



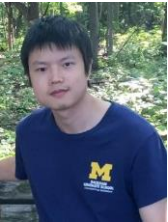



# Diamond – Extreme Material for Nonlinear Optics and Raman Lasers

Presented by:



## Technical Group Leadership



 <p>Fatima Toor University of Iowa Chair</p>	 <p>David James Spence Macquarie University Vice Co-Chair</p>	 <p>Tong Zhou Thorlabs Inc Vice Co-Chair</p>
 <p>Shamsul Arafin UCSB Events and Social Media Officer</p>	 <p>Austin Cyphersmith, UIUC, Webinar and Social Media Officer</p>	 <p>Kaikai Xu Univ. Electro Sci &amp; Tech of China Vice Co-Chair</p>



## Laser Systems (PL)

### Get Involved

#### Technical Divisions +

Bio-Medical Optics

Fabrication, Design & Instrumentation

Information Acquisition, Processing & Display

Optical Interaction Science

#### Photonics and Opto-Electronics +

Fiber Optics Technology (PF)

Integrated Optics (PI)

Laser Systems (PL)

Optical Communications (PC)

Optics for Energy (PS)

Optoelectronics (PO)

Photonic Detection (PD)

Vision and Color Division

Technical Group Newsletter

Technical Group Webinars

#### OSA CONNECT

Minorities and Women in OSA (MWOSA)

Diversity & Inclusion in OSA

Young Professionals

Corporate Members

Optics & Photonics Regions

## Laser Systems (PL)

This group encompasses novel laser system development for a broad range of scientific, industrial, medical, remote sensing and other directed-energy applications. The group addresses technical issues concerning sources that cover the full spectral range, including: ultraviolet, visible, infrared, terahertz and microwave. Strong overlap with other technical groups that study and develop laser techniques and technologies brings together researchers and engineers to produce sources with unique performance, such as high-power, ultra-short pulses and high coherence.

GROUP LEADERSHIP	UPCOMING MEETINGS	RECENTLY PUBLISHED
<b>Name</b>	<b>Affiliation</b>	<b>Title</b>
Fatima Toor	University of Iowa	Chair
Muhammad Faryad	Lahore University of Management Sciences	Conference Events Officer
Shamsul Arafin	University of California Santa Barbara	Events and Social Media Officer
David James Spence	Macquarie University	Vice Co-Chair
Tong Zhou	Thorlabs Inc	Vice Co-Chair
Austin Joseph Cyphersmith	University of Illinois Urbana-Champaign	Webinar and Social Media Officer

## Announcements

Register now for the Laser Systems Technical Group's upcoming webinar on III-nitride nanowire light-emitting diodes grown by molecular beam epitaxy. The webinar, featuring a presentation from Dr. Hieu Nguyen of the New Jersey Institute of Technology, will be held on 27 July 2016 at 12:00 EDT.

[Register for the webinar now >>](#)

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## Work in Optics

SYSTEM INTEGRATION ENGINEER | 95134  
Thu, 22 Sep 2016 18:15:00 EST

Design Engineer - Laser Products | Thorlabs  
Fri, 16 Sep 2016 12:47:00 EST

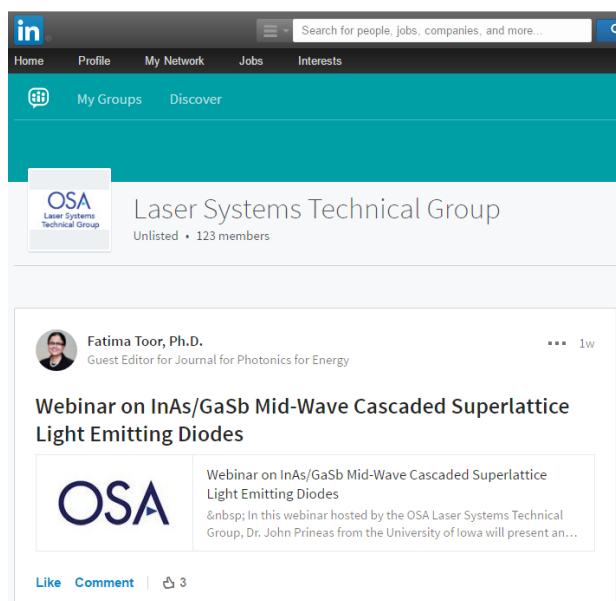
Advanced Development Scientist | Block Engineering  
Thu, 15 Sep 2016 15:09:00 EST

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## Contact your Technical Group and Get Involved!

- LinkedIn site (global reach)
- Announce new activities
- Promote interactions
- Complement the OSA  
Technical Group Member List



Welcome to Today's webinar!



### Diamond - Extreme Material for Nonlinear Optics and Raman Lasers



Overview
Speaker(s)
Register



Diamond is an optical material with a suite of properties that are set well apart from most others. Its recent availability in large, high quality, synthesized crystals is bringing forth important opportunities to tackle major challenges in optics. In this webinar hosted by the OSA Laser Systems Technical Group, Dr. Richard Mildren of Macquarie University will provide an introduction to the optical properties of diamond (linear, nonlinear and thermo-optical). Dr. Mildren will then present a detailed description about some of diamond's more outstanding properties and how these are stimulating development of advanced laser-based technologies, such as high power lasers and nonlinear frequency converters.

**What You Will Learn in the Webinar:**

- Detailed understanding of the contrasting nature of diamond against more familiar optical materials
- Design issues for developing diamond optical devices and in particular diamond lasers
- Features of diamond that are likely to impact on the future development of optics

**Who Should Attend the Webinar:**

- Research Students
- Researchers
- Professionals in Industry, Government

Richard Mildren, Physics and Astronomy, Macquarie University, Australia

<https://research.science.mq.edu.au/diamond/>

# Diamond - Extreme material for nonlinear optics and Raman lasers

Rich Mildren

[rich.mildren@mq.edu.au](mailto:rich.mildren@mq.edu.au)

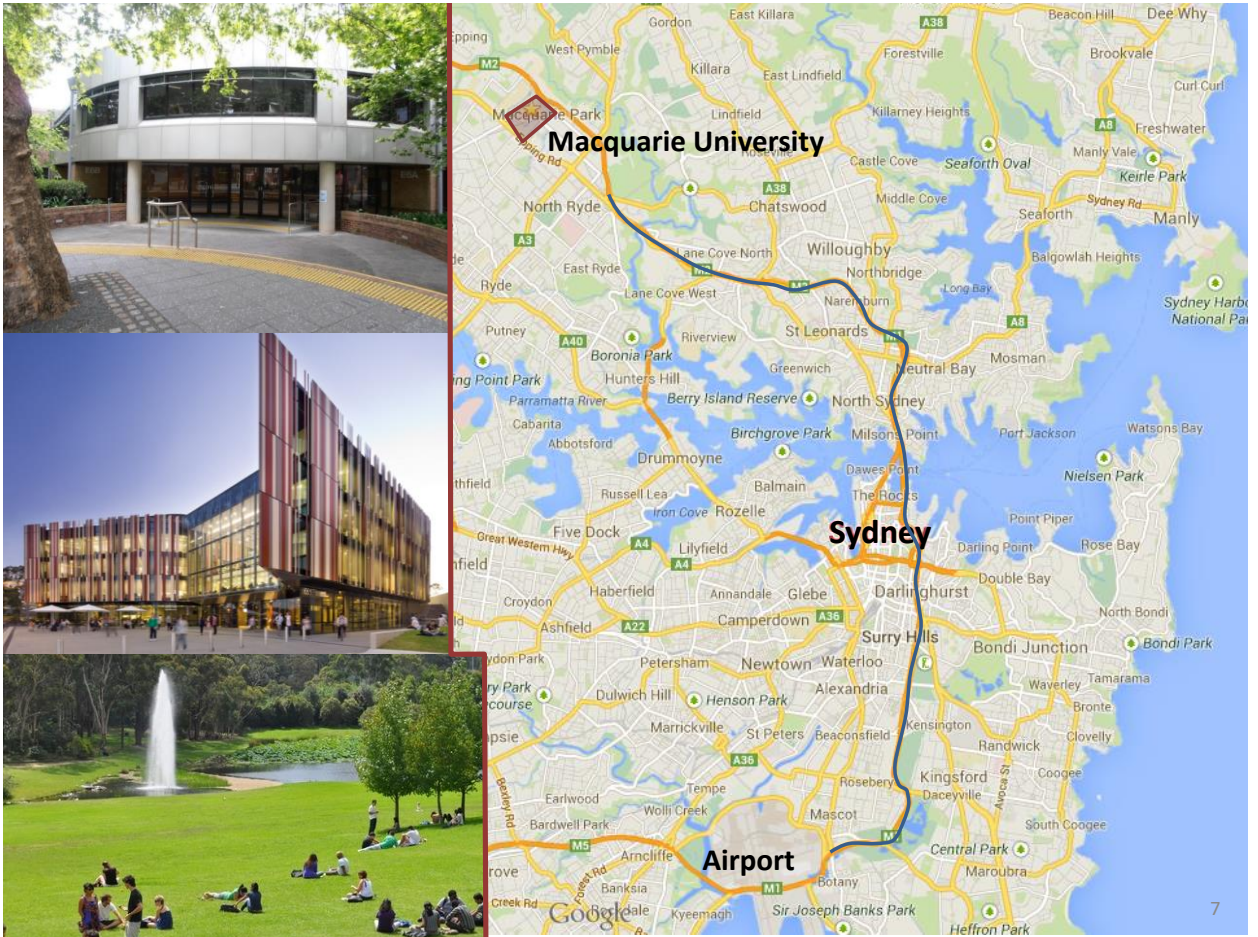
<http://web.science.mq.edu.au/groups/diamond/>

