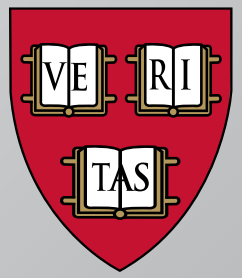
Simultaneous multiphoton absorption in rutile (TiO_2) across the half-bandgap

Christopher C. Evans, Jonathan D. B. Bradley,
Erwin A. Martí-Panameño and Eric Mazur



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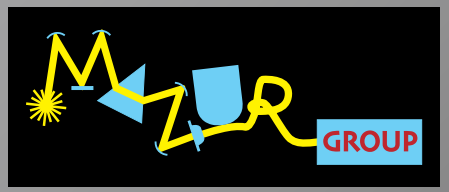


Introduction

TiO_2 Material Properties

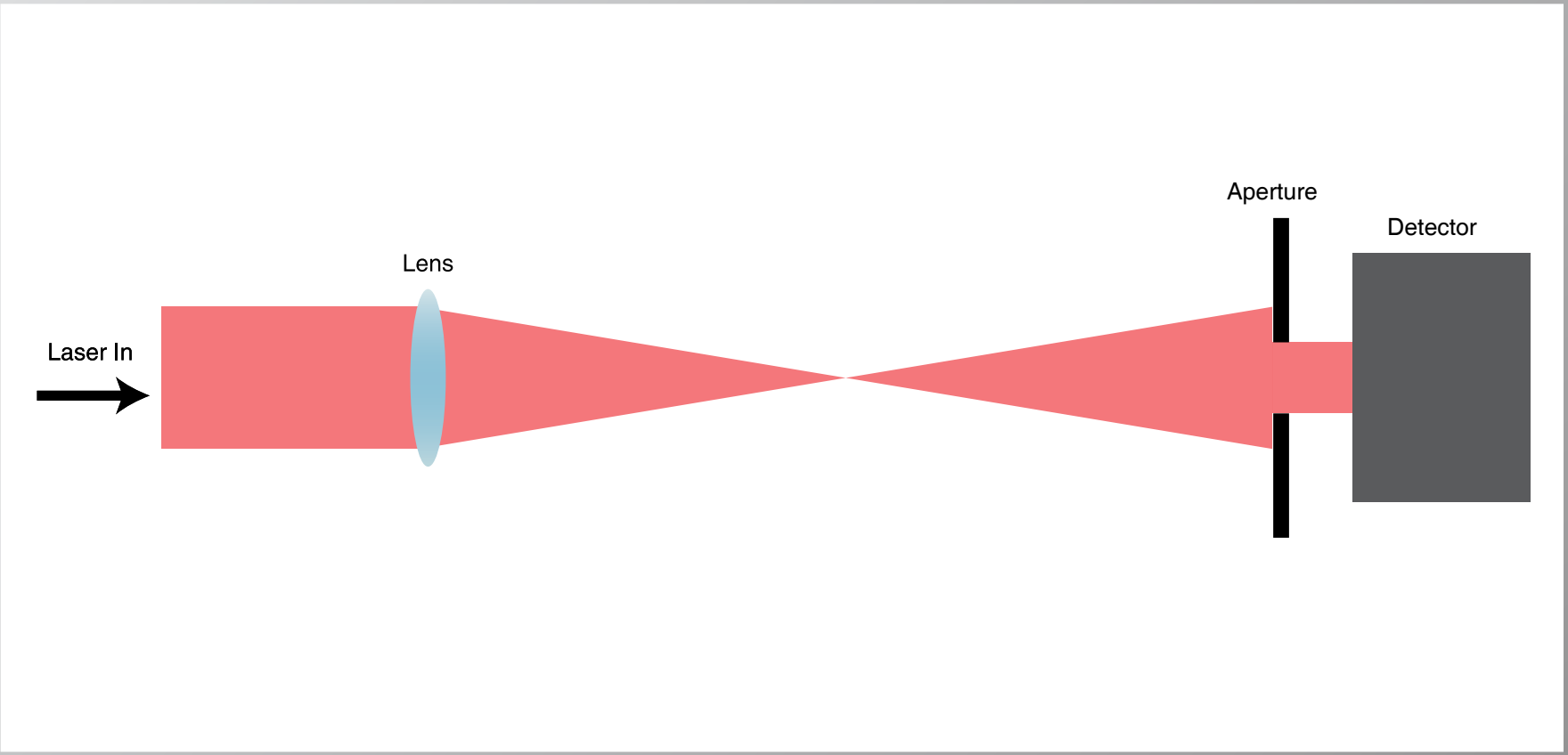
Large ultrafast nonlinearity:	30 x silica
High index of refraction:	2.5
Wide bandgap:	3.1 eV
Low two-photon absorption:	≥ 800 nm

Several polymorphs:

Rutile, Anatase, Brookite and Amorphous

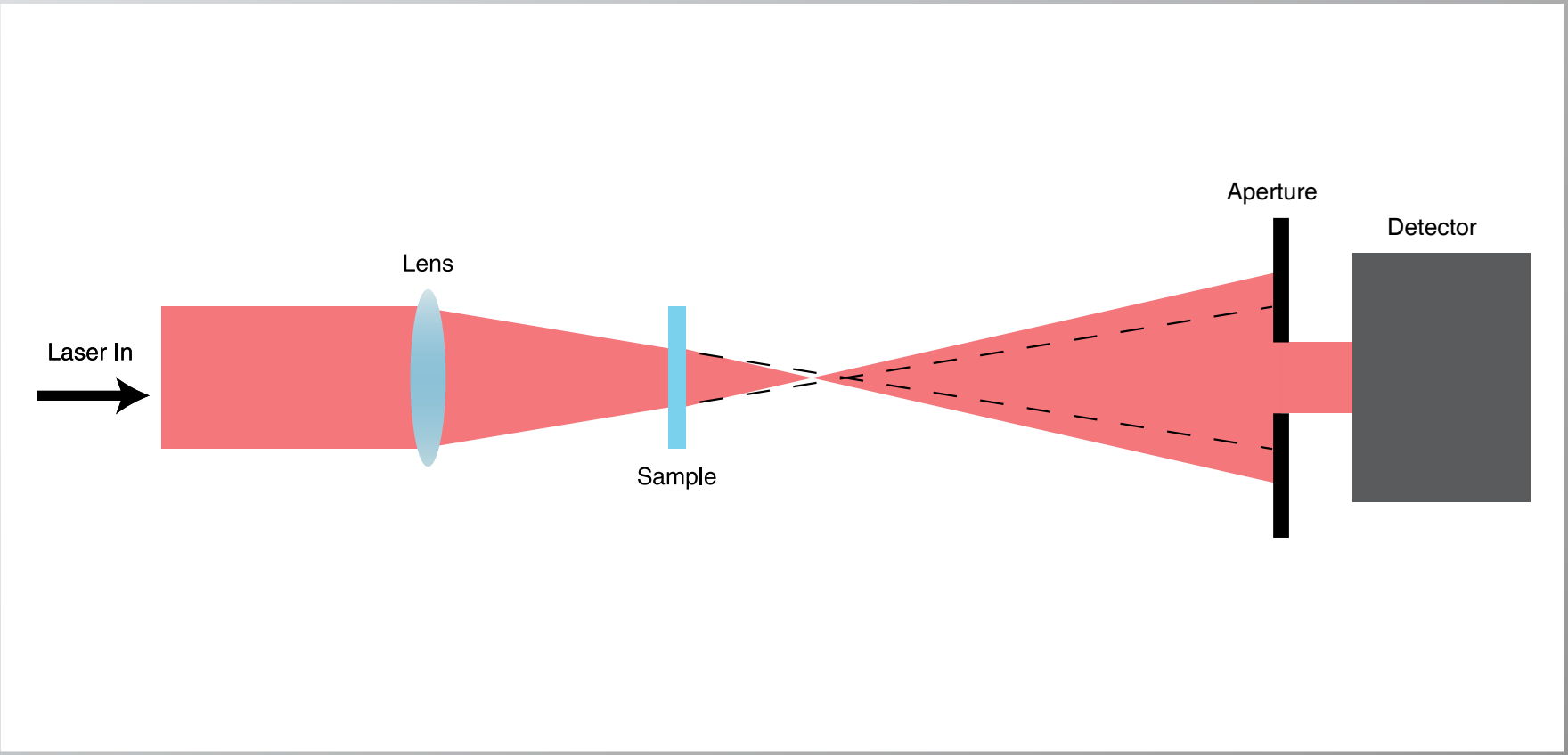
Introduction

Nonlinear material properties: z-scan



Introduction

Nonlinear material properties: z-scan



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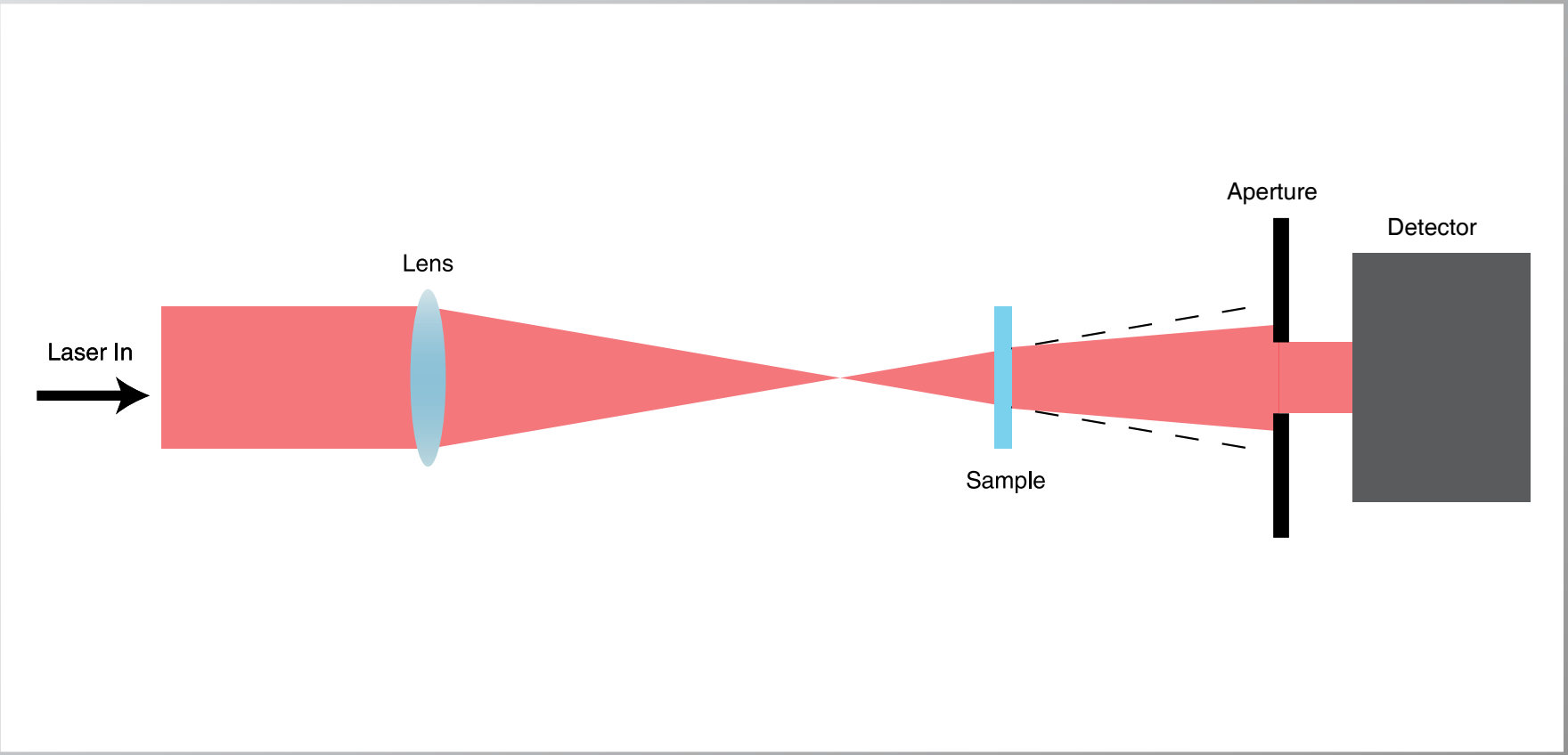
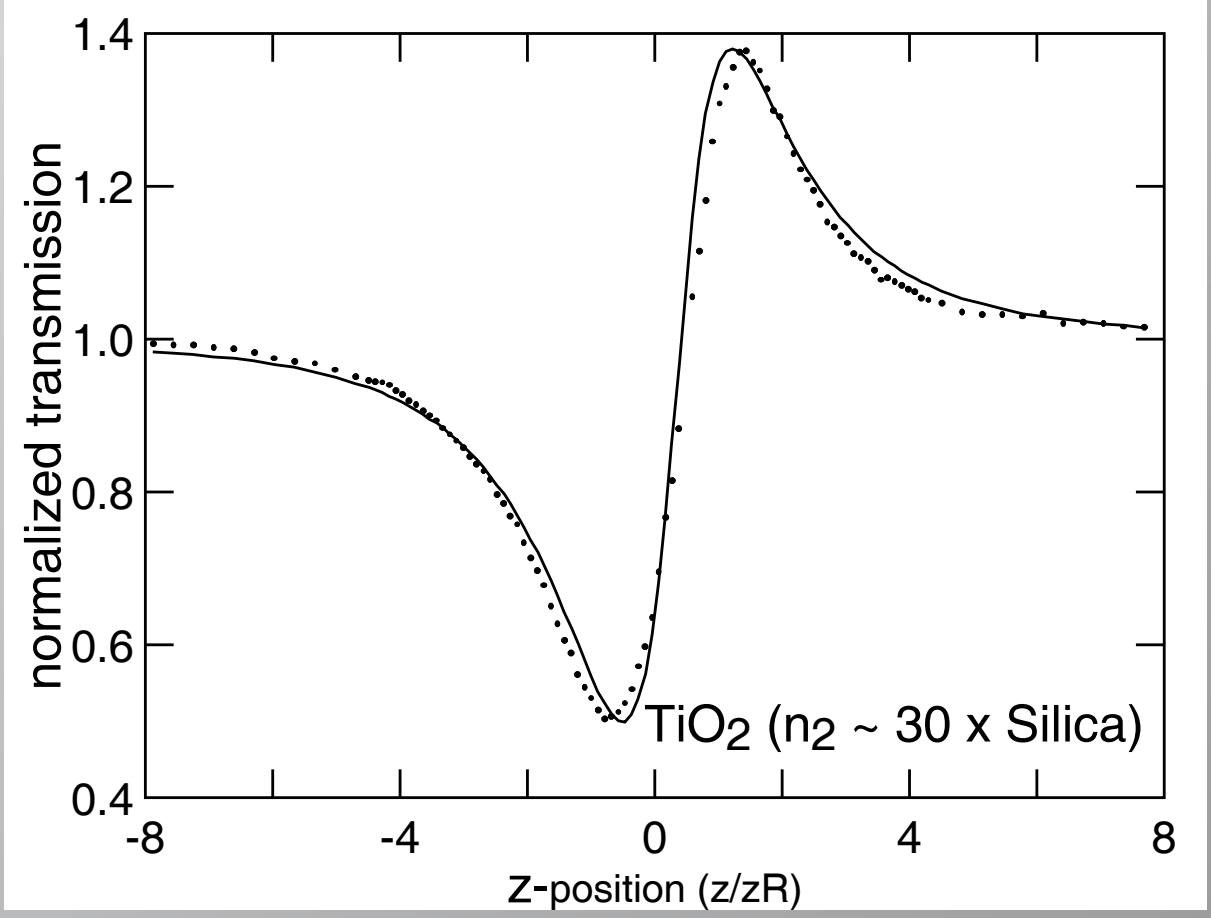
Nonlinear material properties: z-scan



Introduction

Nonlinear refraction (n_2 at 800 nm)



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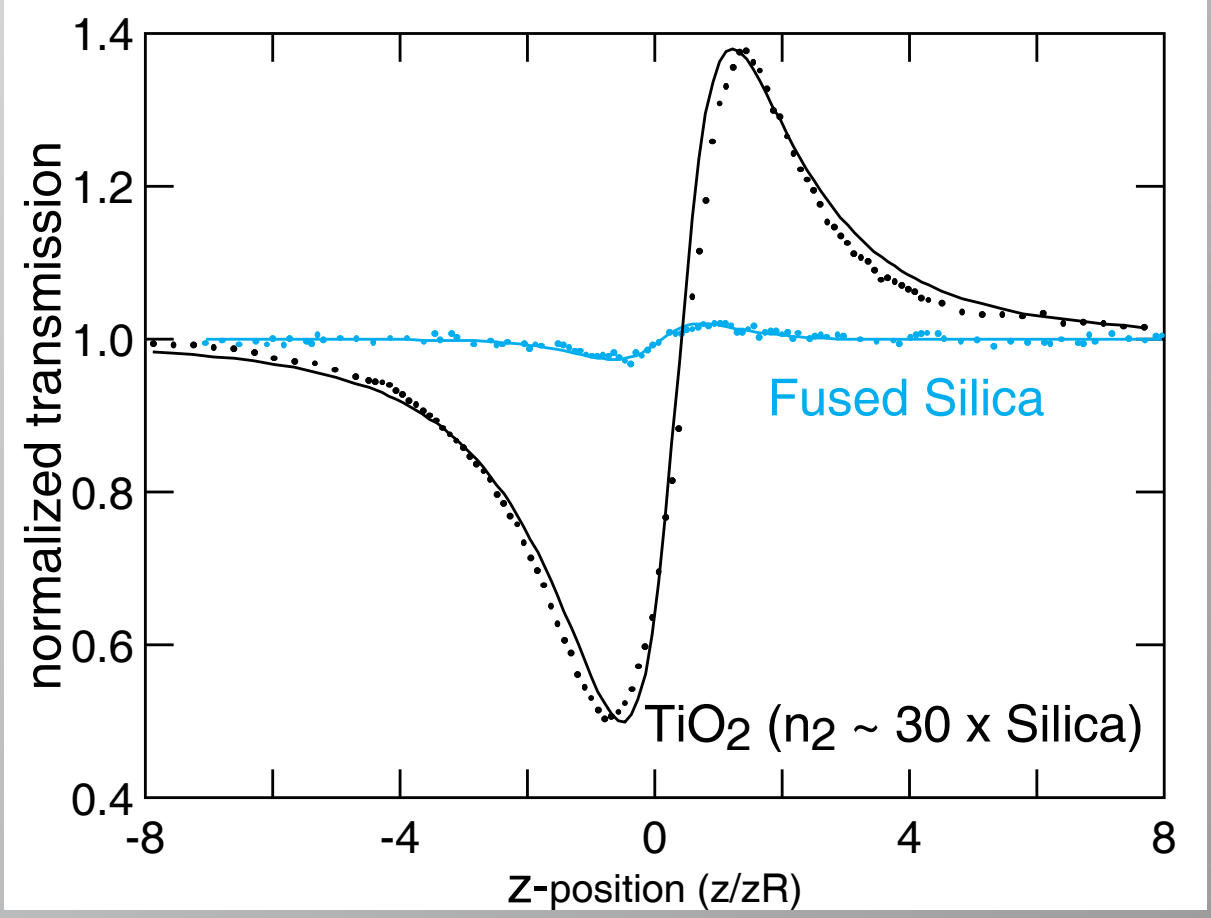
Nonlinear refraction (n_2 at 800 nm)

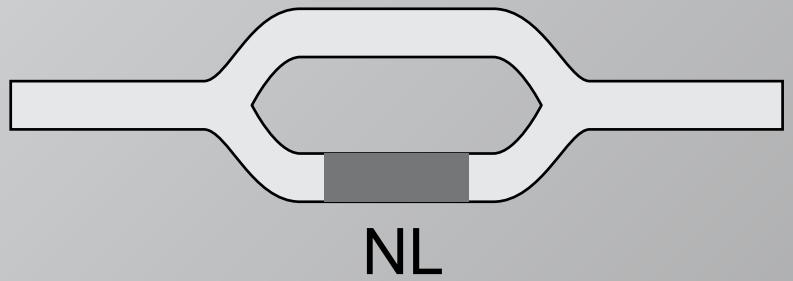


Introduction

high nonlinearity ≠ working device

Introduction

$$n = n_0 + n_2 I$$



Introduction

high nonlinearity ≠ working device

Full modulation requires <u>at least</u>
a 2π nonlinear phase shift

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$$\phi_{NL} = \frac{2\pi n_2}{\lambda} IL$$

Introduction

high nonlinearity ≠ working device

Full modulation requires <u>at least</u>
a 2π nonlinear phase shift

$$\phi_{NL} = \frac{2\pi n_2}{\lambda} IL$$

$$\phi_{NL} = \frac{2\pi n_2}{\lambda} \int_0^L I(z)dz$$

Introduction

high nonlinearity ≠ working device

Full modulation requires <u>at least</u>
a 2π nonlinear phase shift

$$\frac{n_2}{\beta\lambda} > 1 \qquad\qquad \frac{n_2 I_0}{\alpha\lambda} > 1$$

Introduction

high nonlinearity ≠ working device

Full modulation requires <u>at least</u>
a 2π nonlinear phase shift

$$\frac{n_2}{\beta\lambda} > 1$$

two-photon
absorption
figure of merit

$$\frac{n_2 I_0}{\alpha\lambda} > 1$$

one-photon
absorption
figure of merit

Introduction

high nonlinearity ≠ working device

Full modulation requires <u>at least</u>
a 2π nonlinear phase shift

$$\frac{n_2}{\beta\lambda} > 1$$

material
limit

$$\frac{n_2 I_0}{\alpha\lambda} > 1$$

“fabrication”
limit

Introduction

$$\frac{n_2 I_0}{\alpha\lambda} > 1$$

1PA FOM

$$\frac{n_2}{\beta\lambda} > 1$$

2PA FOM

$$\frac{n_2}{\gamma\lambda I_0} > 1$$

3PA FOM

Introduction

$$\frac{n_2 I_0}{\alpha\lambda} > 1$$

1PA FOM

$$\frac{n_2}{\beta\lambda} > 1$$

2PA FOM

$$I_{\max}^{3\mathrm{PA}} = \frac{n_2}{\gamma\lambda}$$

3PA FOM

Outline

Experimental

Modeling

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Conclusions

Experimental

Single crystal rutile samples:

Floating-zone method

1-mm thick

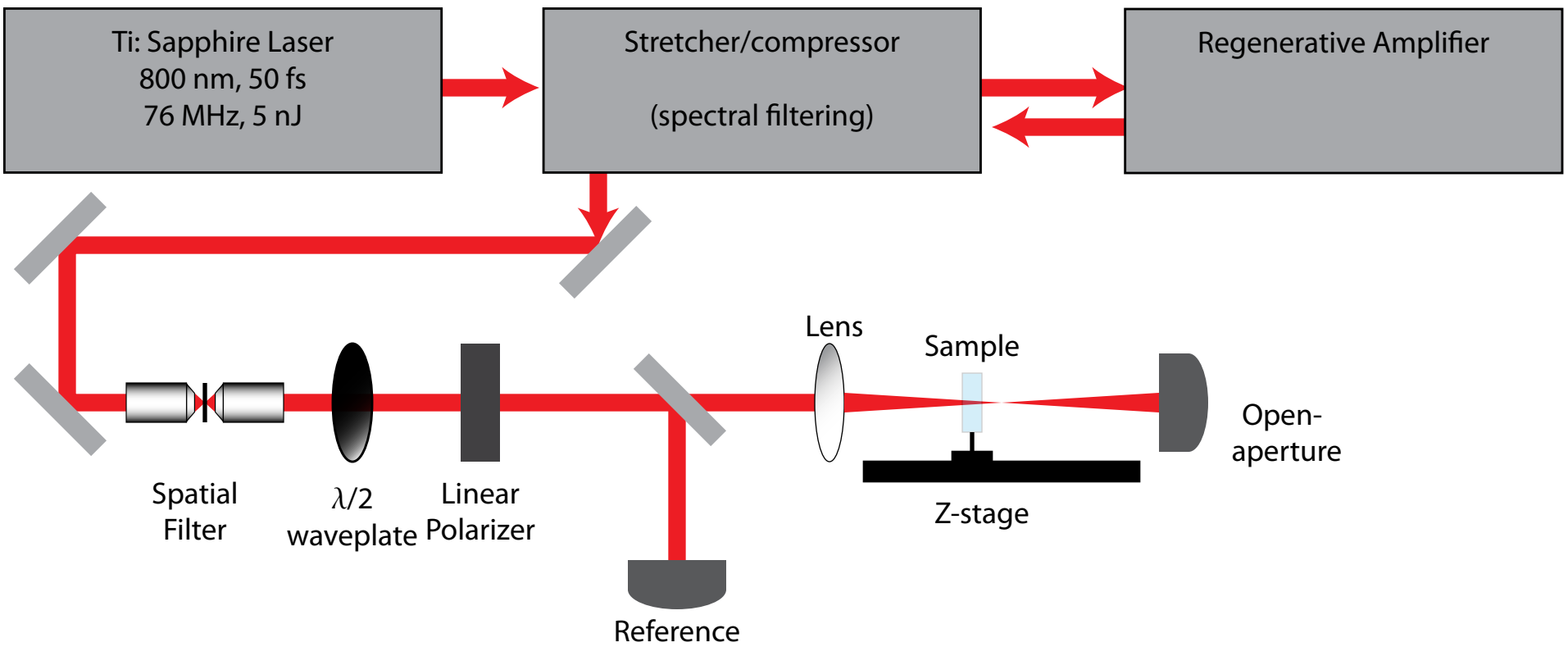
Two-cuts:

(001)-cut, $E \perp c$ only

(100)-cut, $E \perp c$ and $E \parallel c$

Experimental

Experimental setup



Experimental

Laser Parameters

λ_0 (nm)	$2\hbar\omega$ (eV)	$\Delta\lambda$ (nm)	τ (fs)
774	3.20	4	290
813	3.05	10	174
800	3.10	30	50

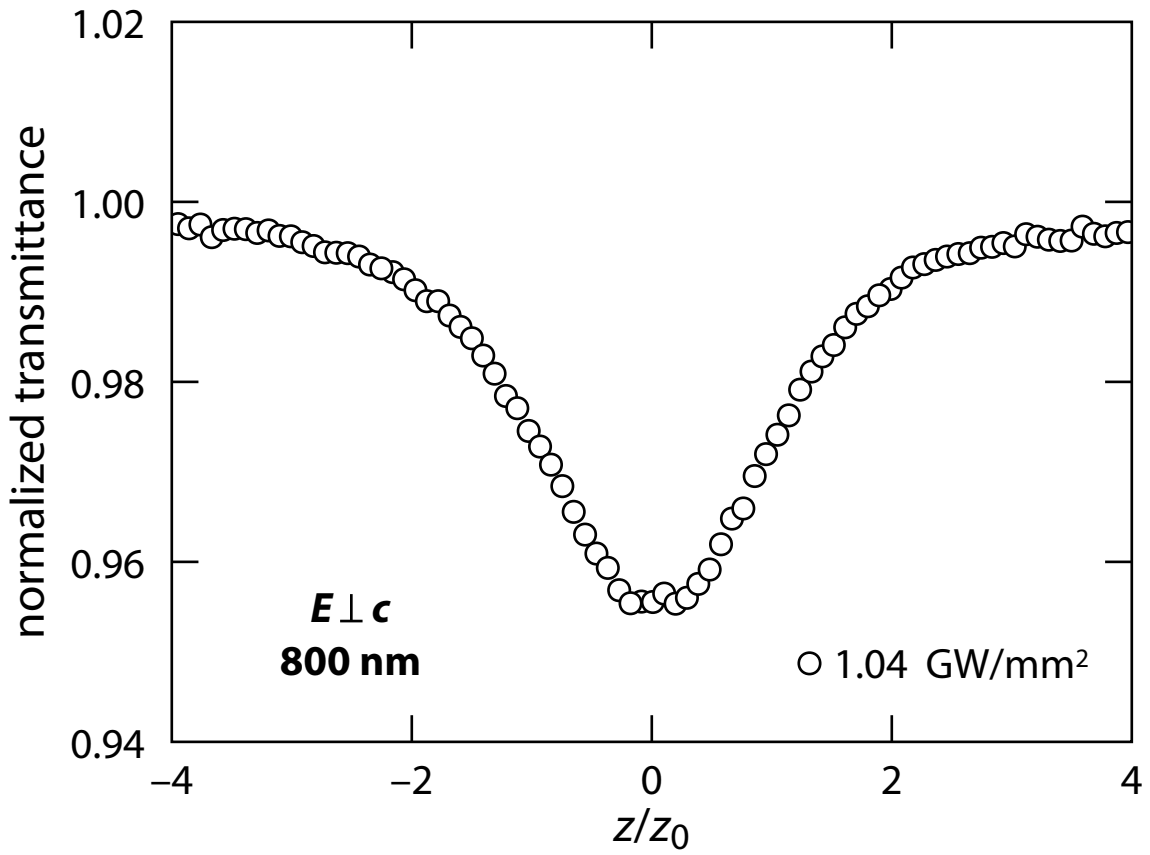
Experimental

Laser Parameters

λ_0 (nm)	M^2	w_{x0}/w_{y0}	$I_{0,\max}$ (GW/mm^2)
774	< 1.13	0.78	0.21
813	< 1.18	0.92	0.32
800	< 1.06	0.95	1.04

Experimental

Open aperture Z-scan of rutile



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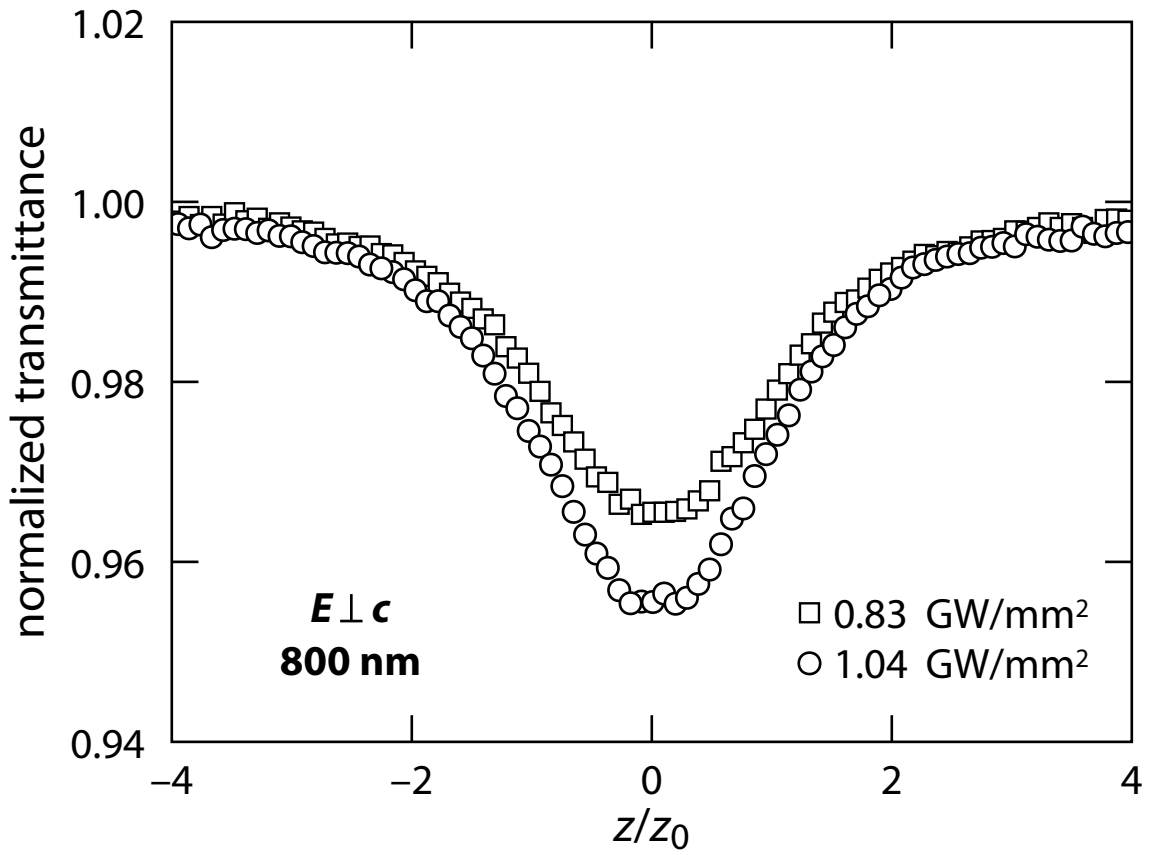

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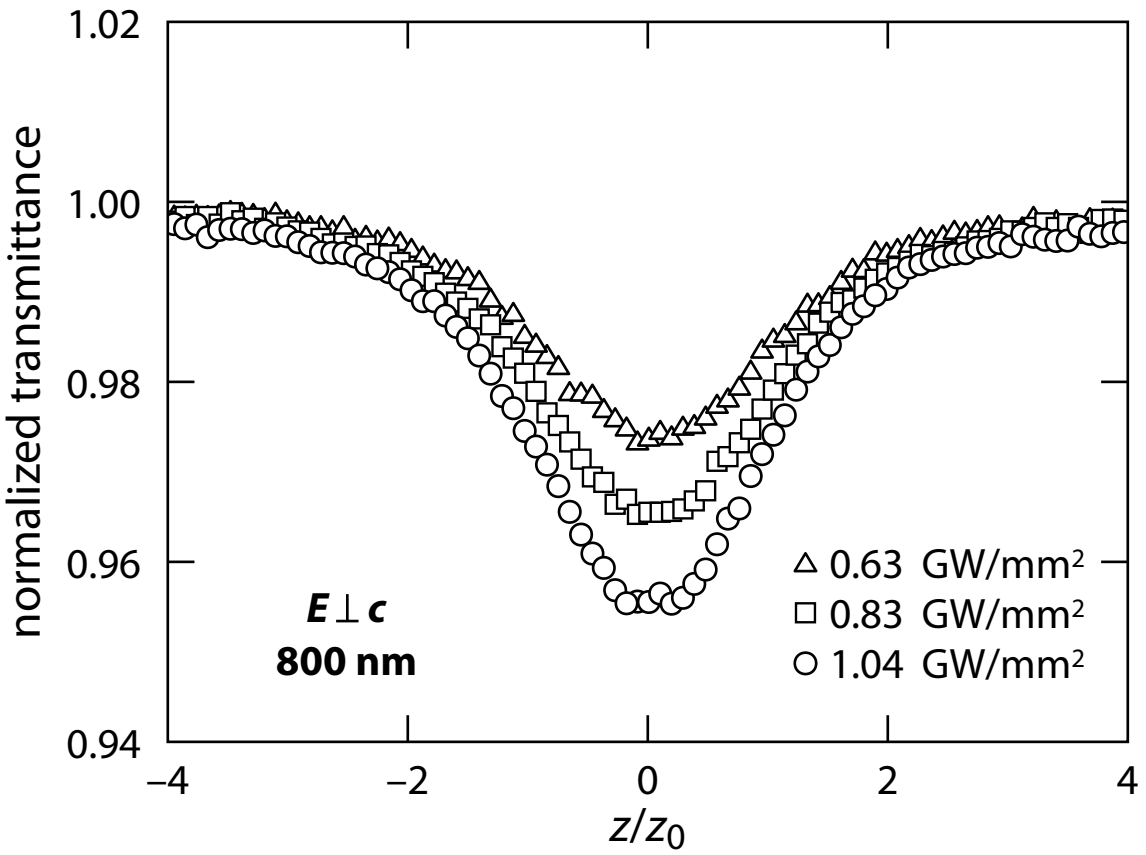

