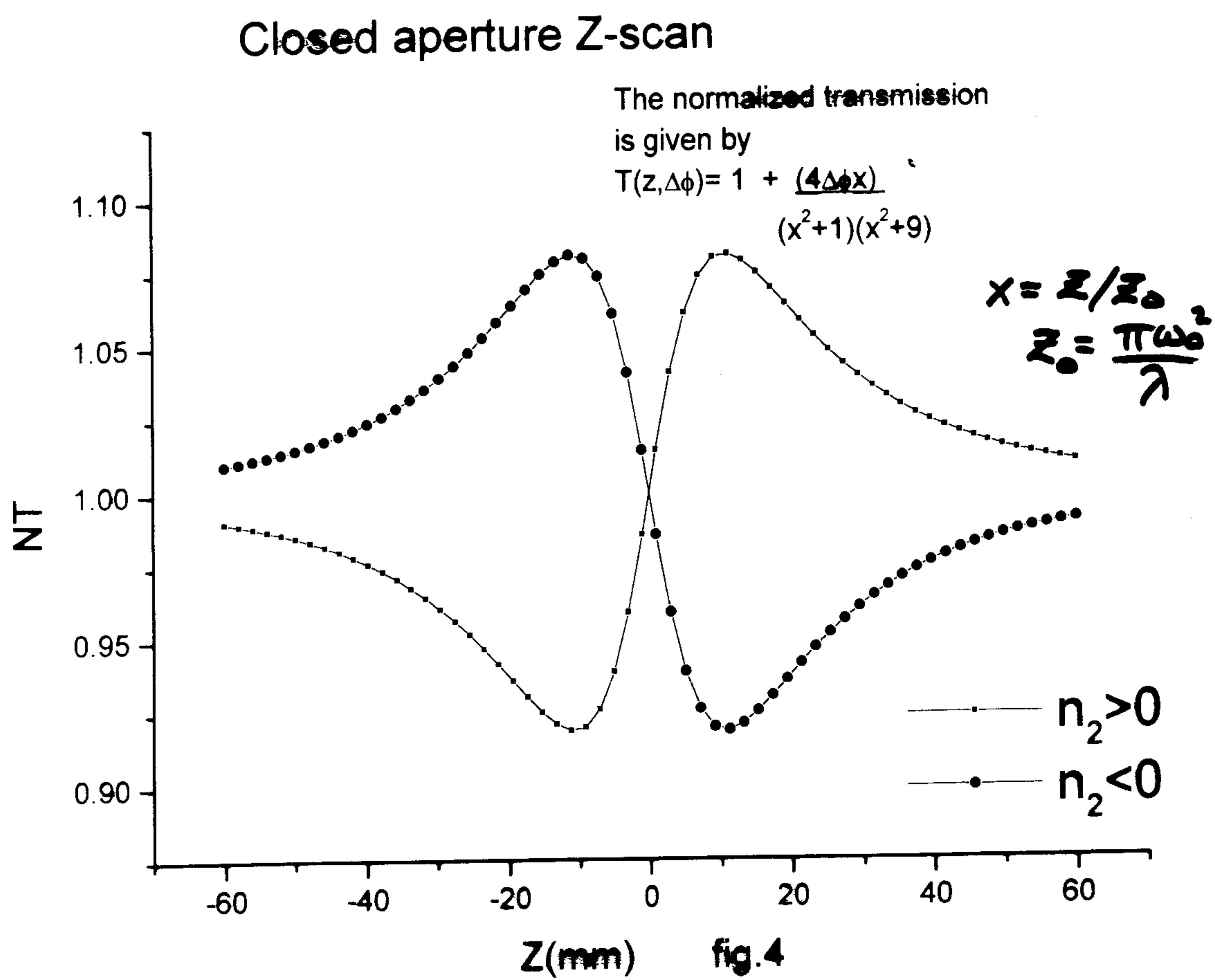
$$x = z/z_0 \; ; \; z_0 = \frac{\pi w_0^2}{\lambda}$$

$$n(I) = n_0 + \gamma I = n_0 + n_2 I$$

$$\gamma = \frac{\Delta\phi \lambda}{2\pi I_0 L_{eff}}$$

$$\chi^{(3)}_{Re} = 2 n_0^3 \epsilon_0 c \gamma$$

$$n_2(esu) = \frac{c n_0}{40\pi} \gamma \left(\frac{cm^2}{W}\right)$$



Closed aperture Z-scan
The normalized transmission
is given by
T(z,Δϕ)= 1 + (4Δϕx) / (x²+1)(x²+9)
x = Z/Z₀
Z₀ = πω₀²/λ
NT
1.10
1.05
1.00
0.95
0.90
-60
-40
-20
0
20
40
60
n₂>0
n₂<0
Z(mm)

fig.4

Z-SCAN (open)