MQ Photonics Research Centre  
Department of Physics and Astronomy  
Macquarie University, **Sydney, Australia**



MACQUARIE UNIVERSITY

**MQ Photonics**



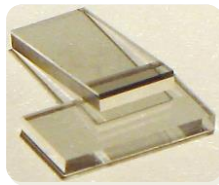
# Diamond technologies

Cutting  
and  
abrasives

Electrochemistry

Gemstones

Electronics,  
LEDs and  
radiation  
detectors



Linear and  
nonlinear optics

- *Windows, diffractive elements, waveguides, heat-spreaders*
- *Four-wave mixing*
- *Raman lasers*
- *Nanophotonics*

Thermal  
Management

Colour Centres

- *Quantum*
- *Magnetometry / Sensing*
- *Bio-imaging*

Opto-  
mechanics



## Contents –

### 1. Optical Properties

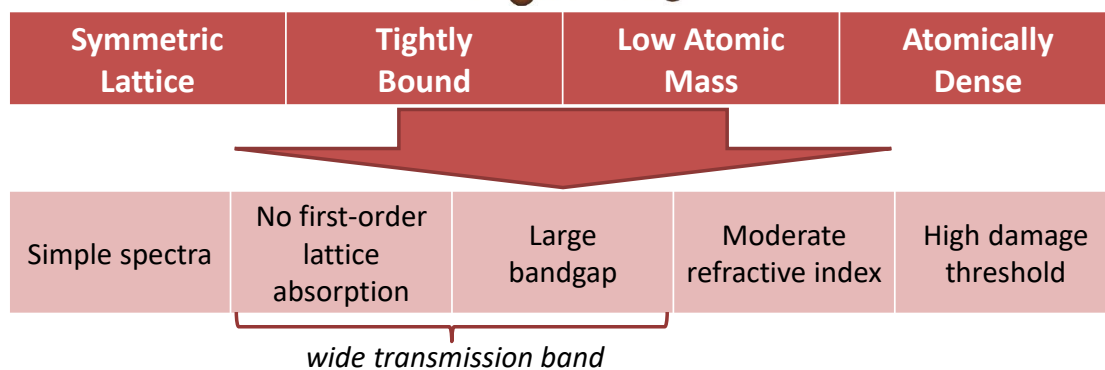
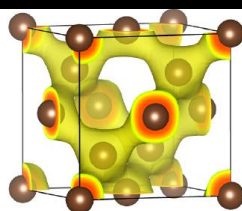
- Linear properties
- Scattering
- Nonlinear properties
- Thermo-optical

### 2. Properties Summary

*“The good, the bad and the ugly”*

### 3. Applications in Raman lasers

## Origins ...



Detailed optical properties depend on subtle effects



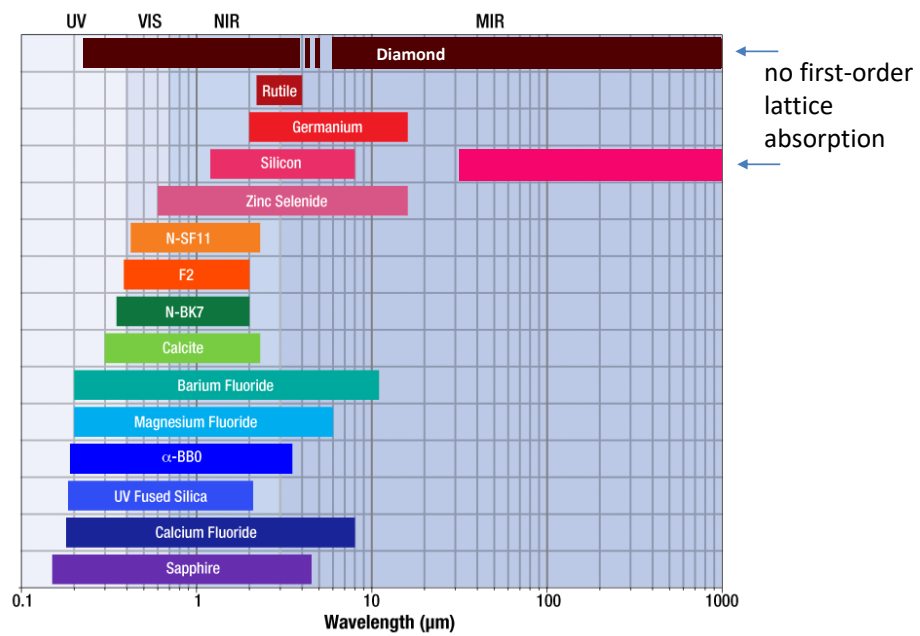
- Electronic band structure
- Phonon dispersion
- Zero point energy
- Anharmonicity
- Surface effects



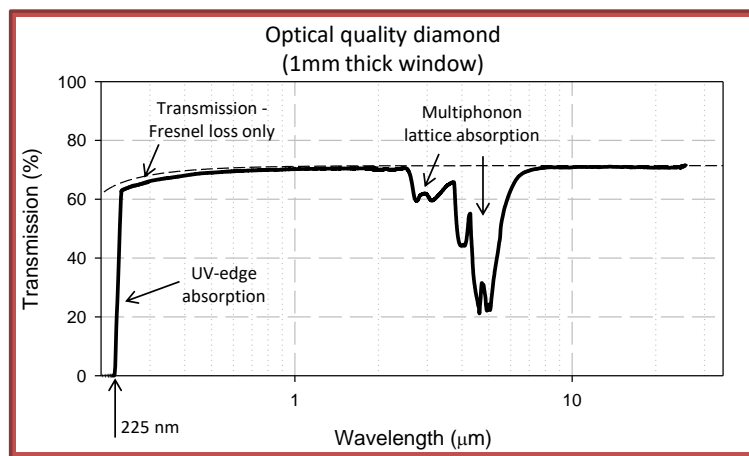
**DEEP PHYSICS**  
~  
**MANY POSSIBILITIES**