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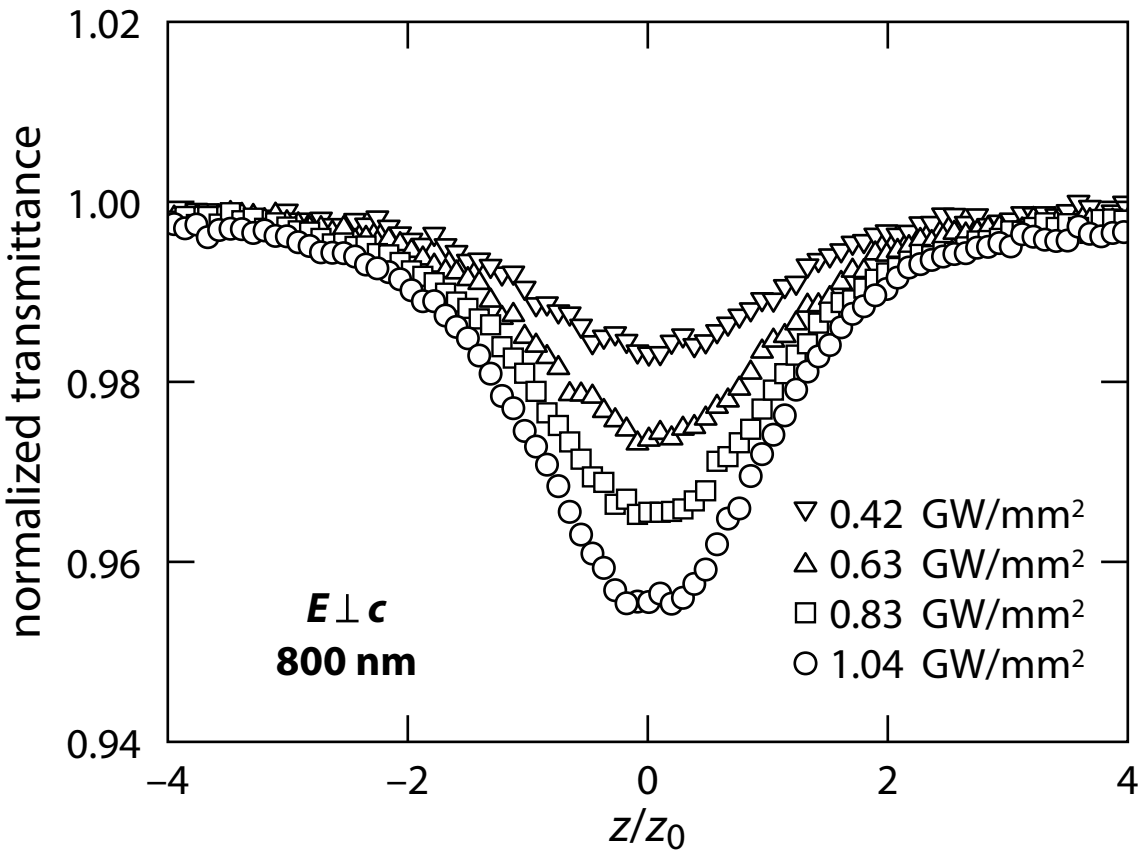

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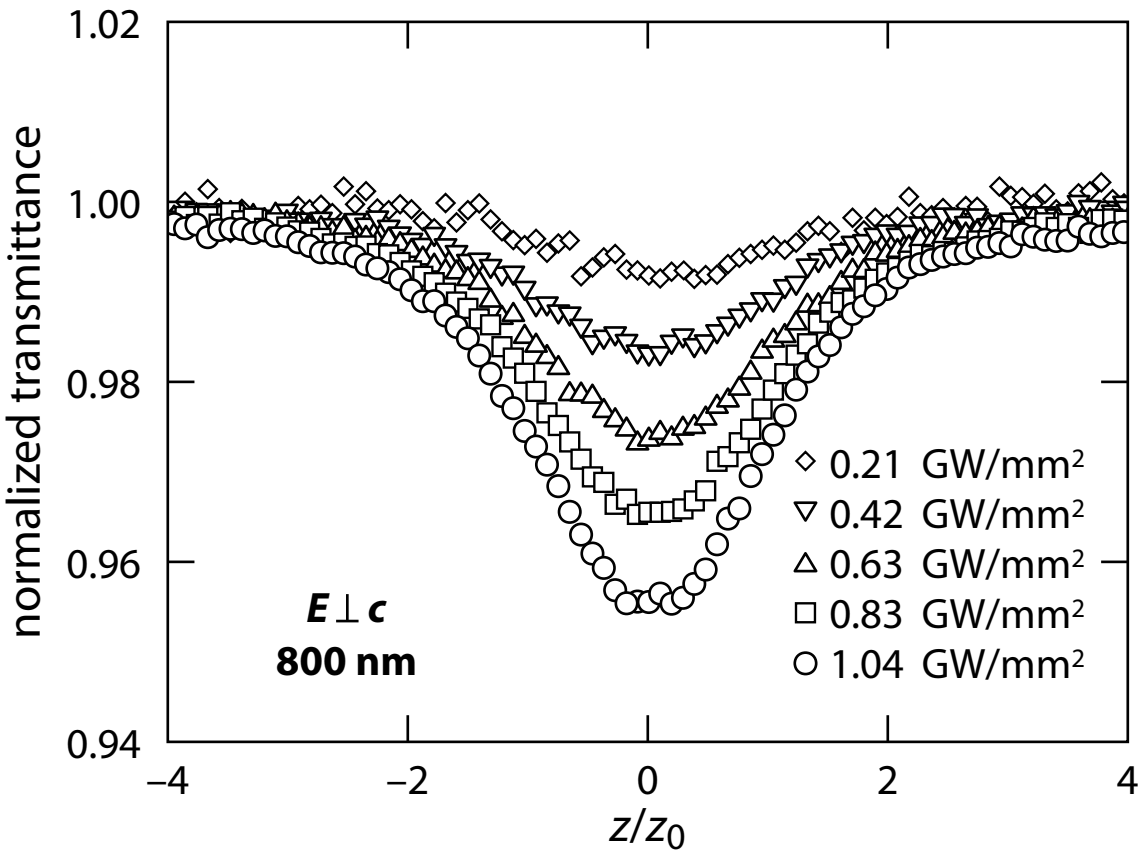

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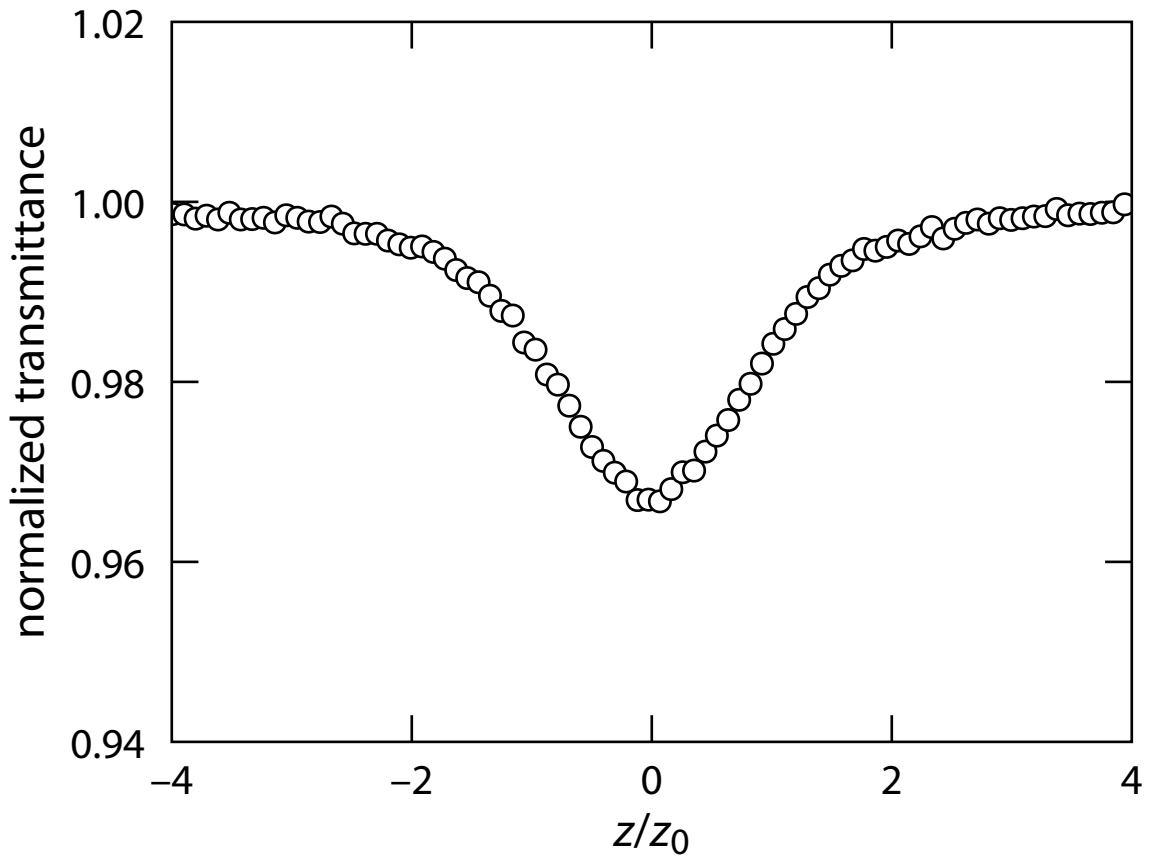
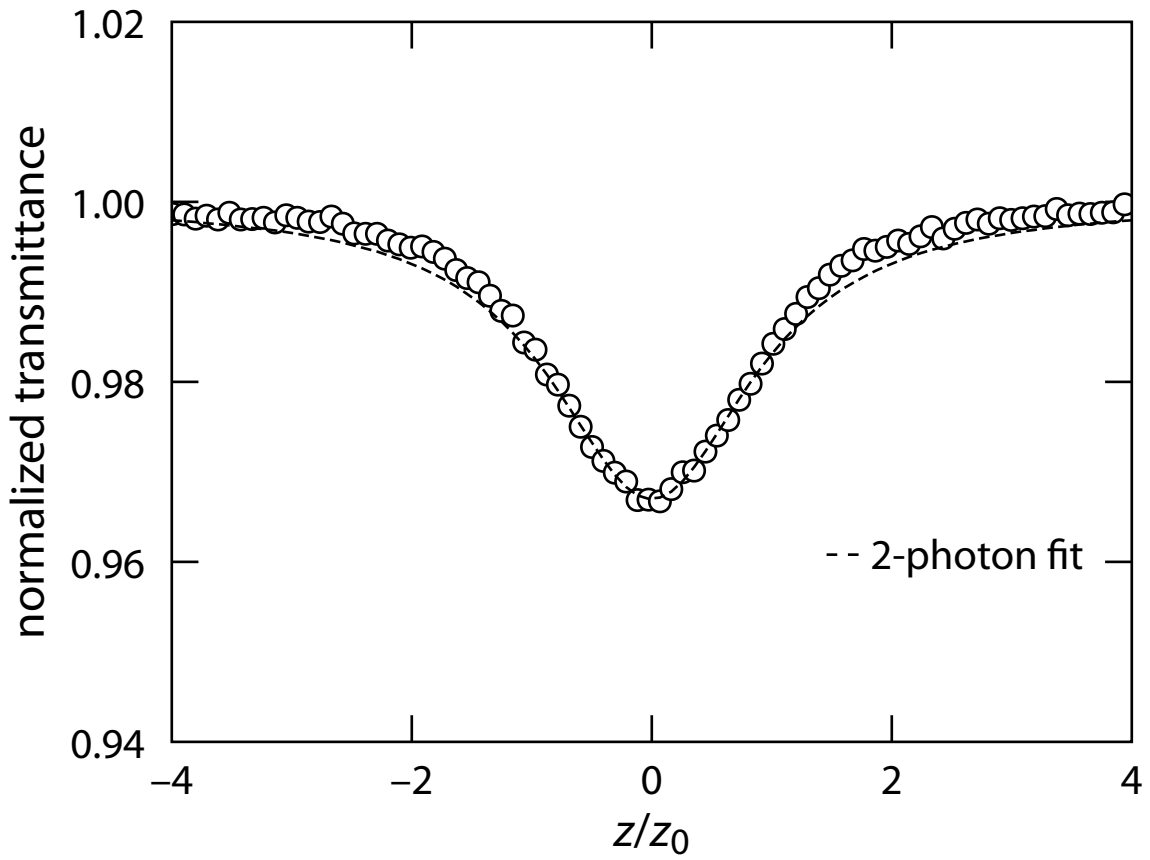
Modeling