Open apertured transmission
- sensitive to nonlinear absorption only

$$T = 1 - \frac{\beta I_0 L_{eff}}{2\sqrt{2}(1+x^2)}$$

$$x = z/z_0 \; ; \; z_0 = \frac{\pi \omega_0^2}{\lambda}$$

$$\alpha = \alpha_0 + \beta I$$

$$\chi^{(3)}_{Im} = \frac{n_0^2 \epsilon_0 c^2}{\omega} \beta$$

$$\omega = 2\pi\nu$$

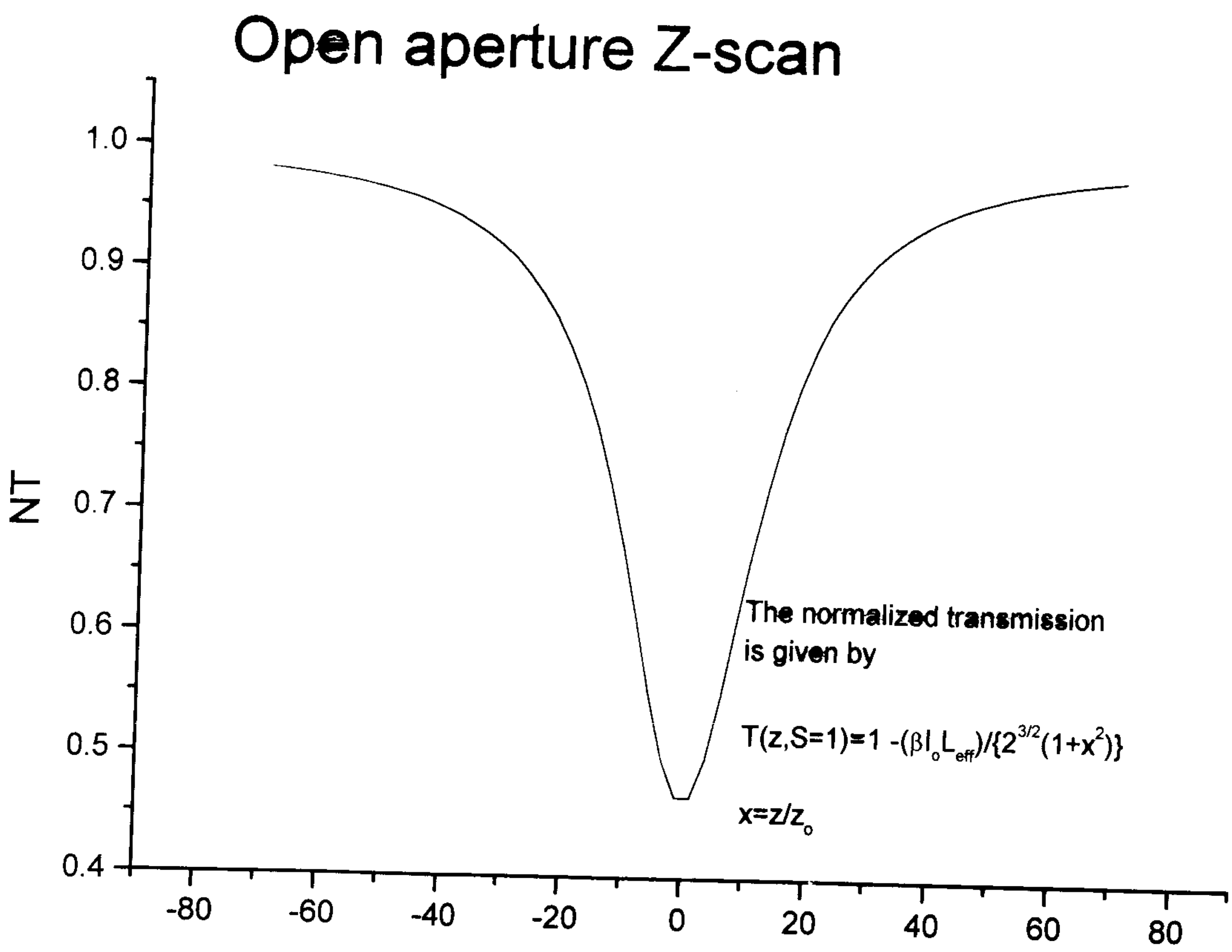
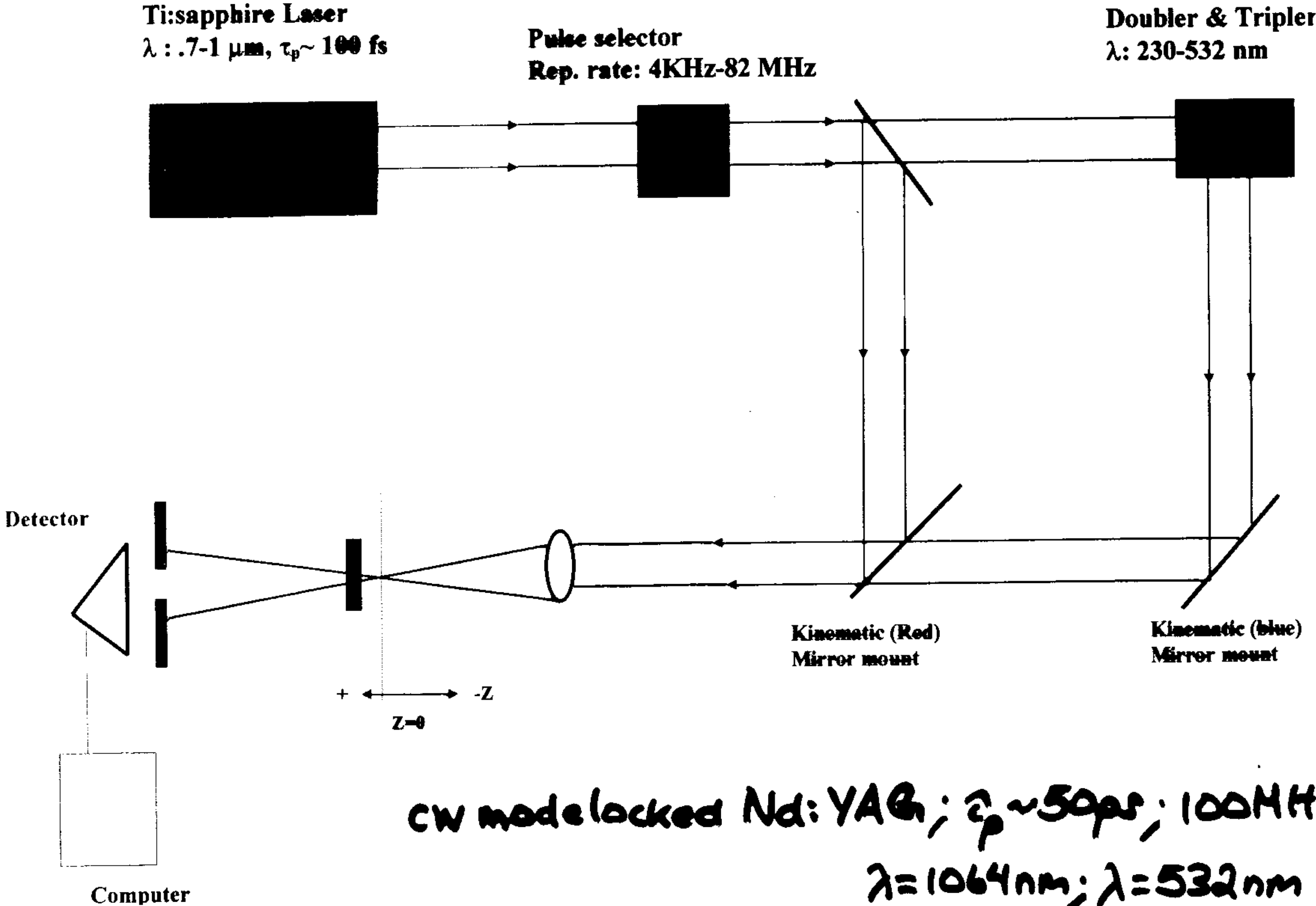