# Transmission Range

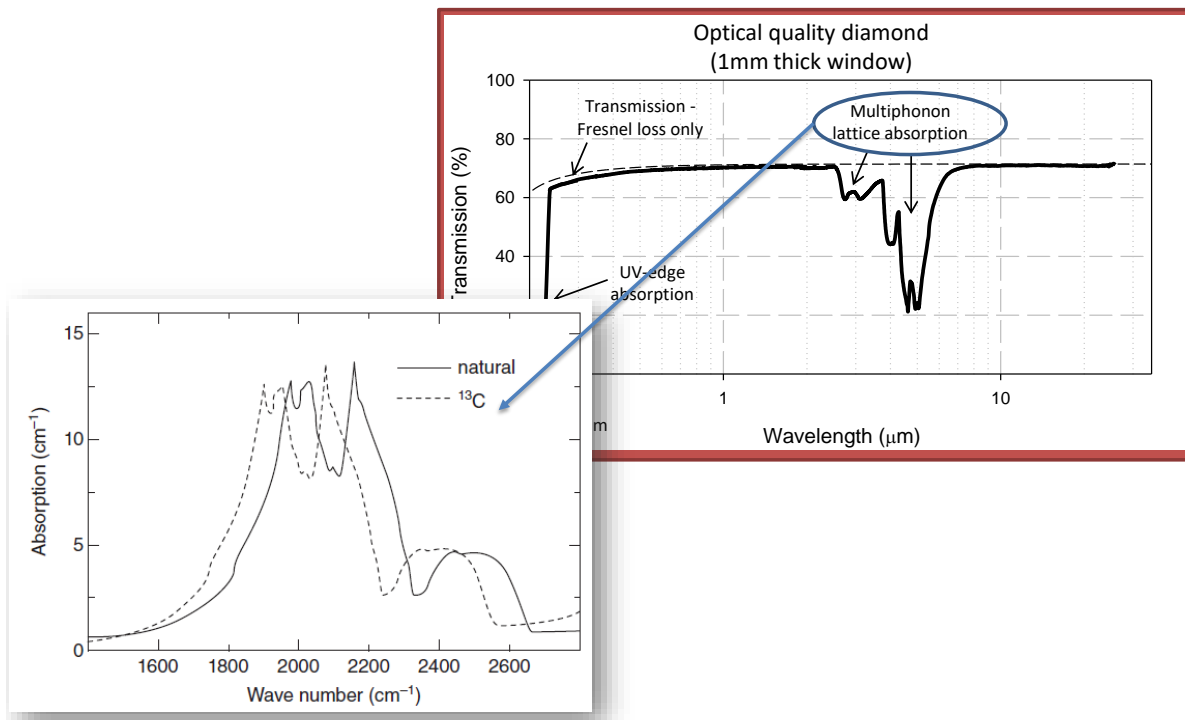
versus other optical materials



# Transmission / Absorption

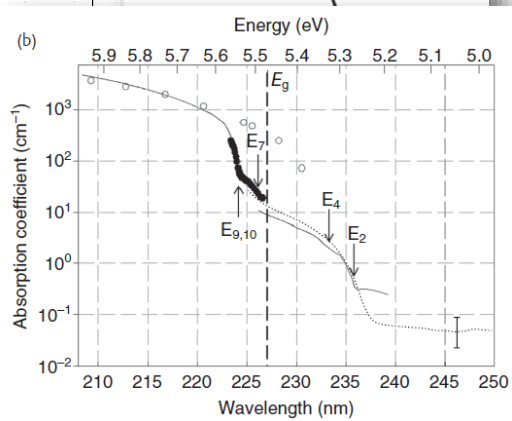
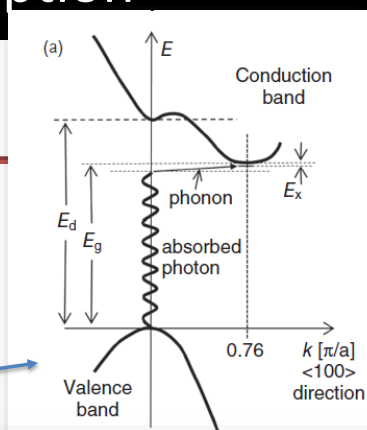
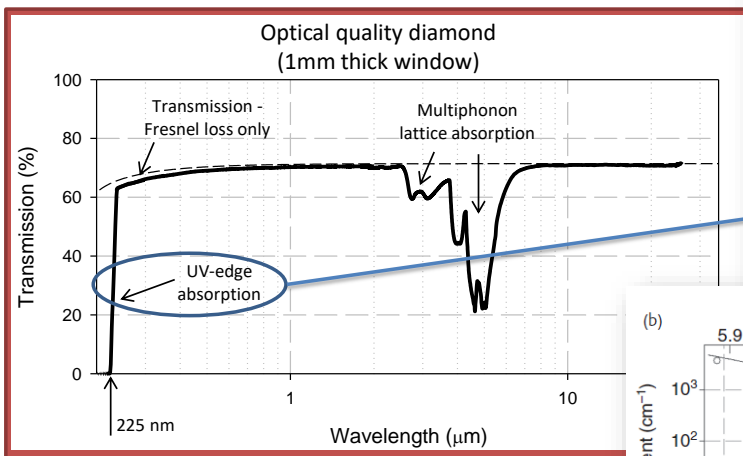


# Transmission / Absorption

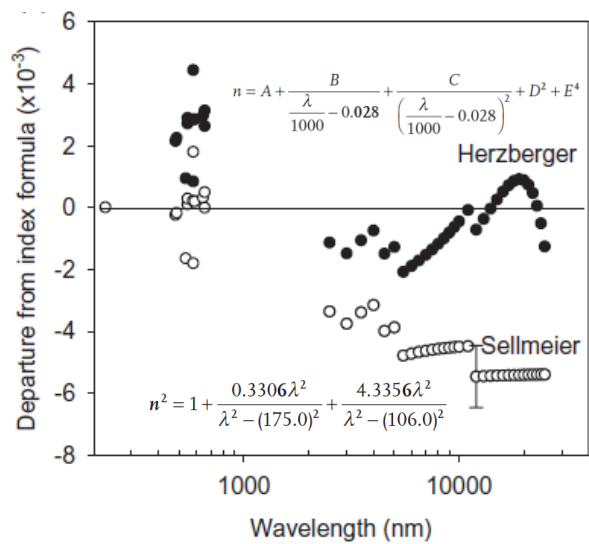
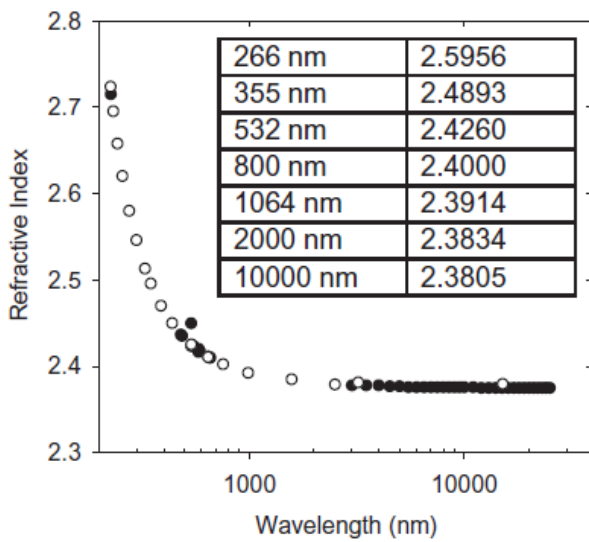


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# Transmission / Absorption

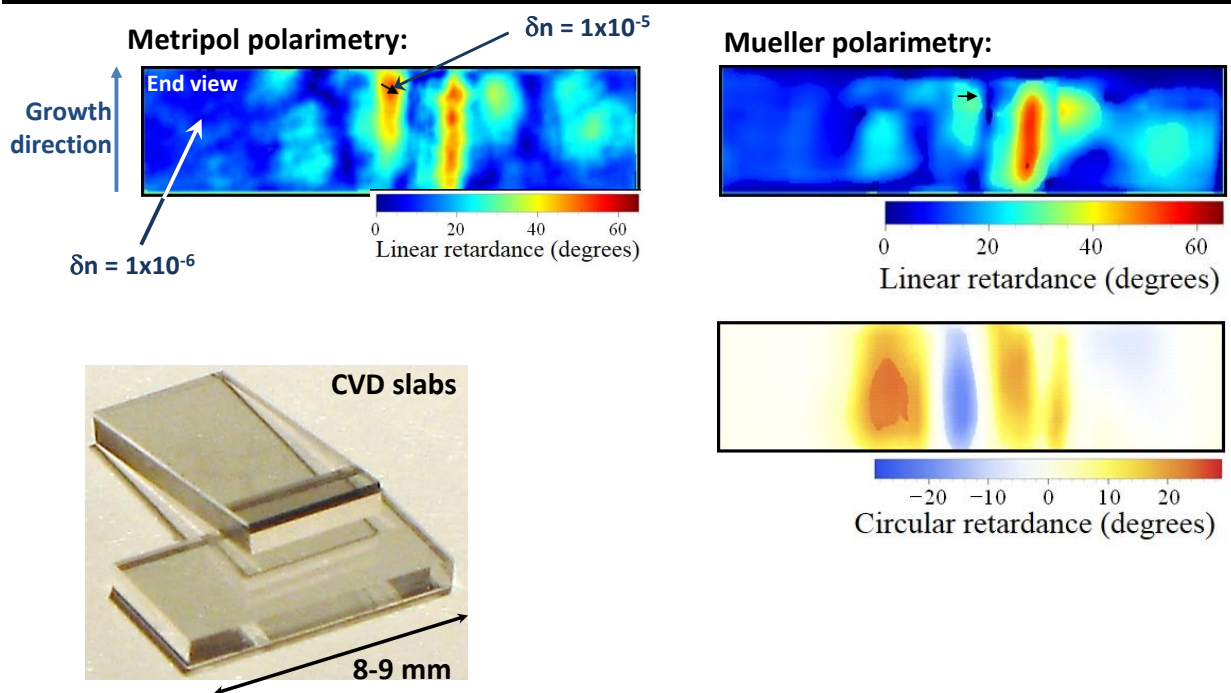


# Refractive index



F. Peter, Z. Phys A Hadrons Nucl. **15** (1923)  
 D.F. Edwards and E. Ochoa JOSA **71**, (1981)

## Birefringence in 'laser grade' material

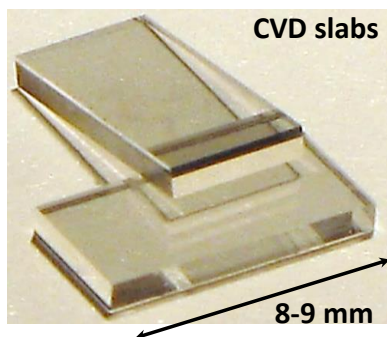
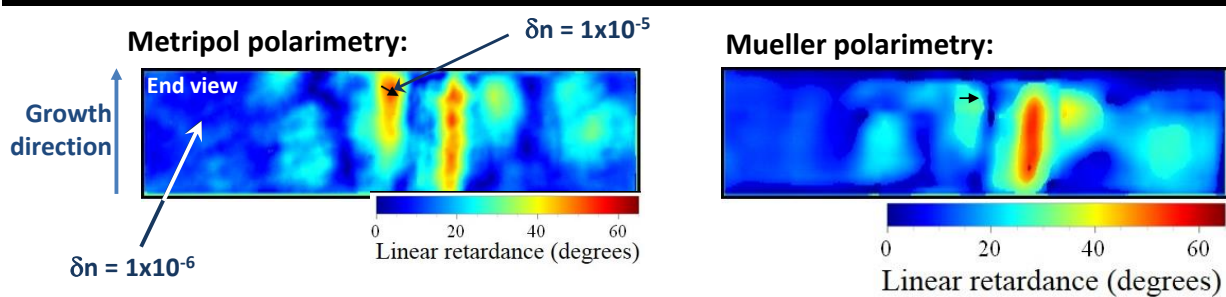