Open aperture Z-scan of rutile



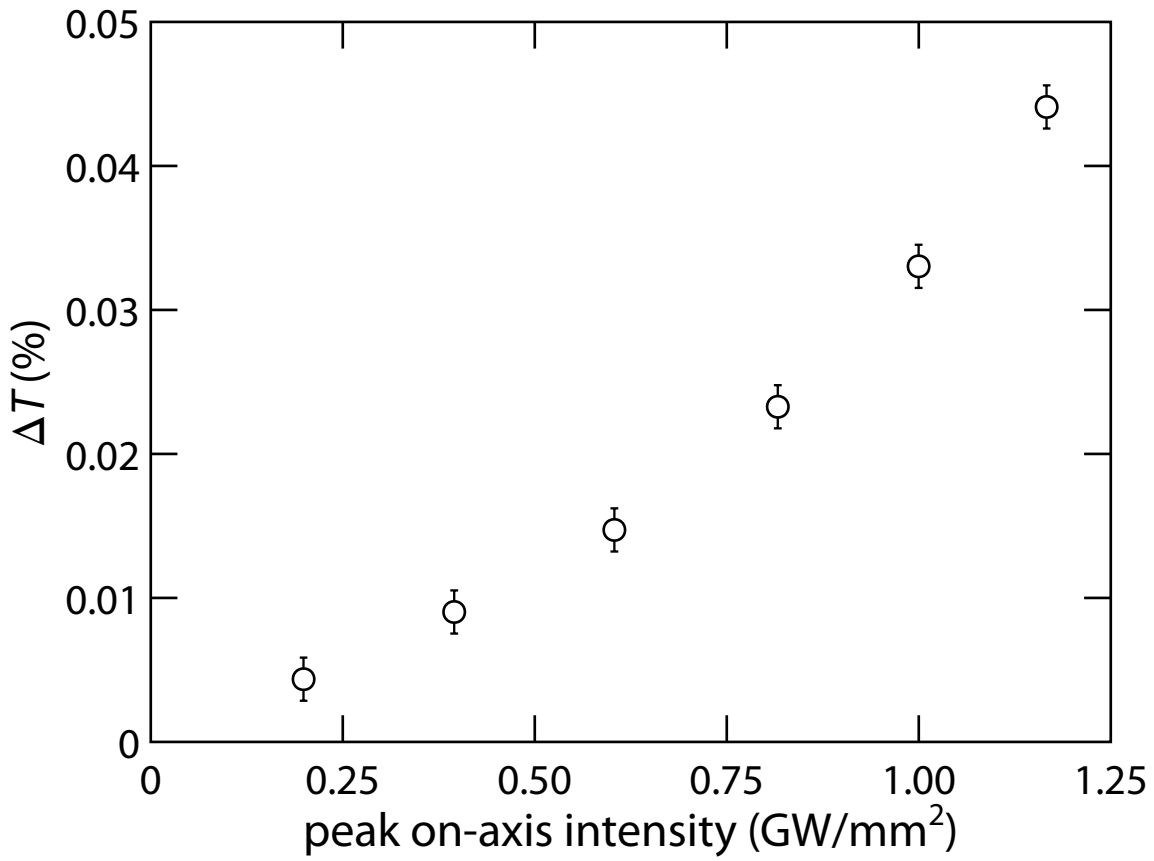
Modeling

Open aperture Z-scan of rutile



Modeling

Open aperture Z-scan of rutile



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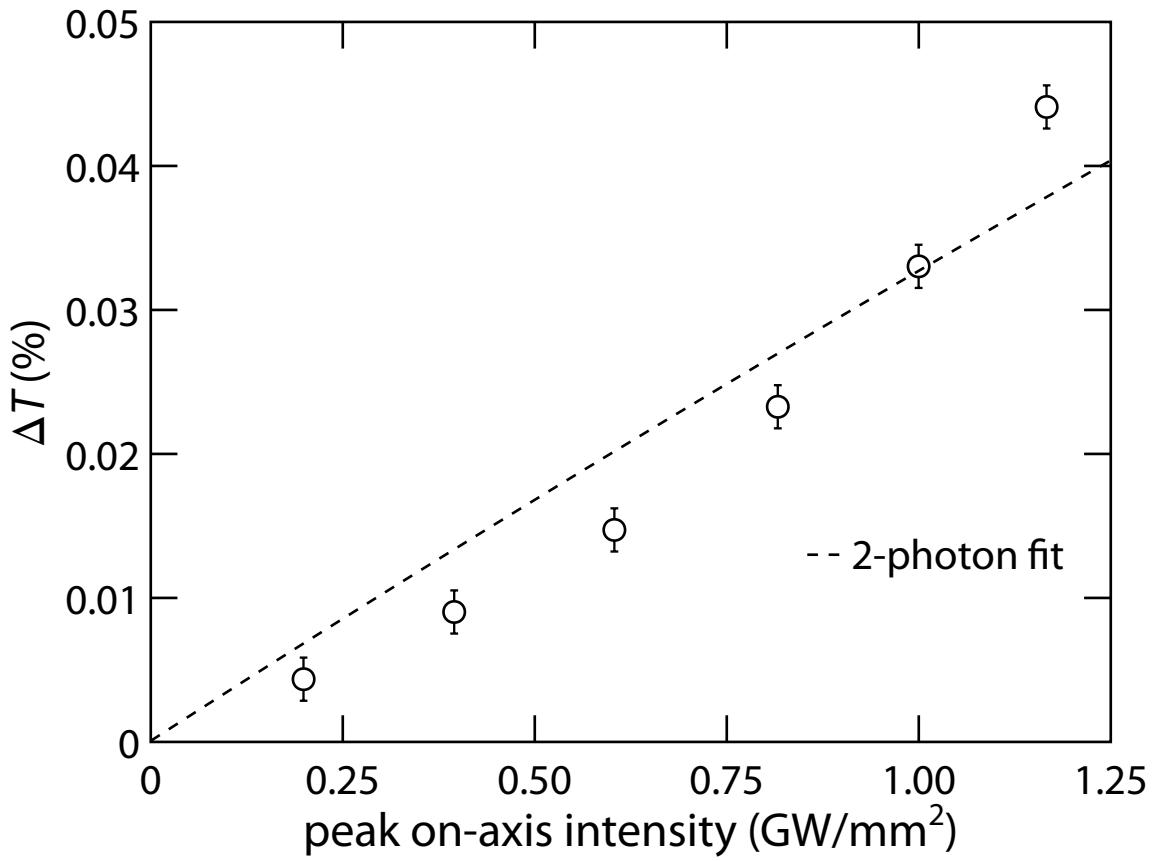

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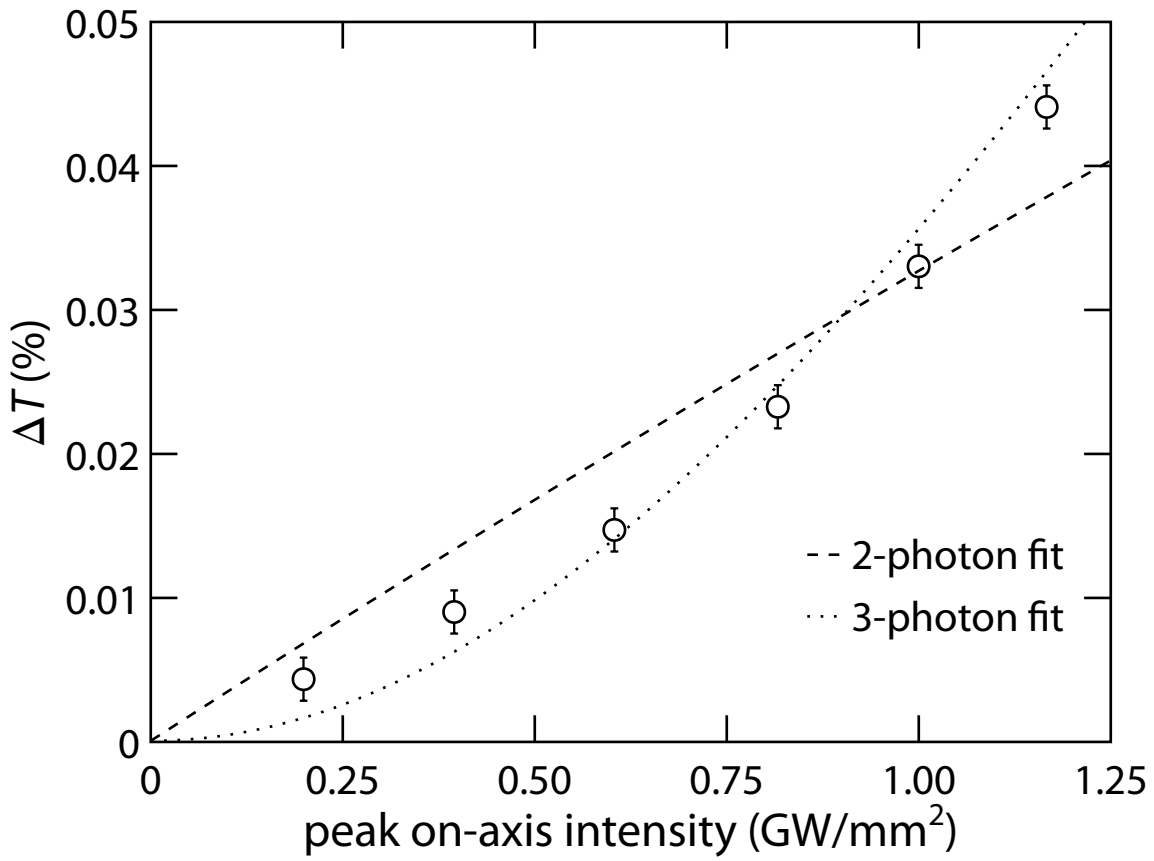