Open aperture Z-scan
NT
1.0
0.9
0.8
0.7
0.6
0.5
0.4
The normalized transmission
is given by
$T(z,S=1)=1-(\beta I_o L_{eff})/\{2^{3/2}(1+x^2)\}$
$x=z/z_o$
-80
-60
-40
-20
0
20
40
60
80
Z(mm)

fig.2

Z-SCAN

(M. Sheik-bahae et al., 1990)

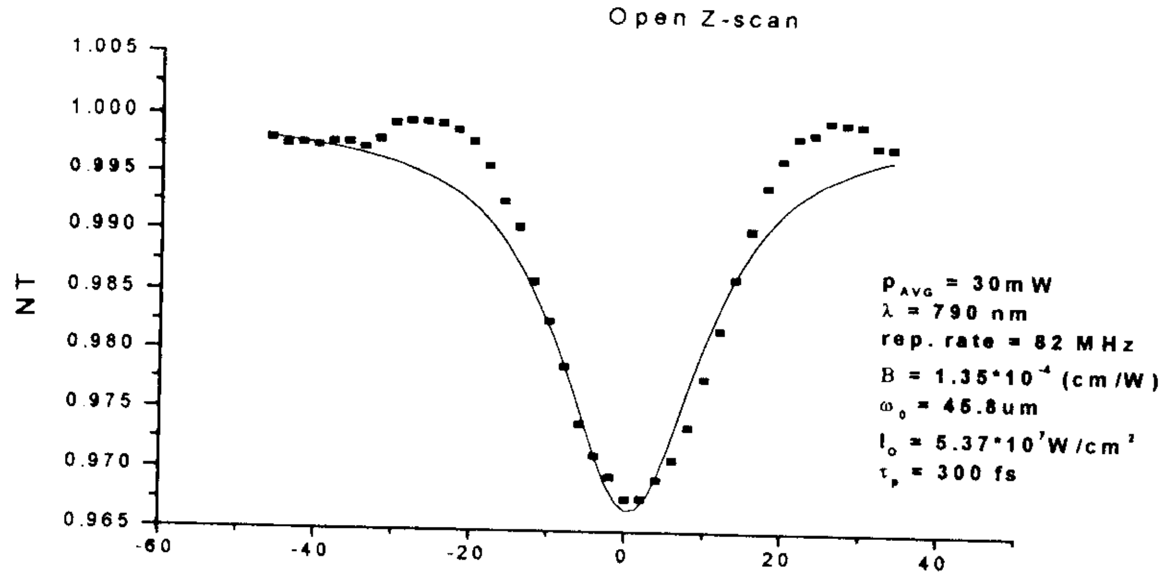
- Sample is scanned about the focal length of a lens and transmission detected as a function of distance from the focal point.
- Sample acts as a thin lens and either self-focuses or defocuses the beam
- Nonlinear refraction estimated by measuring farfield output with an apertured detector and nonlinear absorption without the aperture.
- In the presence of nonlinear absorption: normalized closed aperture is divided by open aperture to retrieve n_2.