H. Jasbeer, R. Williams, O. Kitzler, A. McKay, S. Sarang R. Mildren, *JOSAB*, 33 p56, (2016)

16

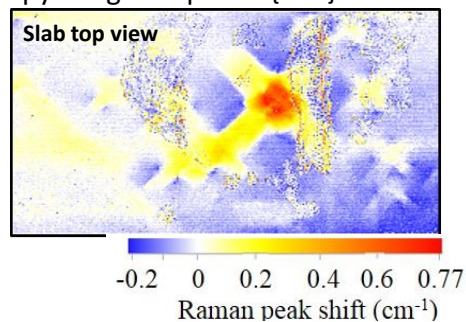


## Birefringence in 'laser grade' material



Raman microscopy image: top face {100}

centre Raman  
frequency:  
 $\omega_0 = 1332.3 \text{ cm}^{-1}$

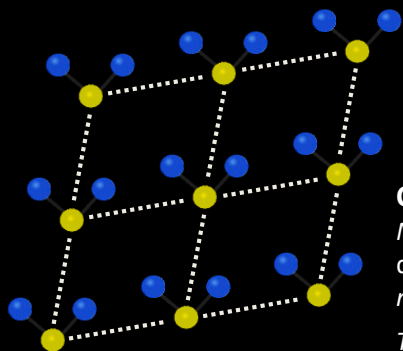


H. Jasbeer, R. Williams, O. Kitzler, A. McKay, S. Sarang R. Mildren, *JOSAB*, 33 p56, (2016)<sup>17</sup>

# Scattering – Raman

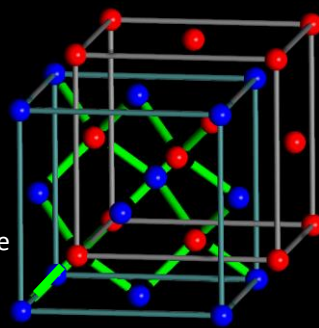
molecular crystals

diamond, silicon, germanium



### Critical variables:

$N$  density of scatterers  
 $d\alpha/dq$  polarizability derivative  
 $m$  reduced mass  
 $T_2$  dephasing time



Differential cross-section

$$\frac{d\sigma}{d\Omega} = k \cdot \frac{\omega_s^4}{\omega_R m} \left( \frac{d\alpha}{dq} \right)^2$$

$$k = \frac{\mu_s h}{4\pi\mu_L c^4}$$

Raman gain coefficient

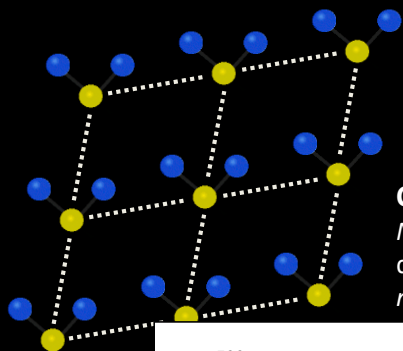
$$g_R = k' \frac{T_2 N}{\omega_s^3} \left( \frac{d\sigma}{d\Omega} \right)$$

$$k' = \frac{4\pi^2}{\mu_L \mu_s c^2}$$

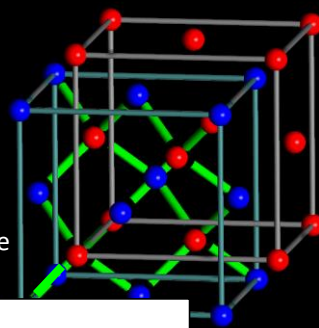
# Scattering – Raman

molecular crystals

diamond, silicon, germanium

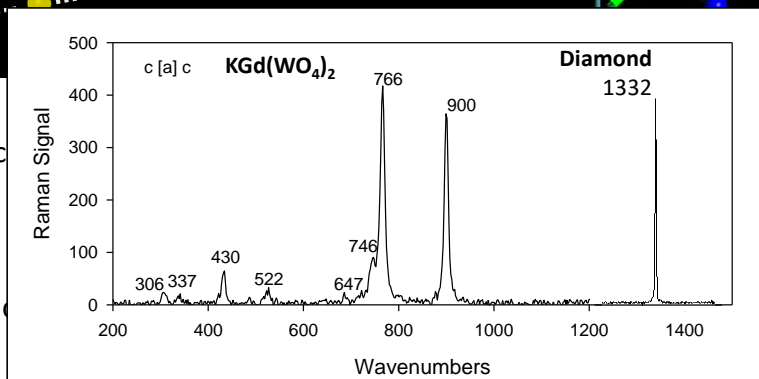


**Critical variables:**  
*N* density of scatterers  
 $d\alpha/dq$  polarizability derivative  
*m* reduced mass



Differential c

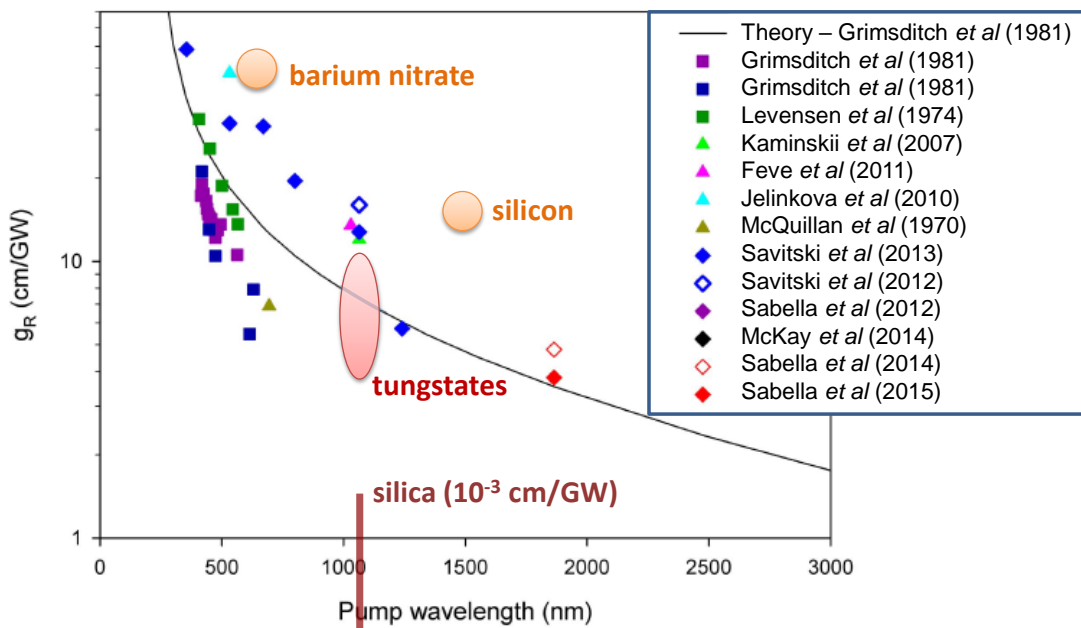
Raman gain



$$\frac{\mu_s h}{4\pi\mu_L c^4}$$

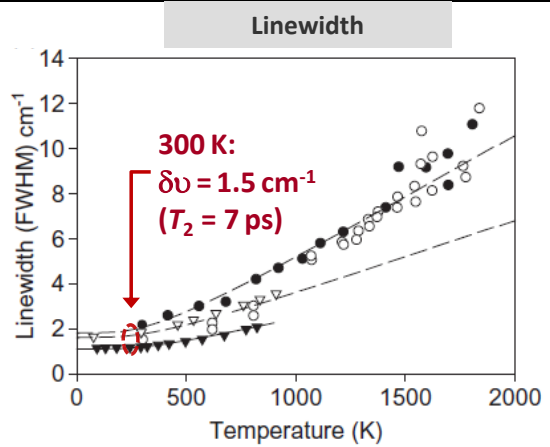
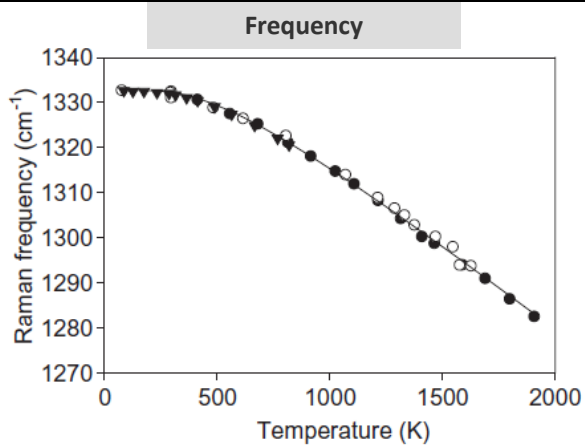
$$\frac{4\pi^2}{\mu_L \mu_s c^2}$$