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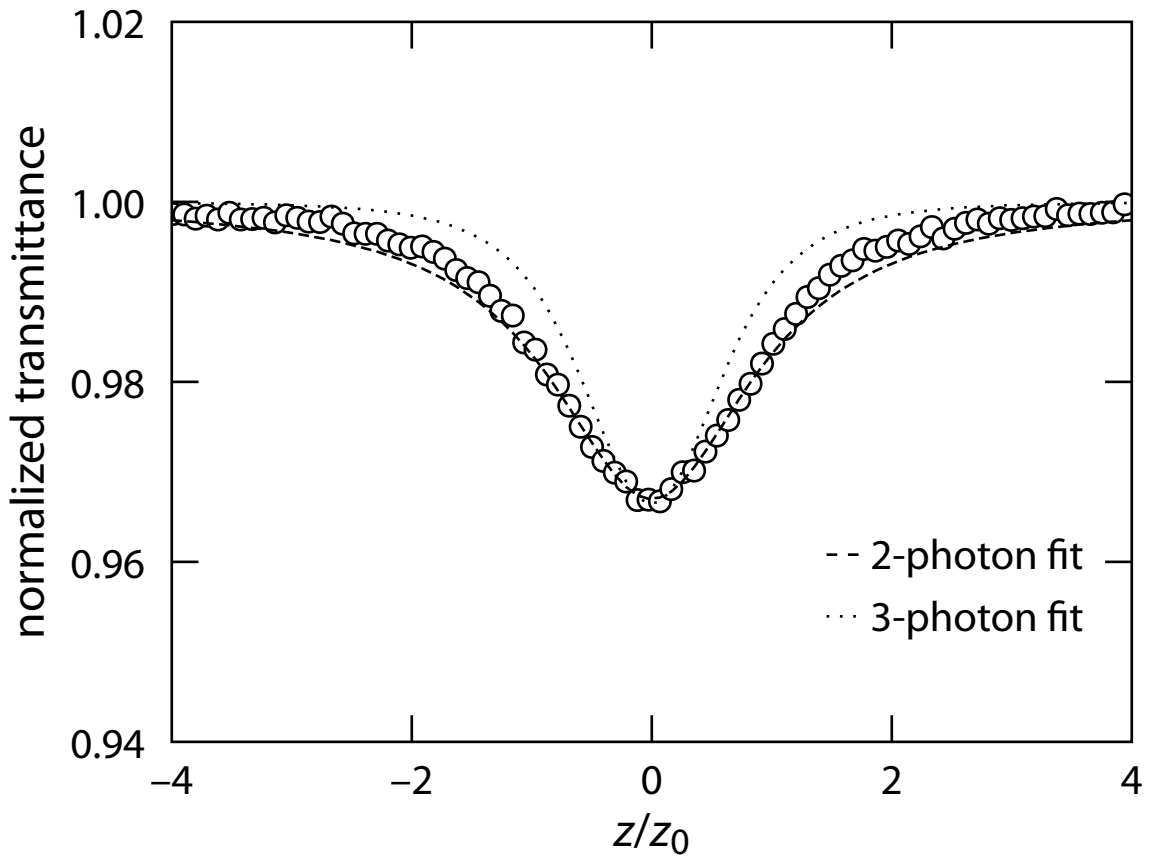

Modeling

Mixed multiphoton model

$$I_{in}\left[x, y, z, P(t)\right] = \frac{2P(t)}{\pi\, w_x(z) w_y(z)} \exp\left[-\frac{2x^2}{w_x^2(z)} - \frac{2y^2}{w_y^2(z)}\right]$$

Modeling

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$$w_x(z) = \sqrt{{w_{x0}}^2 + \left(\frac{M_x^2 \lambda_0}{\pi w_{x0}}\right)^2 \left(z - z_{x0}\right)^2}$$

Modeling

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$$\alpha(I) = \alpha_0 + \beta I + \gamma I^2$$

Modeling

Mixed multiphoton model

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$$\frac{dI}{dz'} = -\alpha(I) I$$

Modeling

Mixed multiphoton model

$$I_{in}[x, y, z, P(t)] = \frac{2P(t)}{\pi\, w_x(z) w_y(z)} \exp\left[-\frac{2x^2}{w_x^2(z)} - \frac{2y^2}{w_y^2(z)}\right]$$

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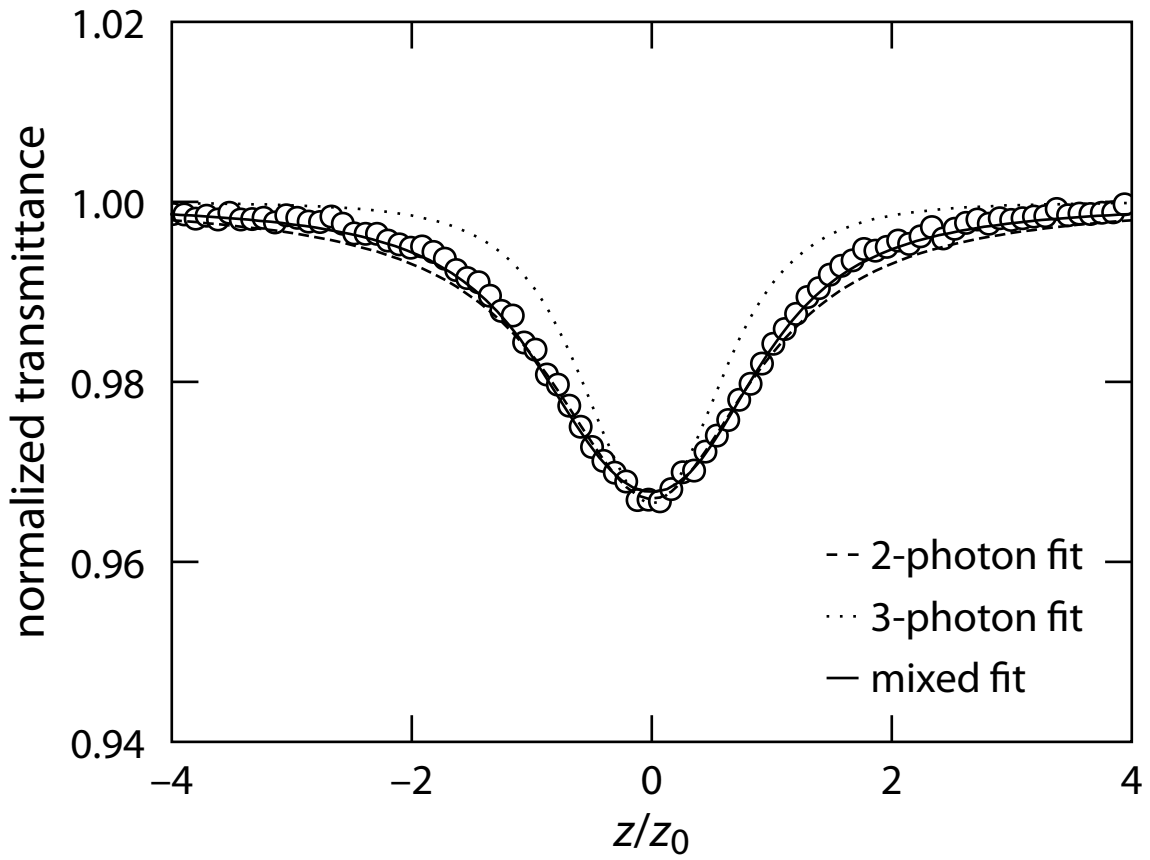
$$\alpha(I) = \alpha_0 + \beta I + \gamma I^2$$

$$\frac{dI}{dz'} = -\alpha(I) I$$

$$T(z) = \frac{\int P(t) T[z, P(t)] dt}{\int P(t)\, dt} = \frac{E_{out}(z)}{E_{in}}$$

Modeling

Open aperture Z-scan of rutile



Modeling

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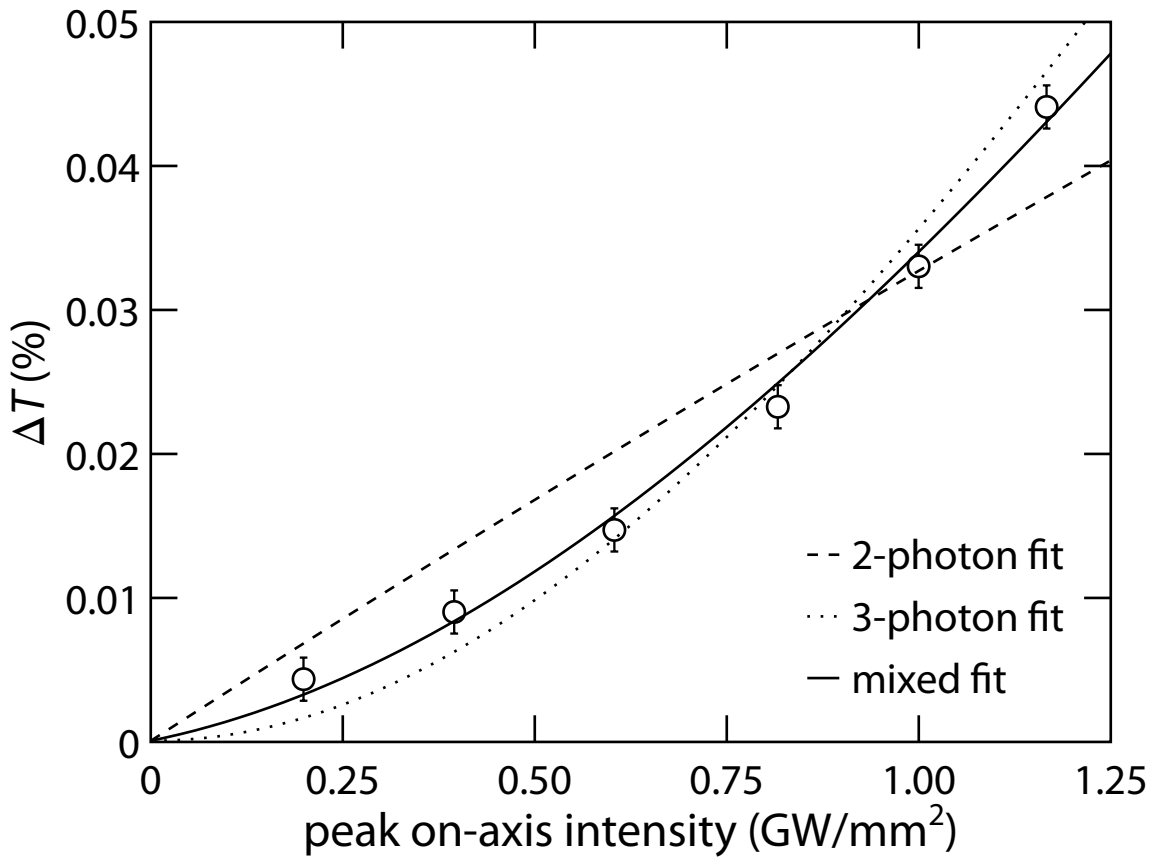
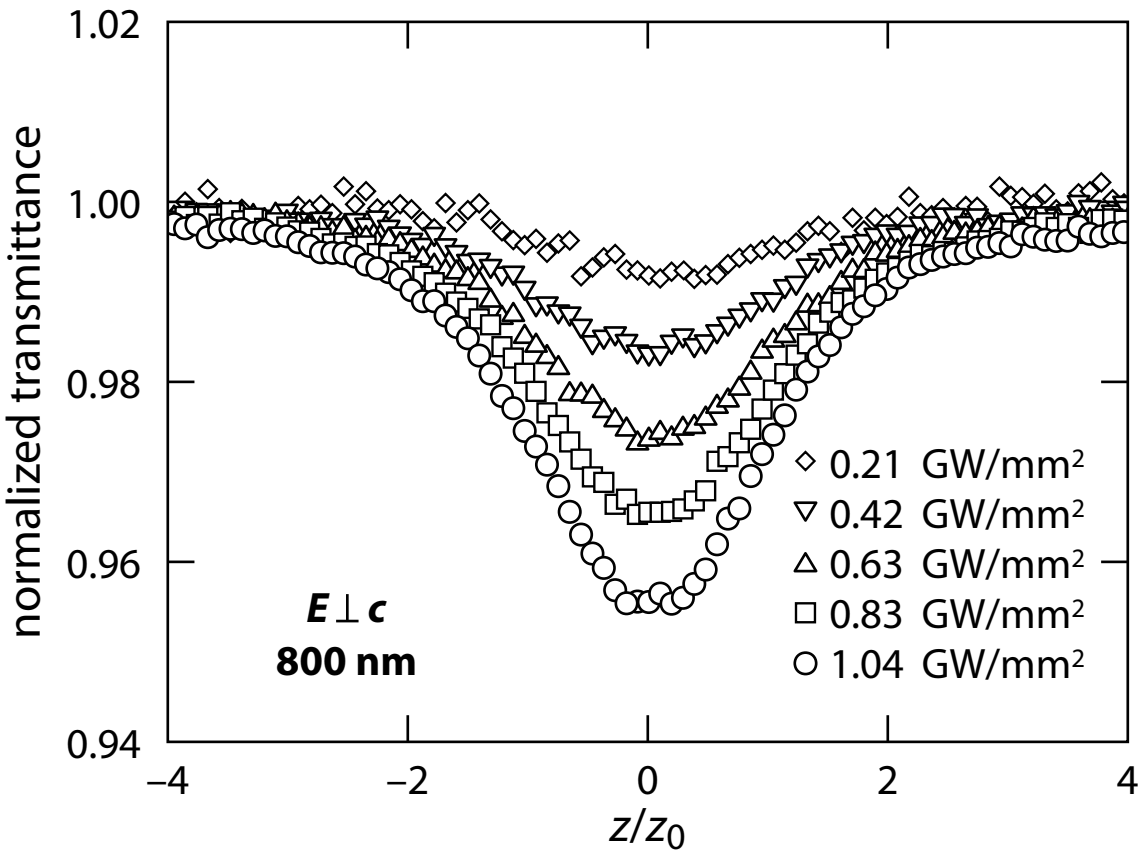