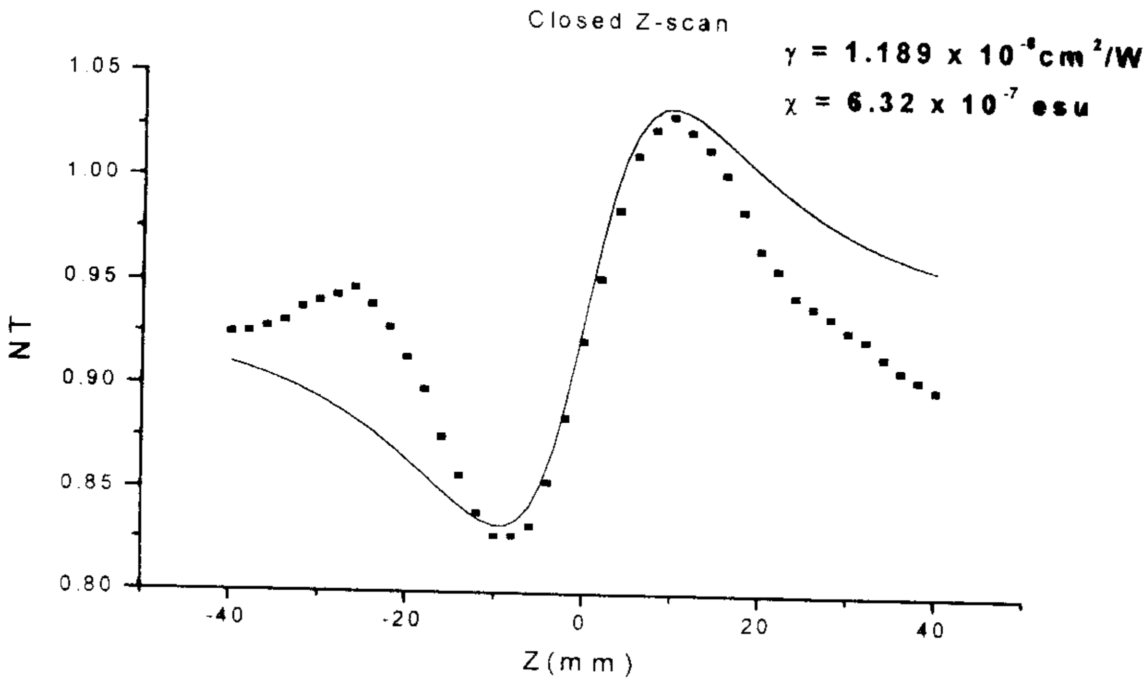


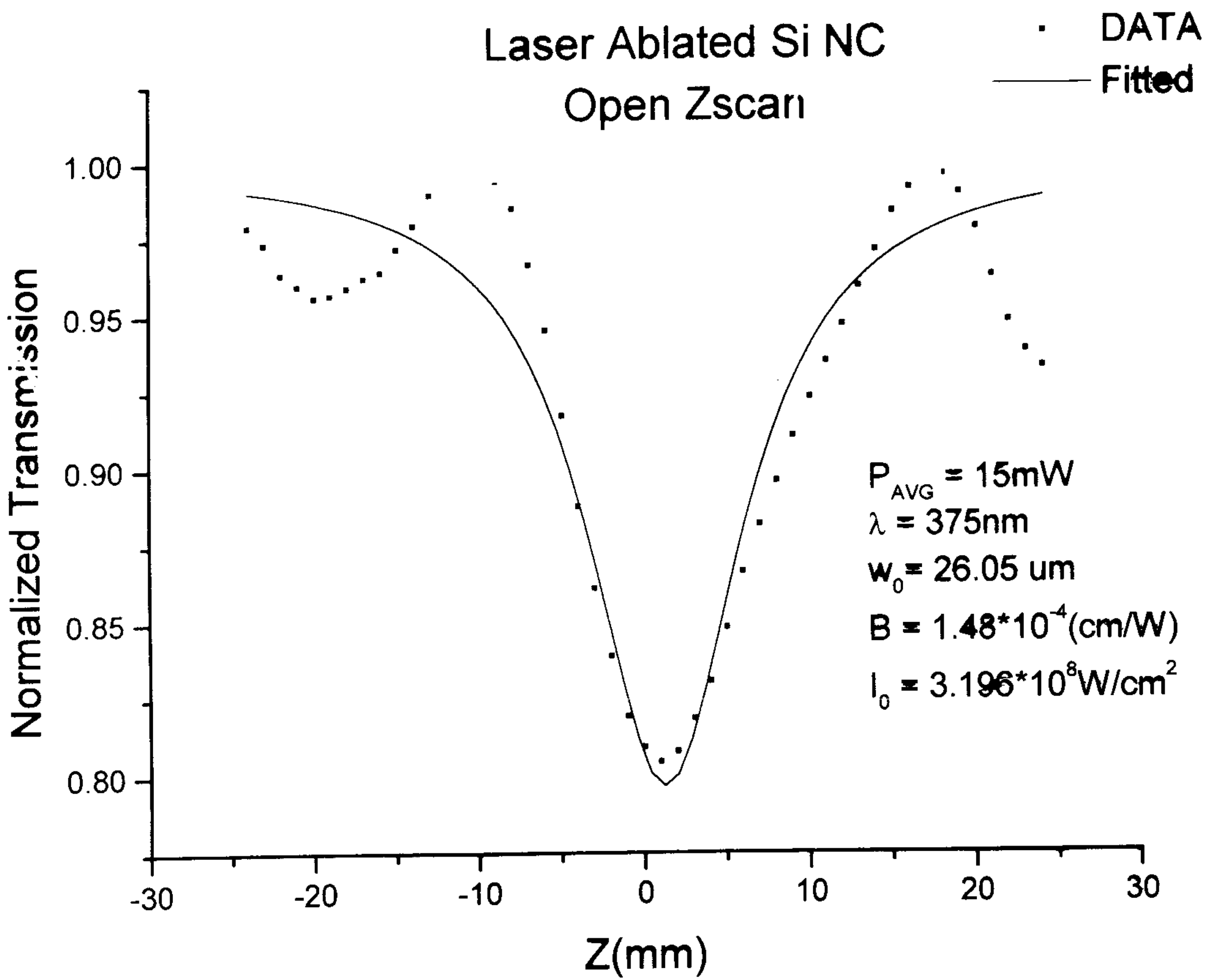
CW modelocked Nd:YAG; τ_p~50ps; 100MHz
λ=1064nm; λ=532nm