# Wavelength dependence of Raman gain



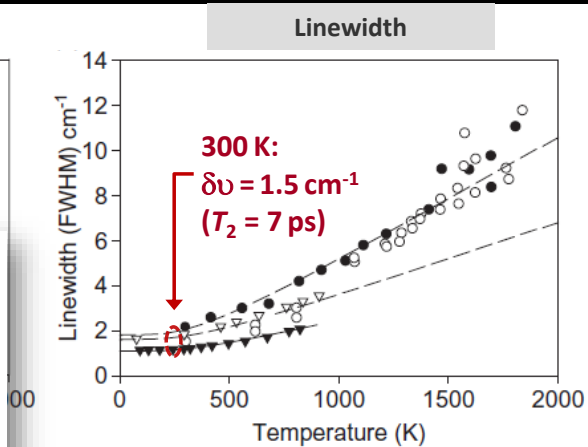
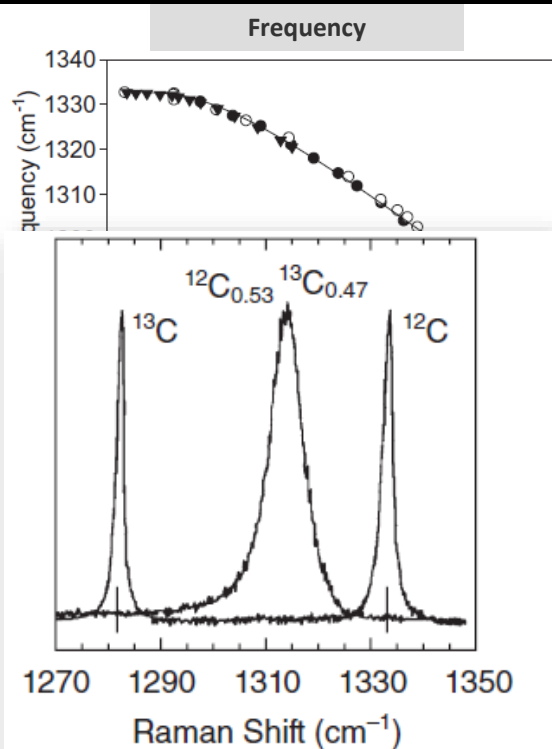
Sabella, Spence, Mildren, J Quantum Electron. IEEE JQE vol. 51, 1000108 (2015) 20

# Raman: Temperature and isotopic Purity



barium nitrate	silicon	diamond
$T_2 = 26 \text{ ps}$	$\sim 6 \text{ ps}$	$7 \text{ ps}$

# Raman: Temperature and Isotopic Purity

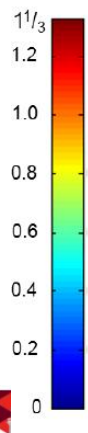
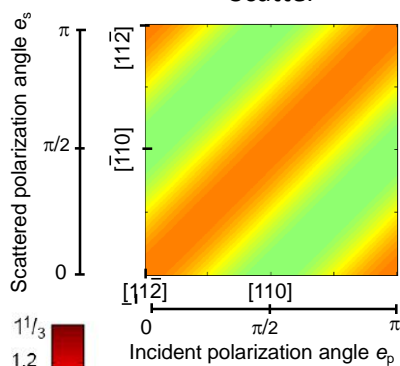


barium nitrate	silicon	diamond
$T_2 = 26 \text{ ps}$	$\sim 6 \text{ ps}$	$7 \text{ ps}$

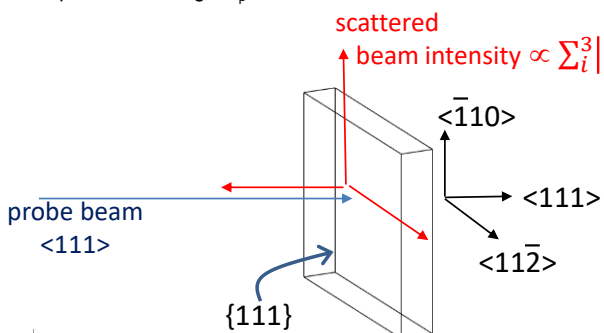
Cardona & Ruf,  
 Solid State Commun. 117 (2001)

# Raman: Polarization dependence

For / Back Scatter



UNIVERSITY  
otonics



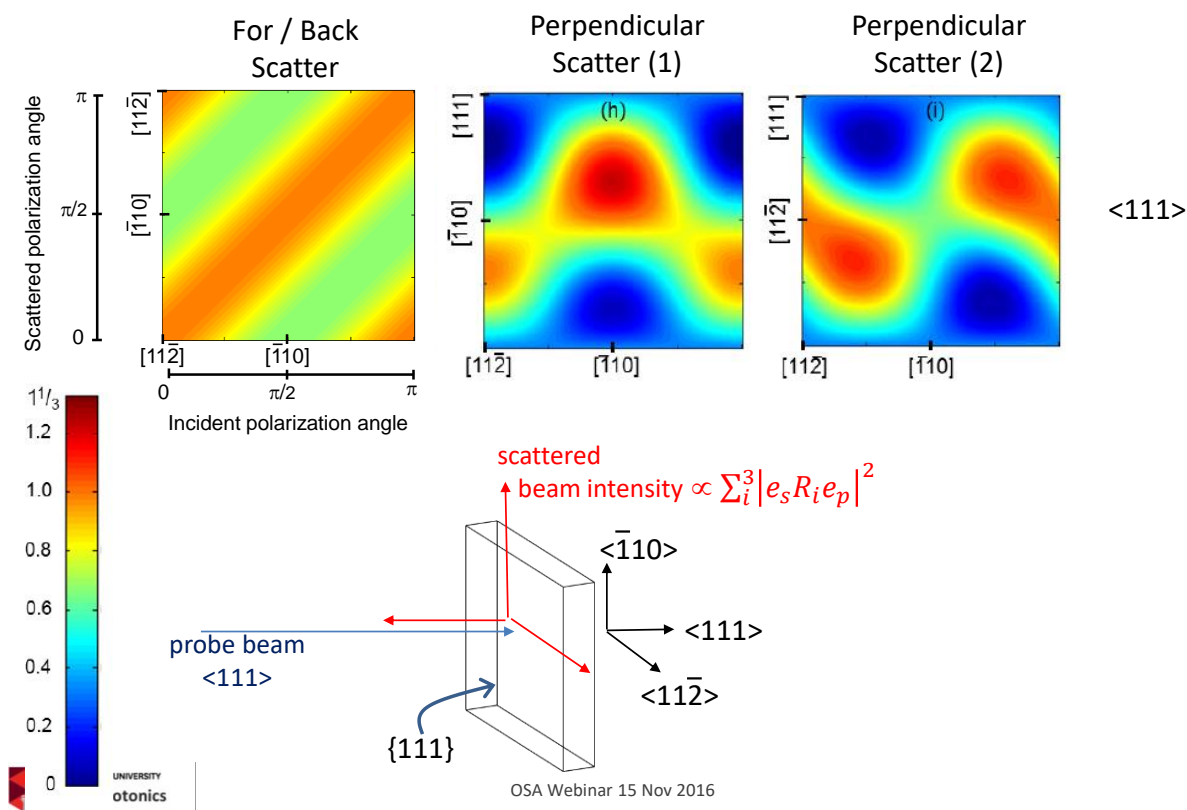
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$$R_1 = \begin{bmatrix} 0 & \frac{d}{\sqrt{3}} & \frac{d}{\sqrt{6}} \\ \frac{d}{\sqrt{3}} & \frac{2d}{3} & -\frac{d}{3\sqrt{2}} \\ \frac{d}{\sqrt{6}} & -\frac{d}{3\sqrt{2}} & -\frac{2d}{3} \end{bmatrix}$$

$$R_2 = \begin{bmatrix} 0 & \frac{d}{\sqrt{3}} & \frac{d}{\sqrt{6}} \\ \frac{d}{\sqrt{3}} & -\frac{2d}{3} & \frac{d}{3\sqrt{2}} \\ \frac{d}{\sqrt{6}} & \frac{d}{3\sqrt{2}} & \frac{2d}{3} \end{bmatrix}$$

$$R_3 = \begin{bmatrix} d & 0 & 0 \\ 0 & -\frac{d}{3} & \frac{\sqrt{2}d}{3} \\ 0 & \frac{\sqrt{2}d}{3} & -\frac{2d}{3} \end{bmatrix}$$