Modeling

Fitting method



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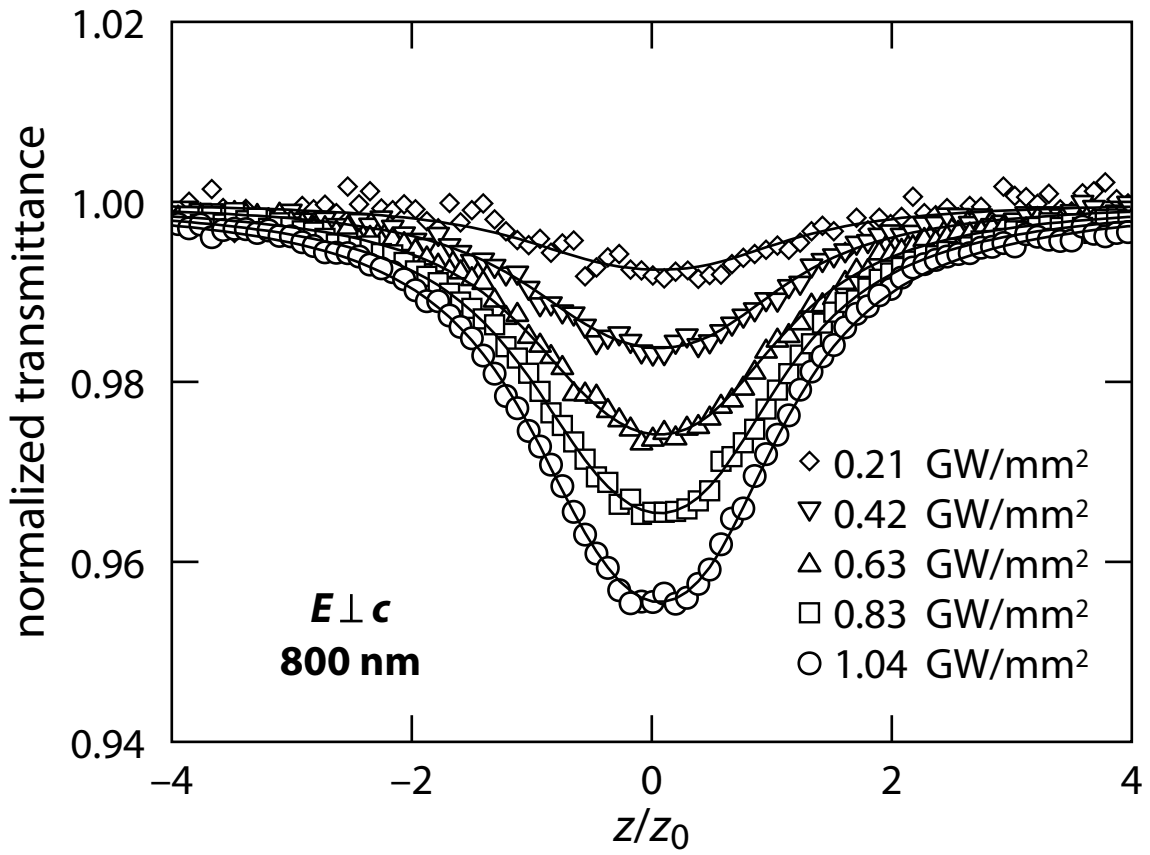

Outline

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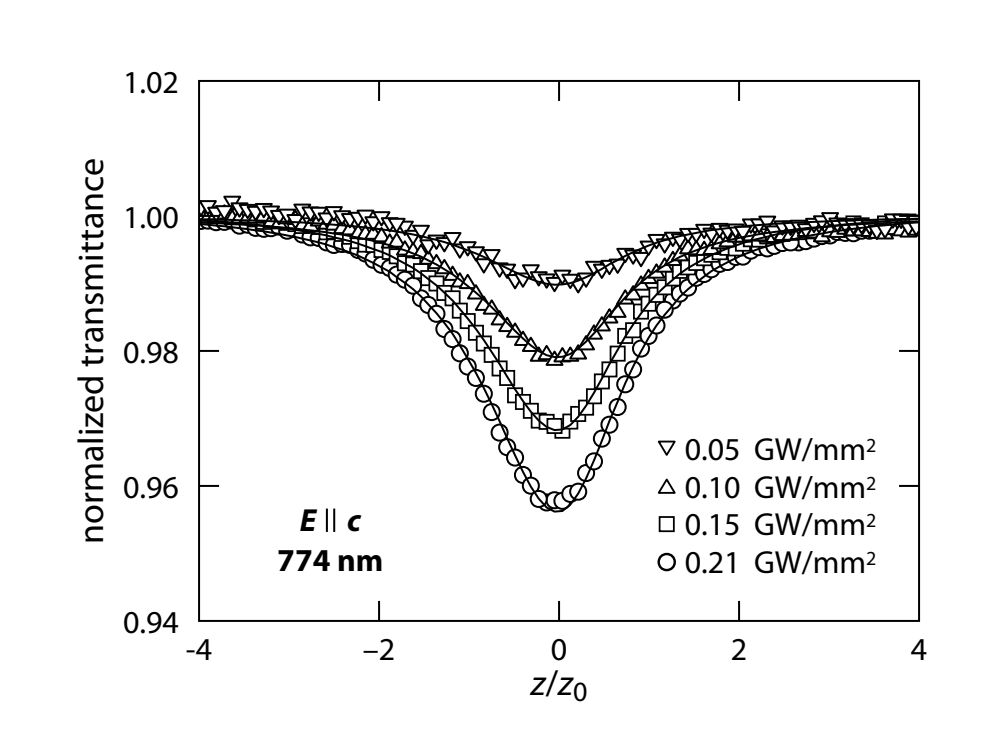
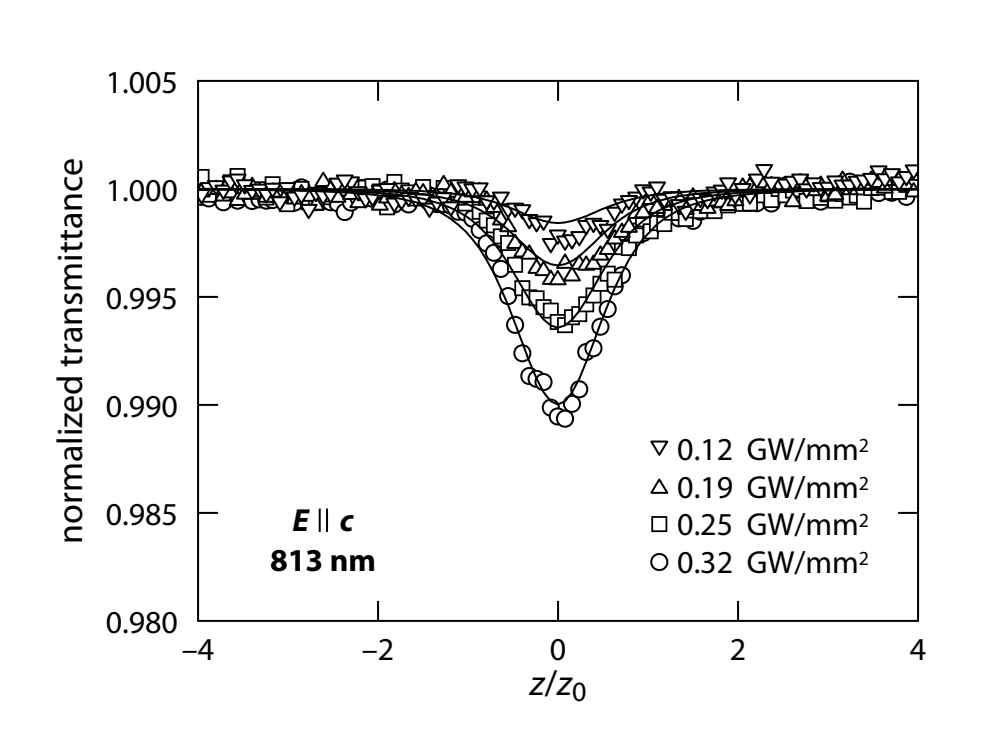
Analysis