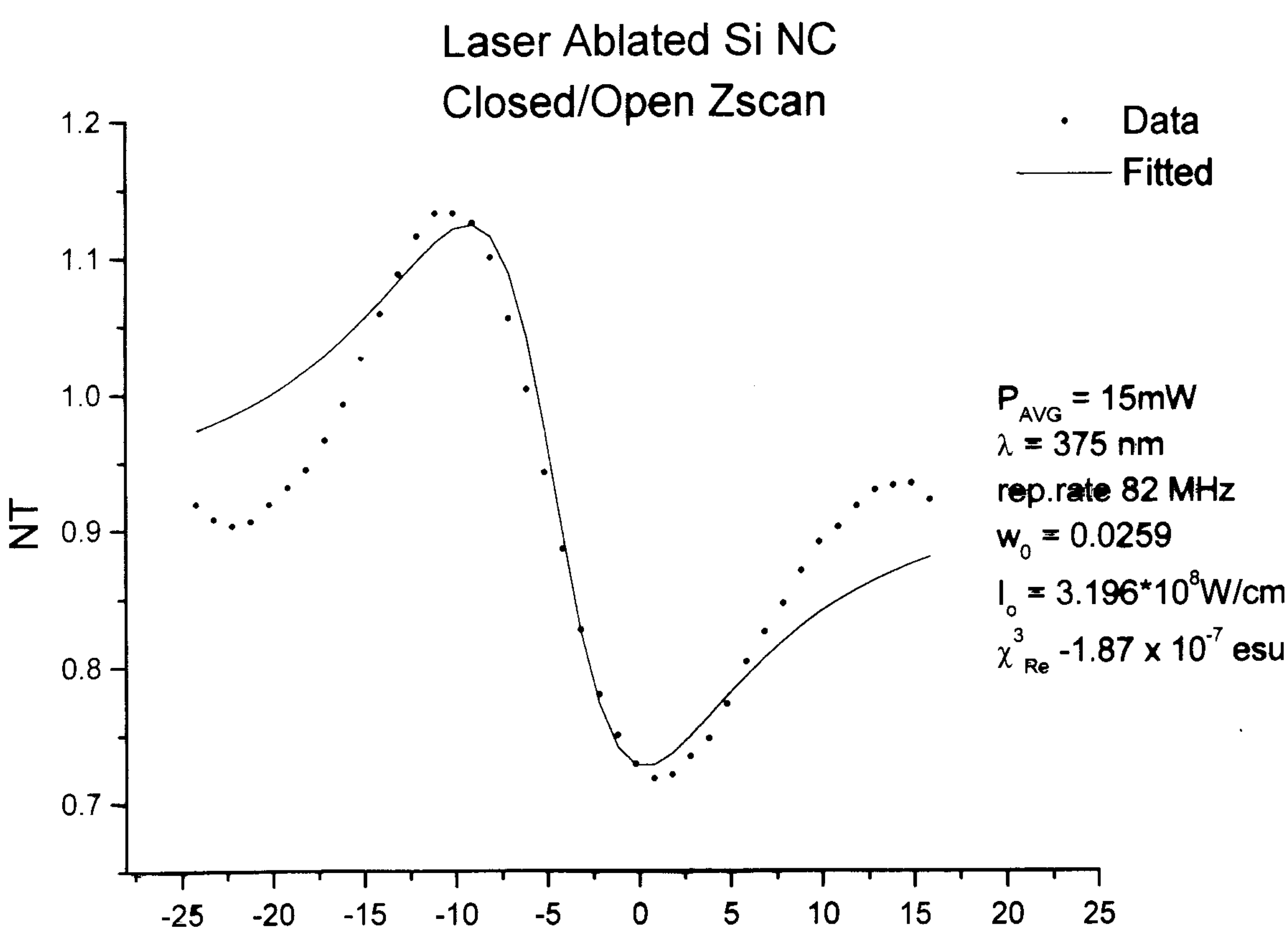
Experimental setup for the Z-scan measurement *(fig.1)*

Laser Ablated Si NC






Laser Ablated Si NC
Open Zscan
DATA
Fitted
1.00
0.95
0.90
0.85
0.80
Normalized Transmission
P_AVG = 15mW
λ = 375nm
w_0 = 26.05 um
B = 1.48*10^-4(cm/W)
I_0 = 3.196*10^8W/cm^2
-30
-20
-10
0
10
20
30
Z(mm)