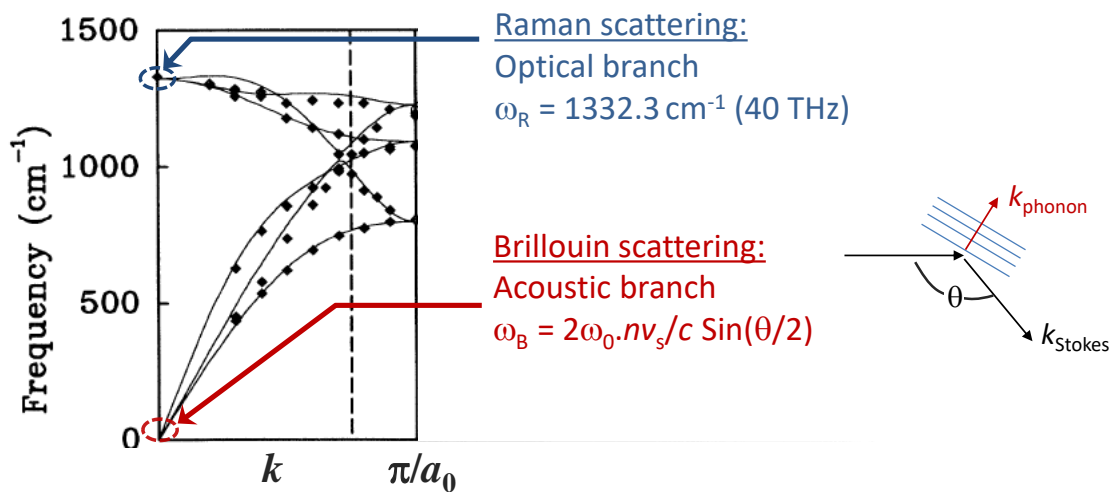
# Polarization dependence





# Brillouin scattering

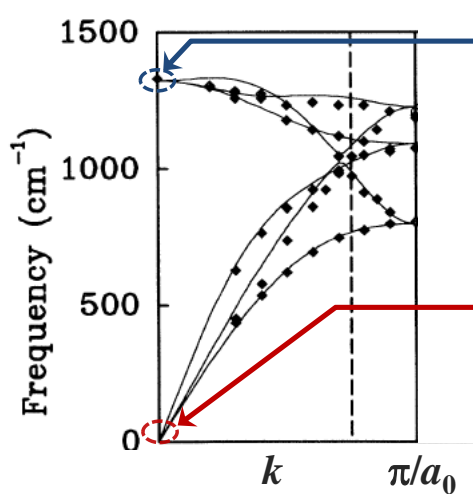
**Optical and Acoustic Phonon Branches** [Pavone *et al*, PRB, 48 3156 (2003)]



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# Brillouin scattering

**Optical and Acoustic Phonon Branches** [Pavone *et al*, PRB, 48 3156 (2003)]



Raman scattering:

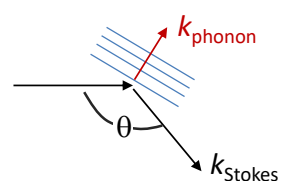
Optical branch

$$\omega_R = 1332.3 \text{ cm}^{-1} \text{ (40 THz)}$$

Brillouin scattering:

Acoustic branch

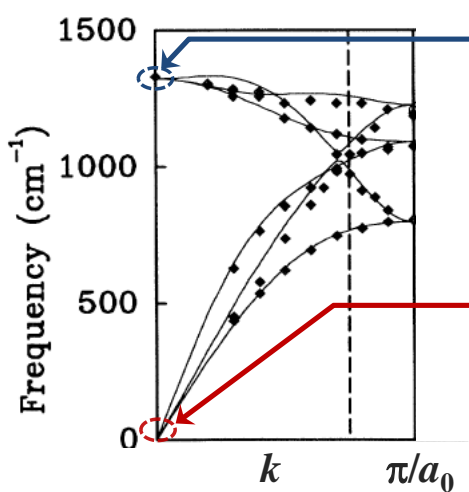
$$\omega_B = 2\omega_0 \cdot n v_s / c \sin(\theta/2)$$



	Glass	Silicon	Diamond
Speed of sound $v_s$ (Longitudinal) [ $\text{m s}^{-1}$ ]	4000- 6000	5840	18350
$\omega_B$ [GHz] (1500 nm)	8 - 12	26	57

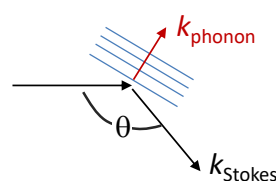
# Brillouin scattering

**Optical and Acoustic Phonon Branches** [Pavone *et al*, PRB, 48 3156 (2003)]



Raman scattering:  
Optical branch  
 $\omega_R = 1332.3 \text{ cm}^{-1}$  (40 THz)

Brillouin scattering:  
Acoustic branch  
 $\omega_B = 2\omega_0 \cdot n v_s / c \sin(\theta/2)$



Scattering efficiency  
 $S \sim \rho^2 n^8$

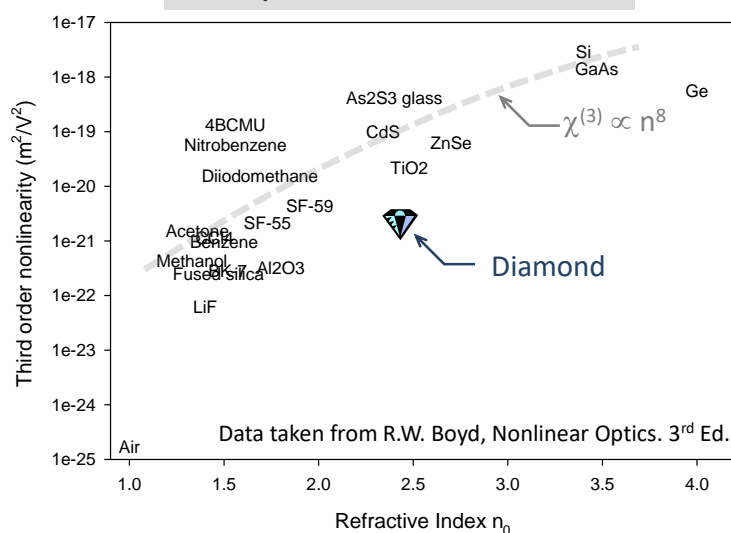
Acousto-optic  
FoM  $\sim \rho^2 n^6 / v_s^3$

SBS gain coefficient  
 $g_B \sim \rho^2 n^7 T_2$

	Glass	Silicon	Diamond
Speed of sound $v_s$ (Longitudinal) [ $\text{m s}^{-1}$ ]	4000-6000	5840	18350
$\omega_B$ [GHz] (1500 nm)	8 - 12	26	57
$\rho_{11}$	0.13	-0.17	-0.25
$\rho_{12}$	0.21	-0.05	0.04
$\rho_{44}$	-0.04	-0.05	-0.17

# Kerr nonlinearity

Comparison with other materials



## Low Kerr nonlinearity:

- High critical power for self-focusing = 1-2 MW
- Low self-phase modulation
- Low four wave-mixing gain

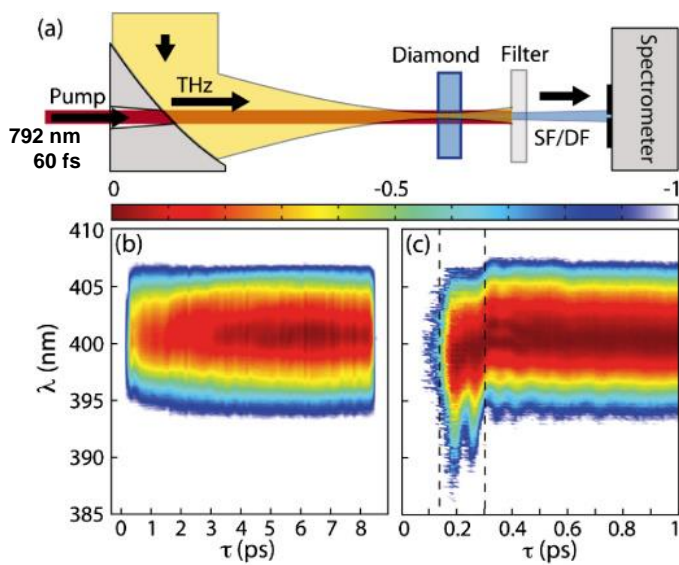
Sheik-Bahae *et al*, Proc. SPIE, 1624, (1992)

Sheik-Bahae *et al* Proc. SPIE, 2428, (1995)

Zhao *et al*, Chin. Opt. Lett., 8 (2010)

Kozák *et al*, JOSAB, 142 (2012)

## Example: Four wave mixing in diamond

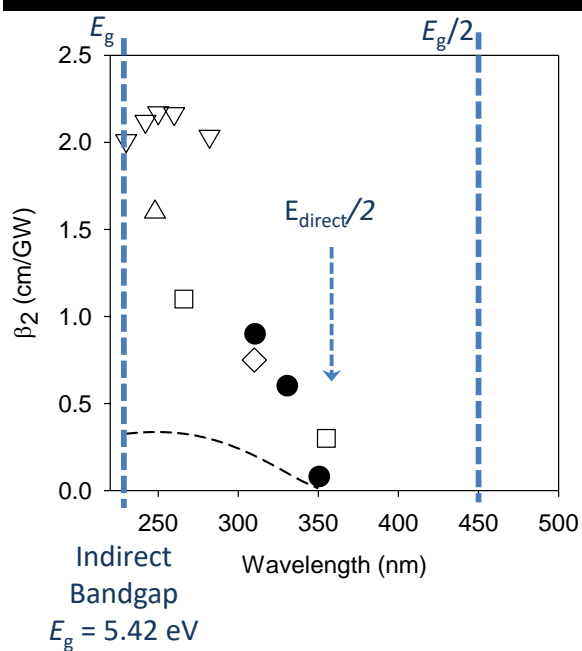


$$\omega_{\text{output}} = 2\omega_p \pm \omega_{\text{THz}}$$

Phase-matching achieved for co- and counter-propagating THz beams

Clerici, et al, *Opt. Lett.* 38, 178-180 (2013)