$E \| c$

$2\hbar\omega = 3.2$ eV



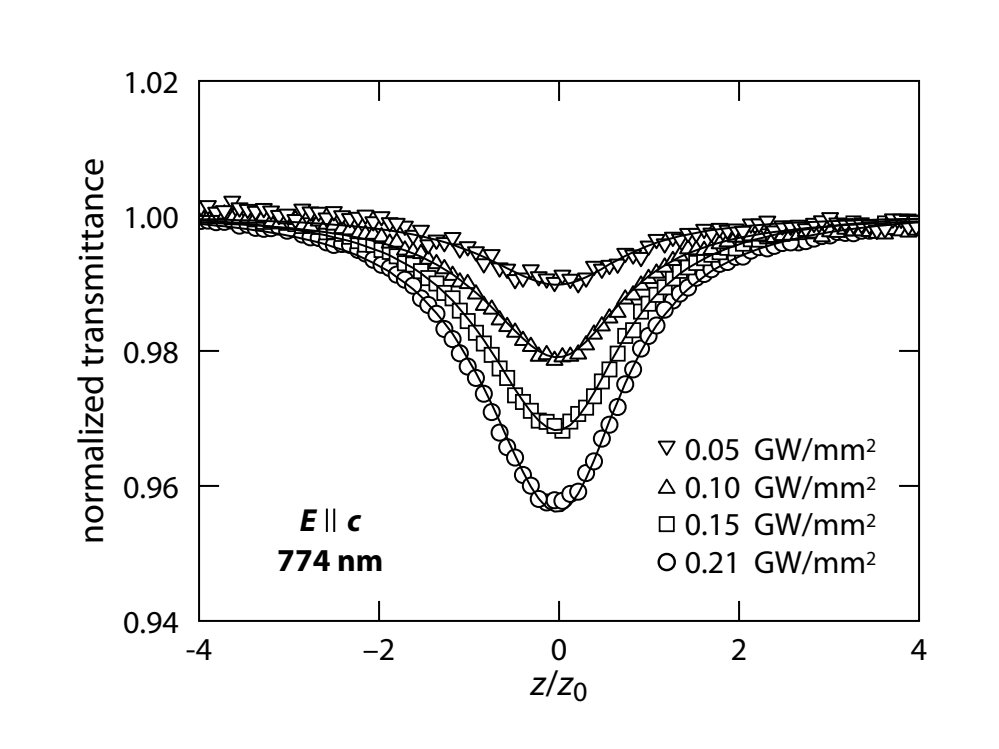
$2\hbar\omega = 3.05$ eV

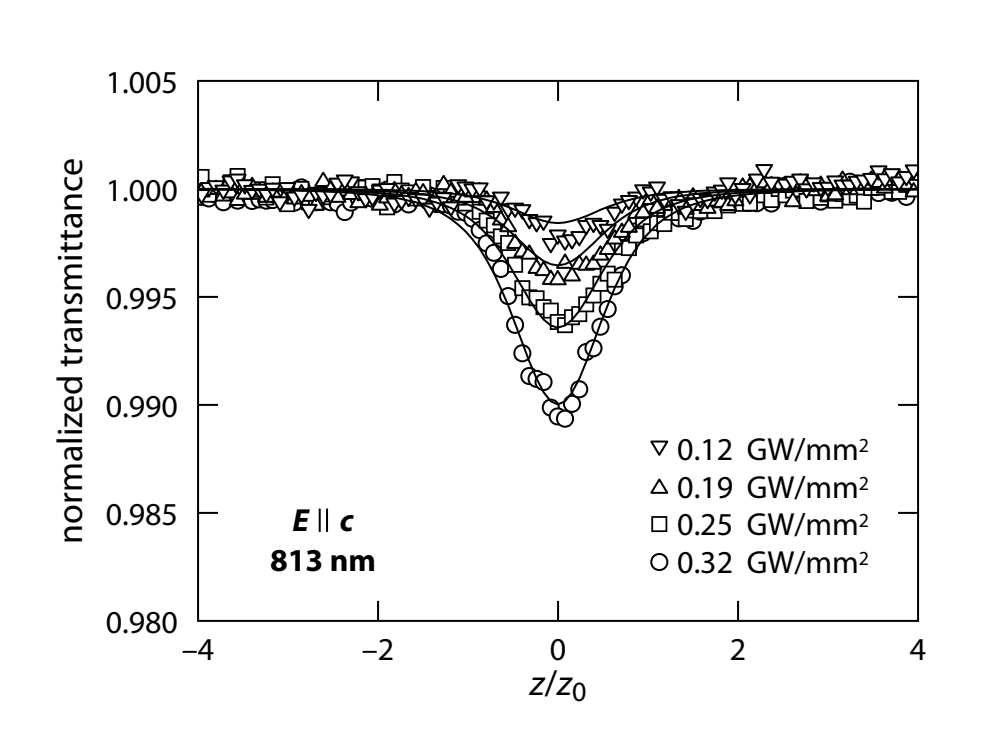


Analysis

$E \| c$

$2\hbar\omega = 3.2$ eV 3.1 eV $2\hbar\omega = 3.05$ eV





Analysis

Fitting Results

		extraordinary polarization ($E \parallel c$)	
λ_0 (nm)	$2\hbar\omega$ (eV)	β (mm/GW)	γ (mm^3/GW^2)
774	3.20	0.89	1.8
813	3.05	$<10^{-7}$	0.9
		±15%	factor of 2

Analysis

Fitting Results

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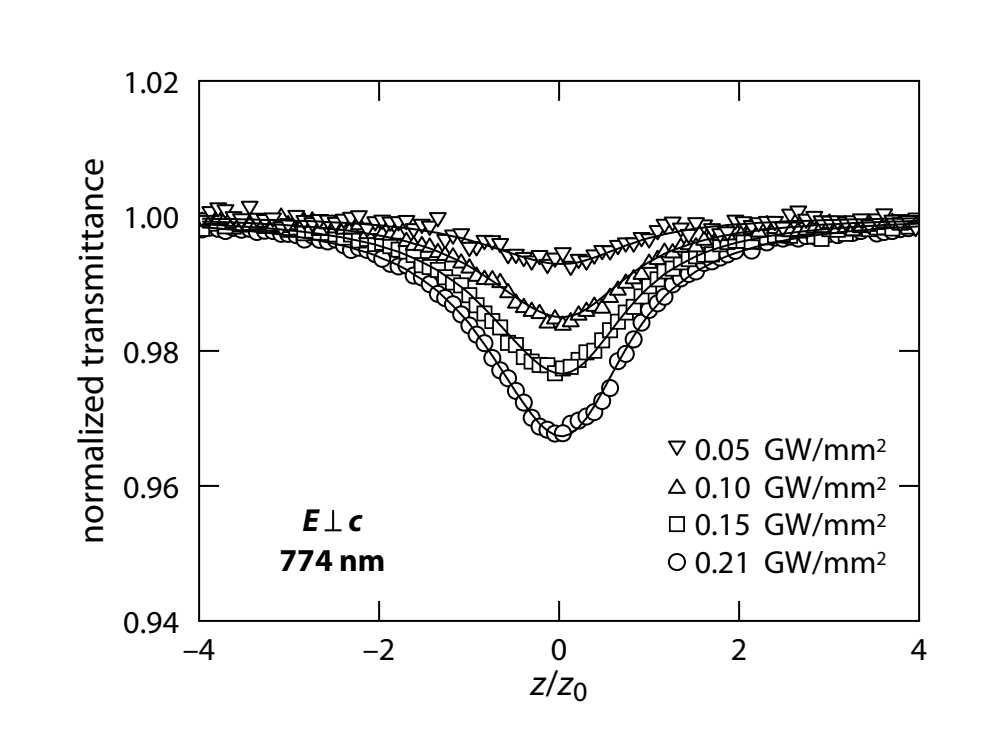
λ_0 (nm)	$2\hbar\omega$ (eV)	β (mm/GW)	γ (mm^3/GW^2)
774	3.20	0.89	1.8
813	3.05	$<10^{-7}$	0.9
		±15%	factor of 2

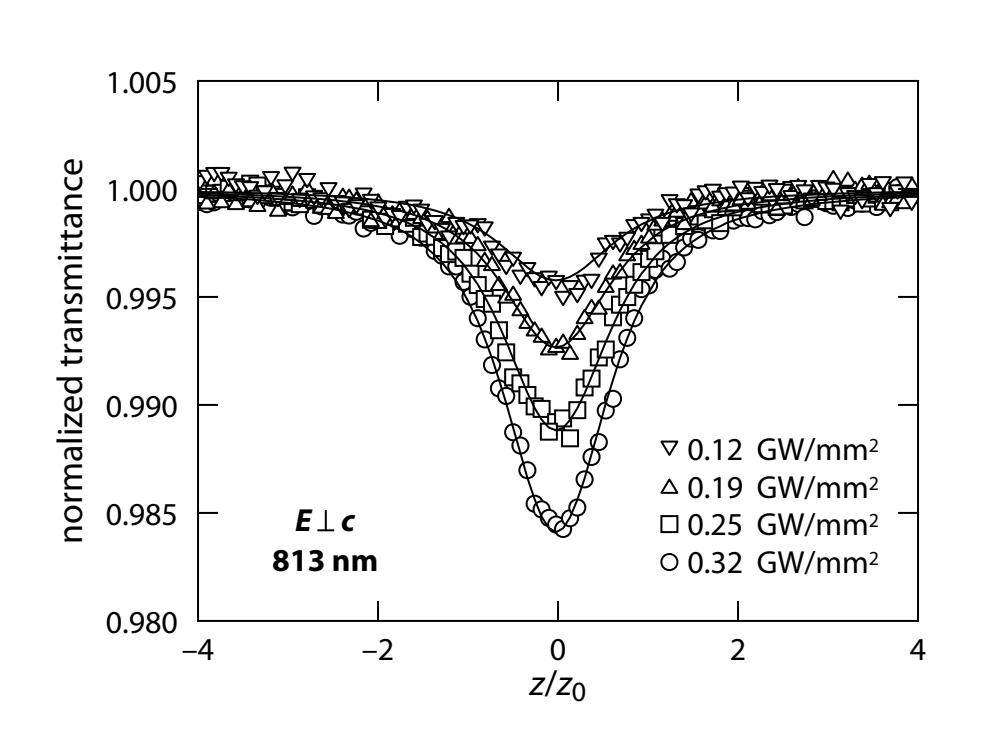
We can see the indirect bandgap at 3.1 eV

Analysis

$E \perp c$

$2\hbar\omega = 3.2$ eV 3.1 eV $2\hbar\omega = 3.05$ eV





Analysis

Fitting Results

		ordinary polarization ($E \perp c$)	
λ_0 (nm)	$2\hbar\omega$ (eV)	β (mm/GW)	γ (mm^3/GW^2)
774	3.20	0.54	1.1
813	3.05	0.08	0.8
		±15%	factor of 2

Direct forbidden bandgap at 3.062 eV

K. Watanabe and K. Inoue, Phys. Rev. B 41, 7957-7960 (1990).

Analysis

Fitting Results

		extraordinary polarization ($E \parallel c$)		
λ_0 (nm)	$2\hbar\omega$ (eV)	β (mm/GW)	γ (mm^3/GW^2)	$\frac{n_2}{\beta\lambda}$
774	3.20	0.89	1.8	1.1
813	3.05	$<10^{-7}$	0.9	$>10^6$
		±15%	factor of 2	

Analysis

Fitting Results

		ordinary polarization ($E \perp c$)		
λ_0 (nm)	$2\hbar\omega$ (eV)	β (mm/GW)	γ (mm^3/GW^2)	$\frac{n_2}{\beta\lambda}$
774	3.20	0.54	1.1	1.9
813	3.05	0.08	0.8	12.1
		±15%	factor of 2	

Analysis

Fitting Results

		extraordinary polarization ($E \parallel c$)			
λ_0 (nm)	$2\hbar\omega$ (eV)	β (mm/GW)	γ (mm^3/GW^2)	$\frac{n_2}{\beta\lambda}$	$I_{\max}^{3PA}$ (GW/mm^2)
774	3.20	0.89	1.8	1.1	0.57
813	3.05	$<10^{-7}$	0.9	$>10^6$	1.10
		±15%	factor of 2		

Analysis

Fitting Results

		ordinary polarization ($E \perp c$)			
λ_0 (nm)	$2\hbar\omega$ (eV)	β (mm/GW)	γ (mm^3/GW^2)	$\frac{n_2}{\beta\lambda}$	$I_{\max}^{3PA}$ (GW/mm^2)
774	3.20	0.54	1.1	1.9	0.93
813	3.05	0.08	0.8	12.1	1.20
		±15%	factor of 2		

Conclusions

Two- and three-photon absorption in rutile

Two-photon absorption is below 1 mm/GW

Three-photon absorption is below 2 mm^3/GW^2

Negligible 2PA at 813 nm for $E \| c$

Conclusions

Two-photon figures of merit greater than 12 for λ_0>813 nm.

Three-photon absorption limits peak intensity to ~0.50 GW/mm^2.

Rutile is a promising for all-optical switching for wavelengths greater than 800 nm.

Funding provided by:

Chris Evans

Jon Bradley

Erwin Martí

Eric Mazur

Any questions?

C. C. Evans, J. D. B. Bradley, E. A. Martí-Panameño, and E. Mazur,
"Mixed two- and three-photon absorption in bulk rutile (TiO_2) around 800 nm,"
Opt. Express 20, 3118-3128 (2012).

Chris Evans

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