Laser Ablated Si NC
Closed/Open Zscan
Data
Fitted
NT
1.2
1.1
1.0
0.9
0.8
0.7
-25
-20
-15
-10
-5
0
5
10
15
20
25
P_{AVG} = 15mW
λ = 375 nm
rep.rate 82 MHz
w_0 = 0.0259
I_o = 3.196*10^8W/cm
χ^3_{Re} -1.87 x 10^{-7} esu

Z(mm) fig.4

Z-scan measurements of Si nanoclusters prepared by laser ablation

Table 1

cw-modelocked Ti-Sapphire laser

λ(nm)	Rep. rate	τ_p	β(cm/W)	$\chi^{(3)}$(esu)	I_0 (W/cm^2)
790	82 MHz	250 fs	1.35×10^{-4}	6.32×10^{-7}	5.37×10^{7}
375	82 MHz	100 fs	1.48×10^{-5}	-1.87×10^{-7}	3.2×10^{8}
375	82 MHz	100 fs	6.07×10^{-4}	-7.64×10^{-7}	1.062×10^{8}

cw-modelocked and frequency doubled Nd:YAG laser

λ(nm)	Rep. rate	τ_p	$\chi^{(3)}$(esu)	I_0(W/cm^2)
532 nm	100 MHz	50 ps	6×10^{-4}	5.1×10^{4}
532 nm	1 MHz	50 ps	4×10^{-4}	1.9×10^{5}

Second & third harmonics of a pulsed Q-switched Nd:YAG Laser

(Vijayalakshmi and Grebel)

λ(nm)	Rep. rate	τ_p	$\chi^{(3)}$(esu)	I_0(W/cm^2)	τ^*
532	10 Hz	8 ns	10^{-3}	3×10^{4}	3.5± .5 ns
355	10 Hz	8 ns	2.28×10^{-5}	5×10^{5}	143± 20 ns

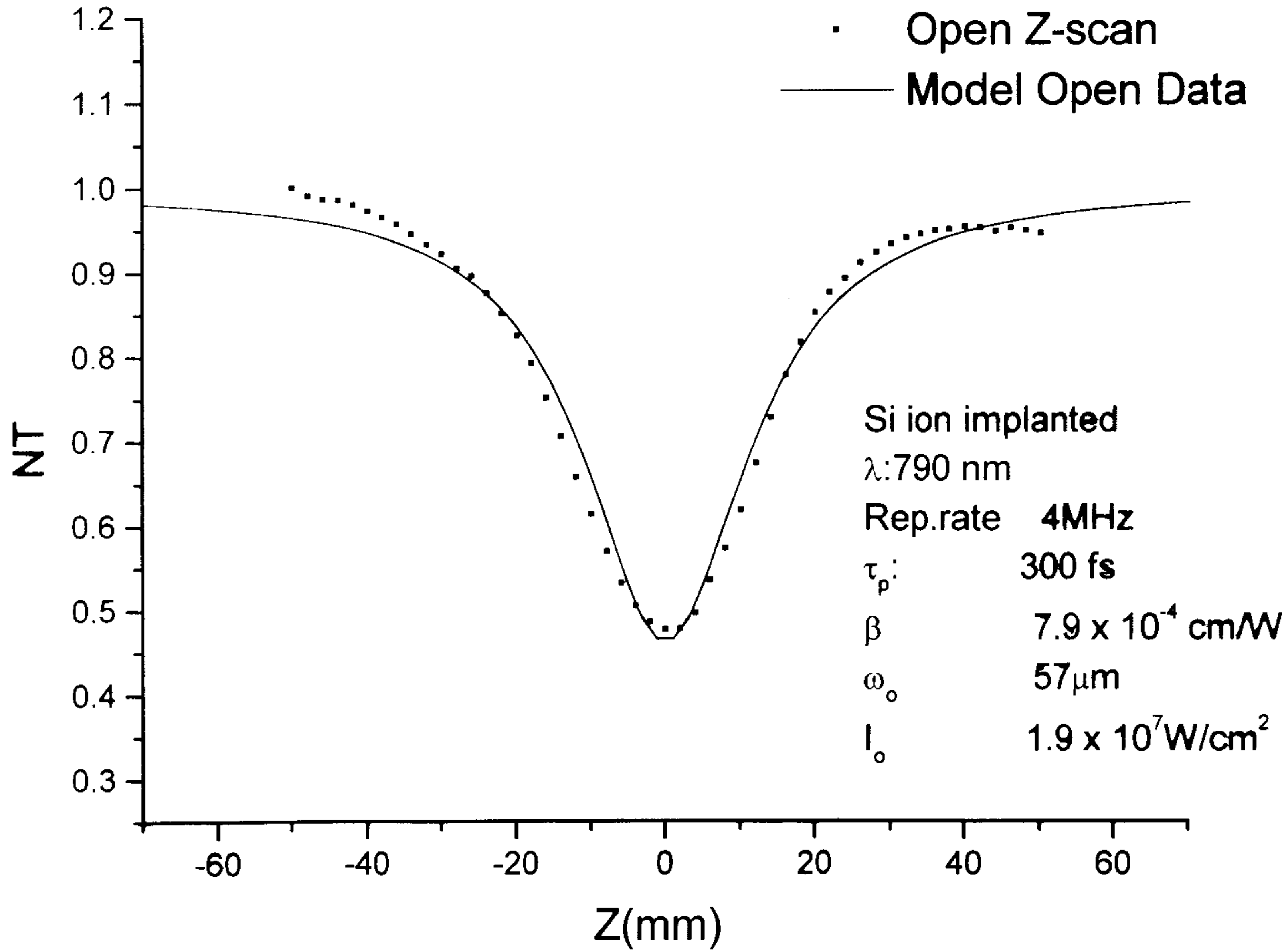
*Limited by laser pulse width

Porous Silicon : $\chi_{Re}^{(3)} \approx 10^{-9}$ esu (Henari et. al, 1995)