## Two-photon absorption

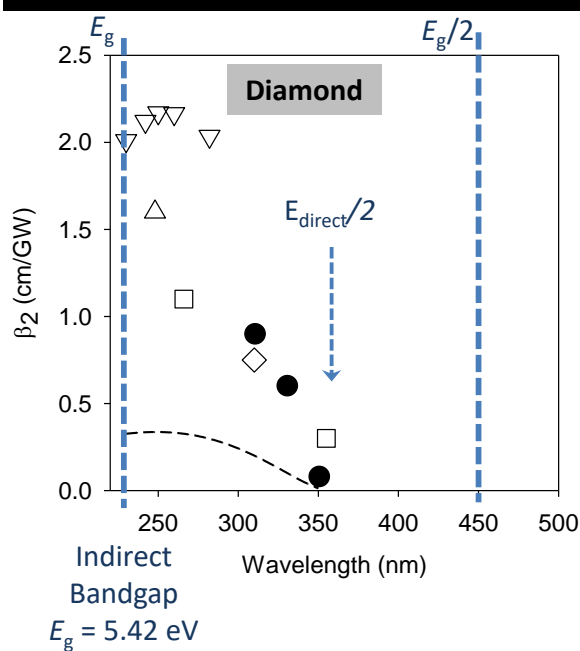


Sheik-Bahae *et al*, Proc. SPIE, **1624**, (1992)  
 Sheik-Bahae *et al* Proc. SPIE, **2428**, (1995)  
 Kozák *et al*, JOSAB, **142** (2012)  
 Preuss and Stuke, Appl. Phys. Lett. , **67** (1995)  
 Gagarskii and Prikhodko J. Opt. Technol., **75** (2008)

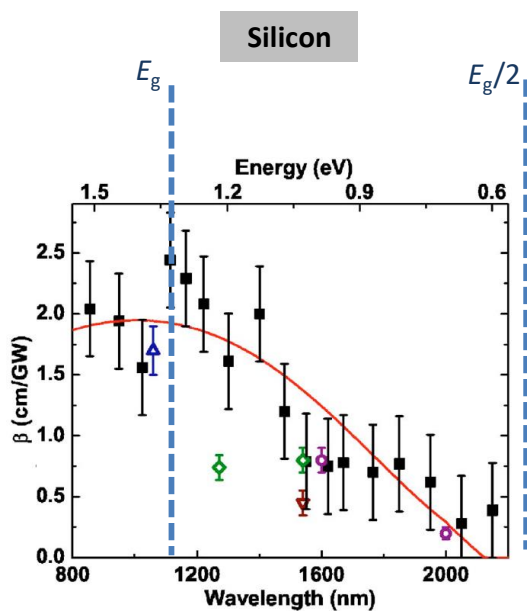
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## Two-photon absorption



Sheik-Bahae *et al.*, Proc. SPIE, **1624**, (1992)  
 Sheik-Bahae *et al.* Proc. SPIE, **2428**, (1995)  
 Kozák *et al.*, JOSAB, **142** (2012)  
 Preuss and Stuke, Appl. Phys. Lett., **67** (1995)  
 Gagarskii and Prikhodko J. Opt. Technol., **75** (2008)

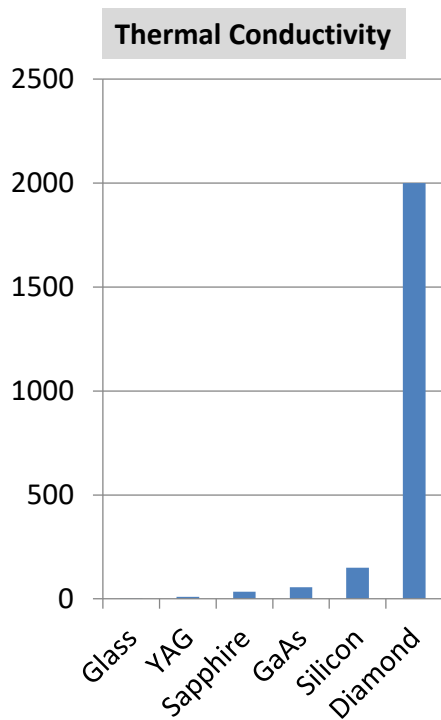


Bristow, *Appl. Phys. Lett.*, **90** 191104 (2004)

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## Thermo-optic properties



**Thermal conductivity**  
**Thermal expansion**  
**Thermo-optic coeff.**

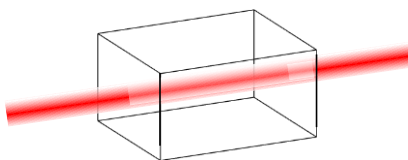
$\kappa = 2000 \text{ W/m.K}$   
 $\alpha_T = 1.1 \times 10^{-6} / \text{K}$   
 $dn/dT = 16 \times 10^{-6} / \text{K}$



## Thermally-induced lens

$$f^{-1} = \frac{P_{\text{dep}}}{2\pi\kappa\omega_0^2} \left( \frac{dn}{dT} + (n-1)(\nu+1)\alpha_T + n^3\alpha_T C_{r,\phi} \right)$$

*Lens strength* = *Thermo-optic* + *End-face distortion* + *Stress Birefringence*



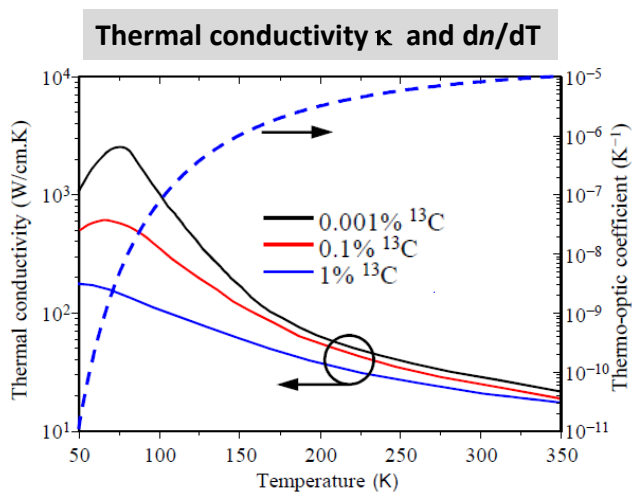
$\tau \sim 10^{-5} \text{ s} !!$

	Key Parameter	Diamond : YAG
Stress fracture limit	$\kappa/\alpha_T$	1100:1
Stress birefringence	$\kappa/\alpha_T$	1100:1
Crystal distortion	$\kappa/\alpha_T$	1100:1
Thermo-optic lens	$\kappa/ \frac{dn}{dT}$	85:1

Thermal conductivity

Thermal expansion coefficient

# Cryo Cooling: Natural and Enriched diamond



At  $T = 100$  K:

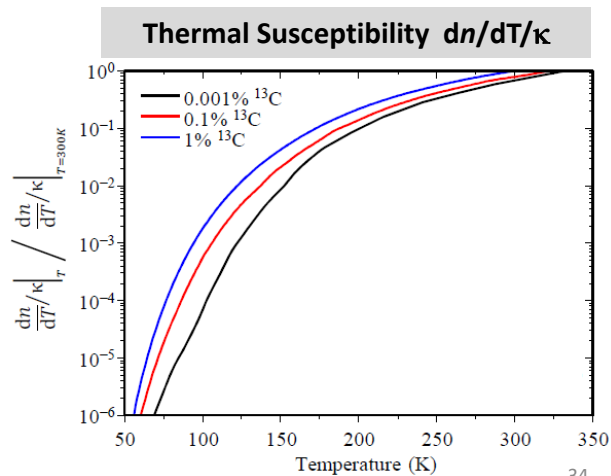
$10^2$  enhancement for natural diamond

$10^4$  enhancement for enriched 0.001%

Wei et al, *Phys Rev Lett*, 70: 3764 (1993)