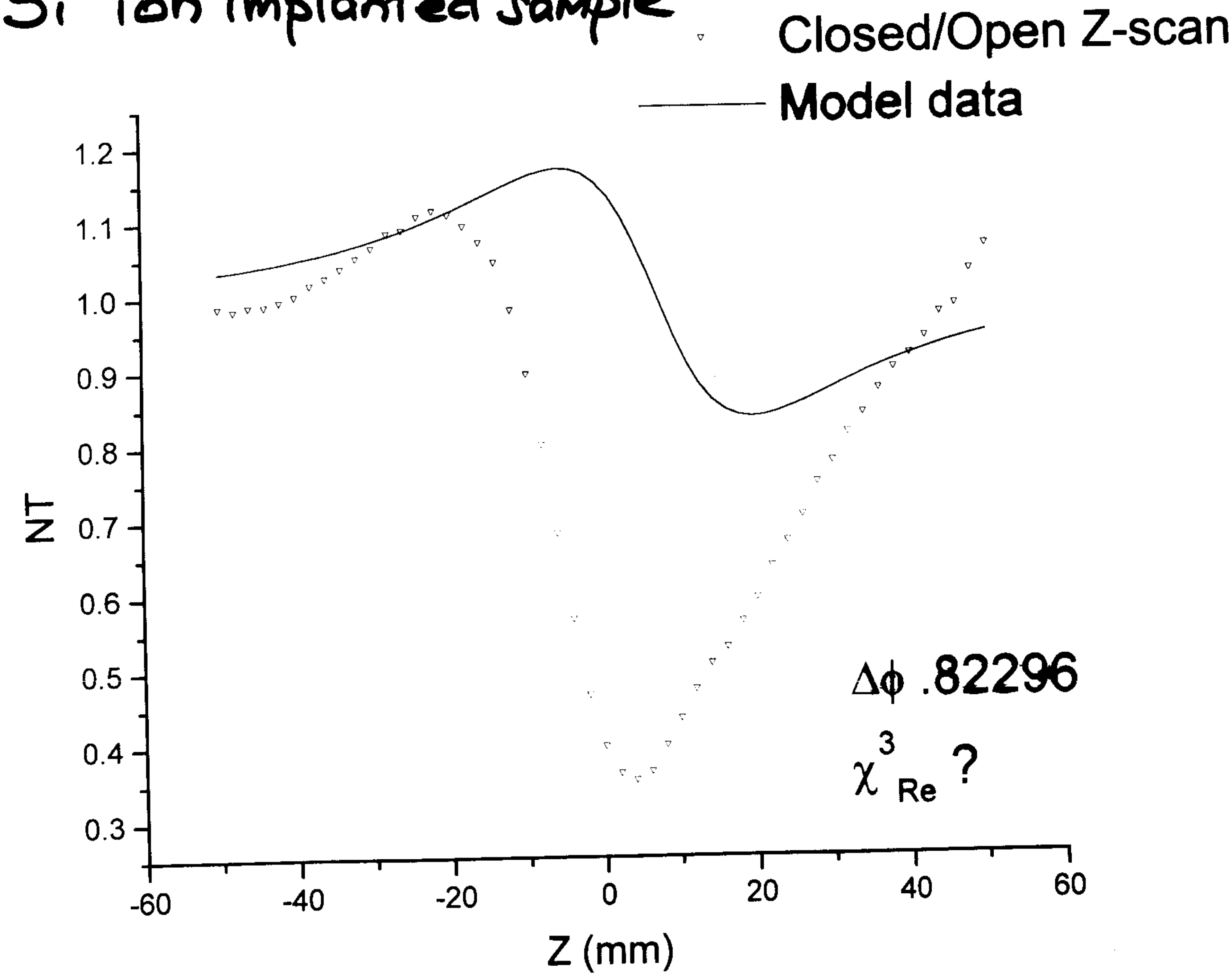
Silica Fiber: $\chi_{Re}^{(3)} \approx 9 \times 10^{-14}$ esu

Lalanne, Johnson et al

Si ion implanted sample



Si ion implanted sample



Micro-Raman Spectra

S1: 1.5×10^{17} cm^{-2}; Si-NC ~ 3-4 nm

S2: 6×10^{17} cm^{-2}; Si-NC ~ 5-6 nm

S3: laser ablated; Si-NC ~ 15 nm

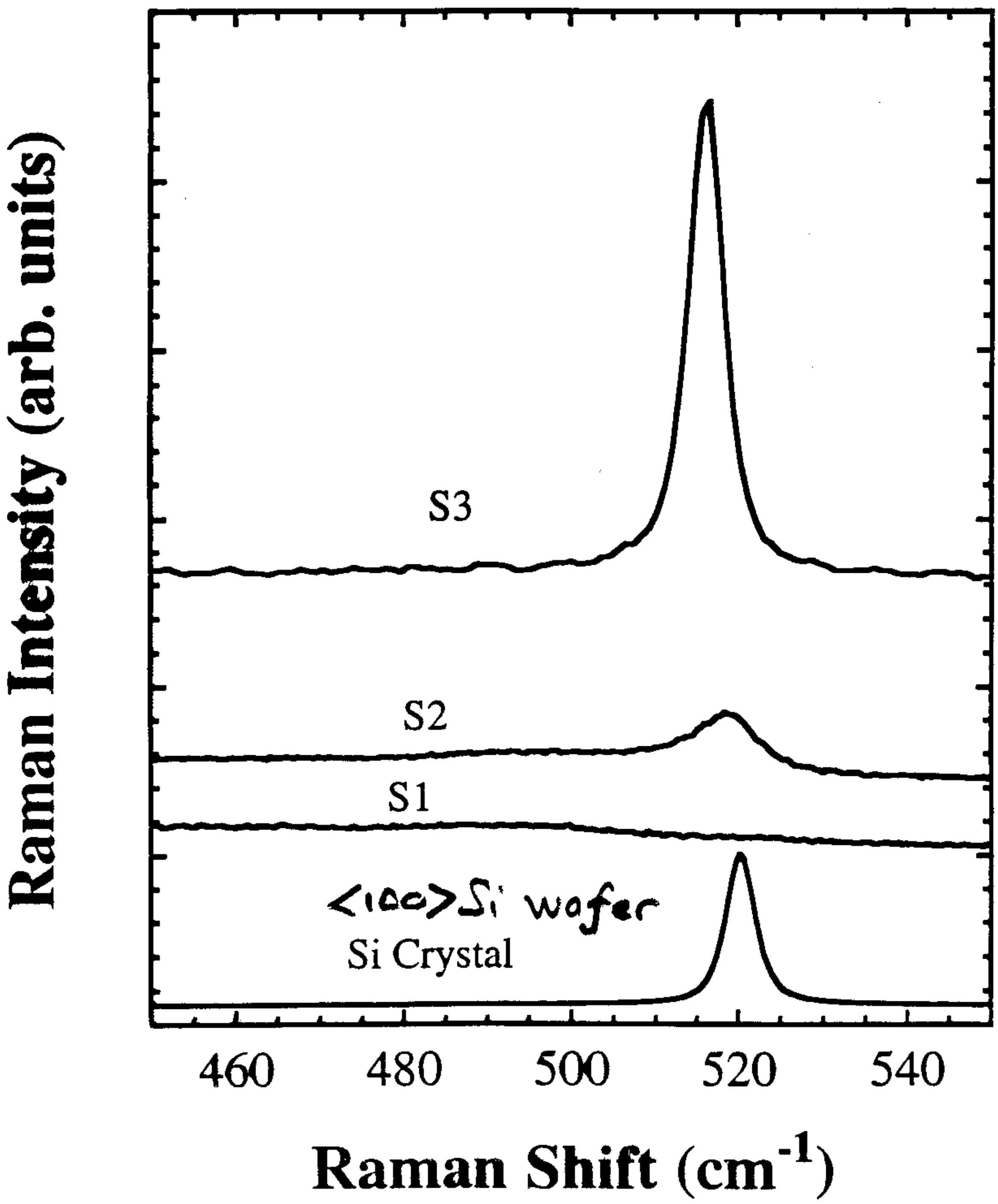


Fig 7

Conclusions (Laser Ablated)

- Large nonlinearity arises from close-packed nanocrystallites of silicon within the micron-sized droplets -- a "cooperative" or "collective" phenomena.
- Thermal contributions to $\chi^{(3)}_{Re}$ appears to be small @ 532 nm

  (τ_p 50 ps) 100 MHz : $\chi_{Re}^{(3)} = 6x10^{-4}$esu

  1 MHz : $\chi_{Re}^{(3)} = 4x10^{-4}$esu
- Consistent with the ns result (532 nm, 355nm) ultrashort pulse excitation results in a peak in χ_{Re} at 532 nm and a nearly 3 order of magnitude drop at 375 nm and 790 nm

  → resonance?

- Underlying Physics is still not well understood
- Future experiment:

  Time resolve the nonlinearity for both the laser ablated and ion implanted Si NC

Conclusions (Ion Implanted)

- Z-scan data shows a large β which is attributed to 2-photon absortption.
- Nonlinearity is dominated by absorption, unable to extract information about $\chi^{(3)}_{Re}$
- Thermal contributions to $\chi^{(3)}_{Im}$ decrease as a function of repetition rate.

@790 nm

Rep. rate	β	I_o
82 MHz	2.193×10^{-3} cm/W	4×10^{6} W/cm^2
4 MHz	6.18×10^{-4} cm/W	8.12×10^{6} W/cm^2
.8 MHz	3.26×10^{-4} cm/W	2×10^{7} W/cm^2

Apparent larger thermal contribution to the nonlinearity than the laser ablated Si NC

Future Experiments

- Efforts to time resolve the nonlinearity have been hampered by low pulse energies (pJ) – 50 kHz Ti:sapphire regenerative amplifier (μJ) on order.
- Time-resolved two-color Z-scan measures nondegenerate nonlinear absorption (β) and nonlinear refraction (n_2) – time resolve separately the sign and the magnitude of β and n_2 at frequency ω_{probe} that are due to the presence of a strong excitation at frequency ω_{exc}.
- For high-speed optical switching a fast electronic nonlinearity is needed over a slow thermal nonlinearity and thus time-resolved nonlinearity measurements. Another approach – use fs pulses **(λ=1550 nm)** to generate a third harmonic signal (λ=517 nm) – a third harmonic signal can only result from an electronic nonlinearity.