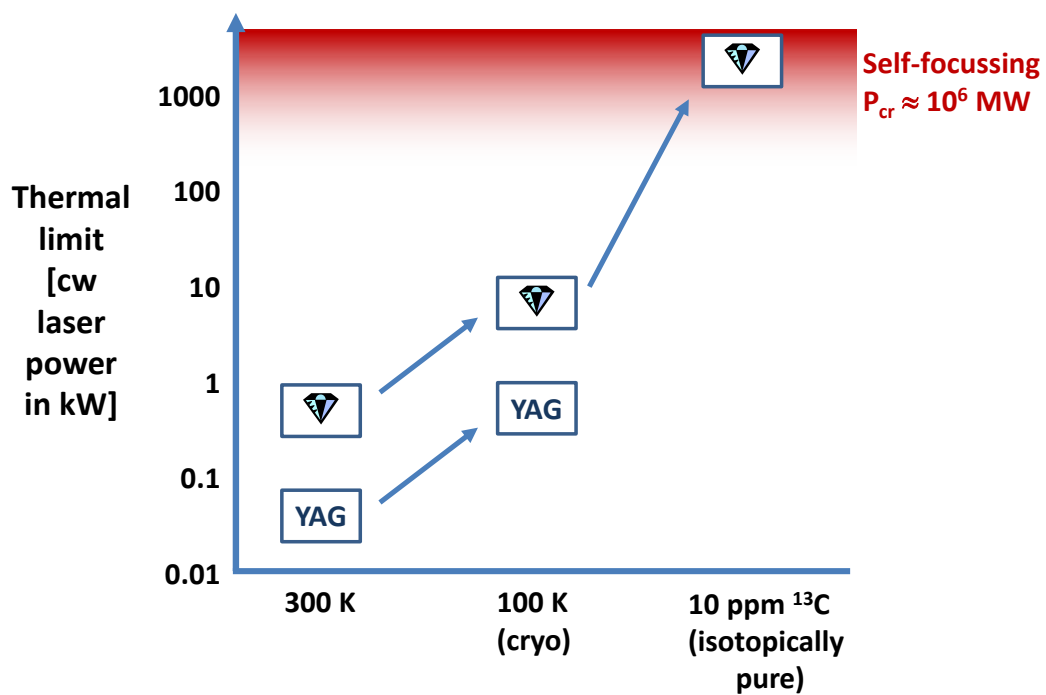
Ruf et al, *Phys Rev B*, 62: 16578 (2000)

McKay et al, *Laser and Photonic Reviews* (2016)



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## Manipulating material properties



## Properties Summary:

### Extremely good

Transmission range  
Thermal conductivity  
UV Raman gain coeff.  
Brillouin frequency  
Low Kerr nonlinearity  
Low optical phonon population at room temperature  
CW damage threshold

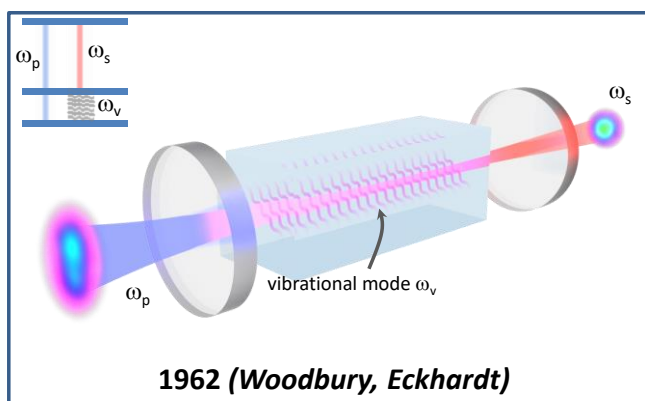
### “Un-exceptional”

Refractive index  
Raman gain coeff.  
(visible and infrared)  
Phonon dephasing time  
Photoelastic coefficient  
Two-photon absorption only strong close to bandgap  
Pulsed damage threshold

### Poor

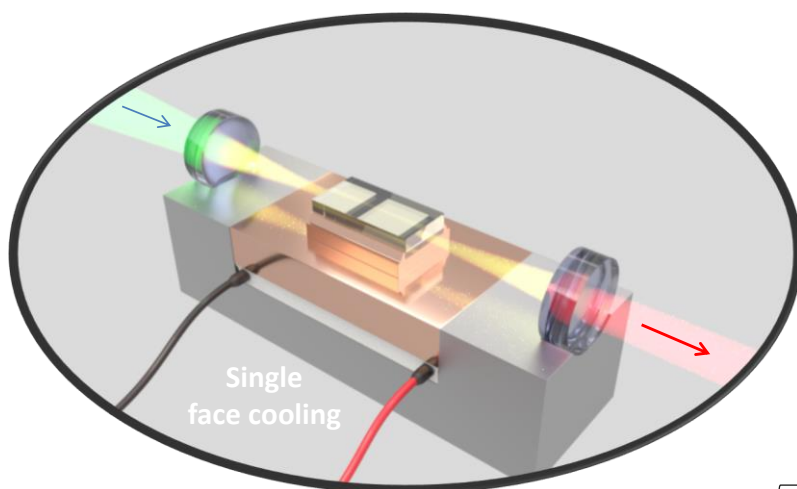
Mid-infrared transmission (4-6  $\mu\text{m}$ )  
Thermo-optic coefficient ( $T > 250\text{K}$ )

# The Raman laser



Optical Mixing	Raman Lasers	Fluorescence Lasers
$\chi^{(2)}, \chi^{(3)}$ harmonics, anti-/Stokes phase matched continuous tuning (OPOs) aberrations preserved	$\chi^{(3)}$ Stokes shift "auto-phasematched" beam cleanup cascadable	energy storage in medium Stokes shift incoherent pumping brightness enhancement

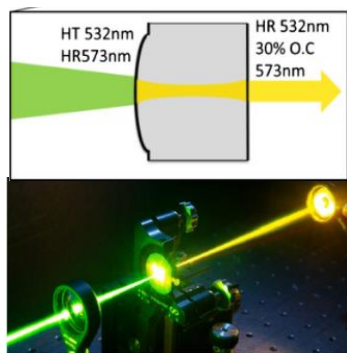
# External Cavity Raman Laser



$$\omega_v = 1332 \text{ cm}^{-1} \rightarrow$$

Wavelengths (nm)	Stokes Order		
	Pump $\lambda$	First	Second
266	276	286	298
532	573	620	676
1064	1240	1485	1851

## Examples ...



### Monolithic External Cavity Raman Laser

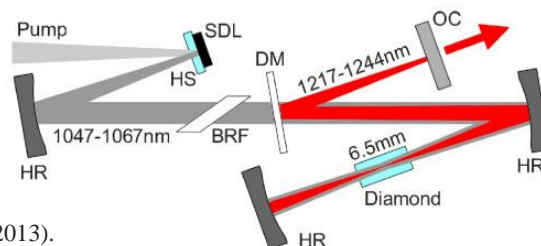
- Record efficiency 84%
- 134 mW, 10 kHz, 1.5 ns pulses

Reilly et al, *Opt. Lett.* 40, 930 (2015)

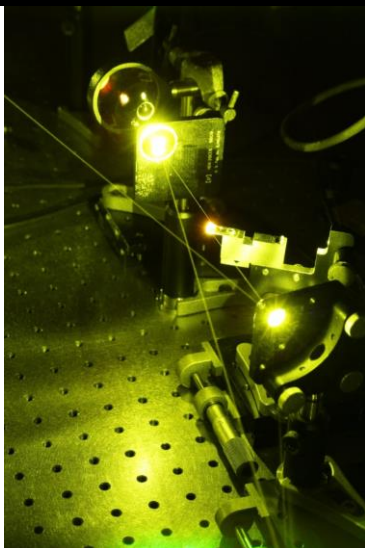
### Intracavity CW Raman Laser

- Tunable output via pump tuning
- 4.4 W max, 1217-1244 nm

Parrotta et al, *Opt. Express* 19, 24165 (2011)  
*IEEE J. Sel. Topics Quantum Electron.* 19, 1400108, (2013).



## Examples ...



### Synchronously Pumped, Ultrafast

- 2.2 W, 21 ps, 573 nm

Spence et al, *Opt. Lett.*, vol. 35, 556, (2010).

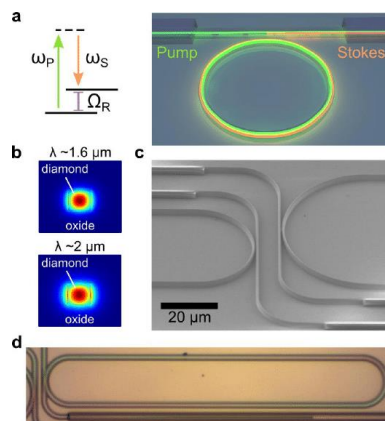
(25 fs recently demonstrated)

Lin et al, *Opt. Lett.*, 41, 1861 (2016)

### On-Chip Racetrack Resonator

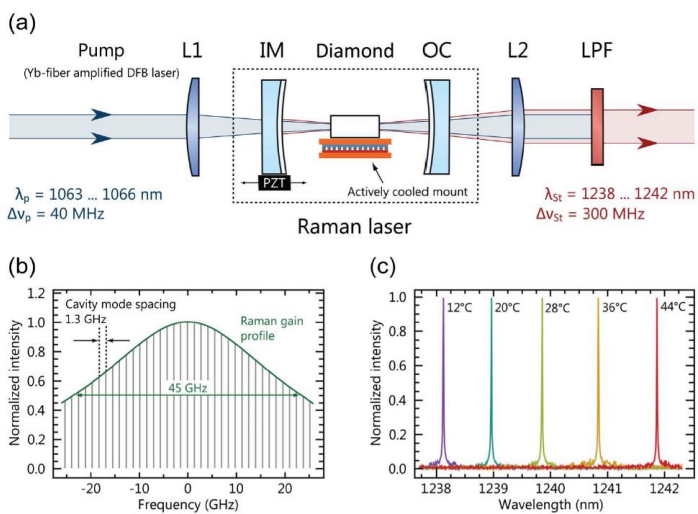
- Tunable output near 2 micron
- 85 mW threshold (no free carrier absorption)

Latawiec, et al., *Optica*, 2, 11, 924 (2015).





## Examples ...

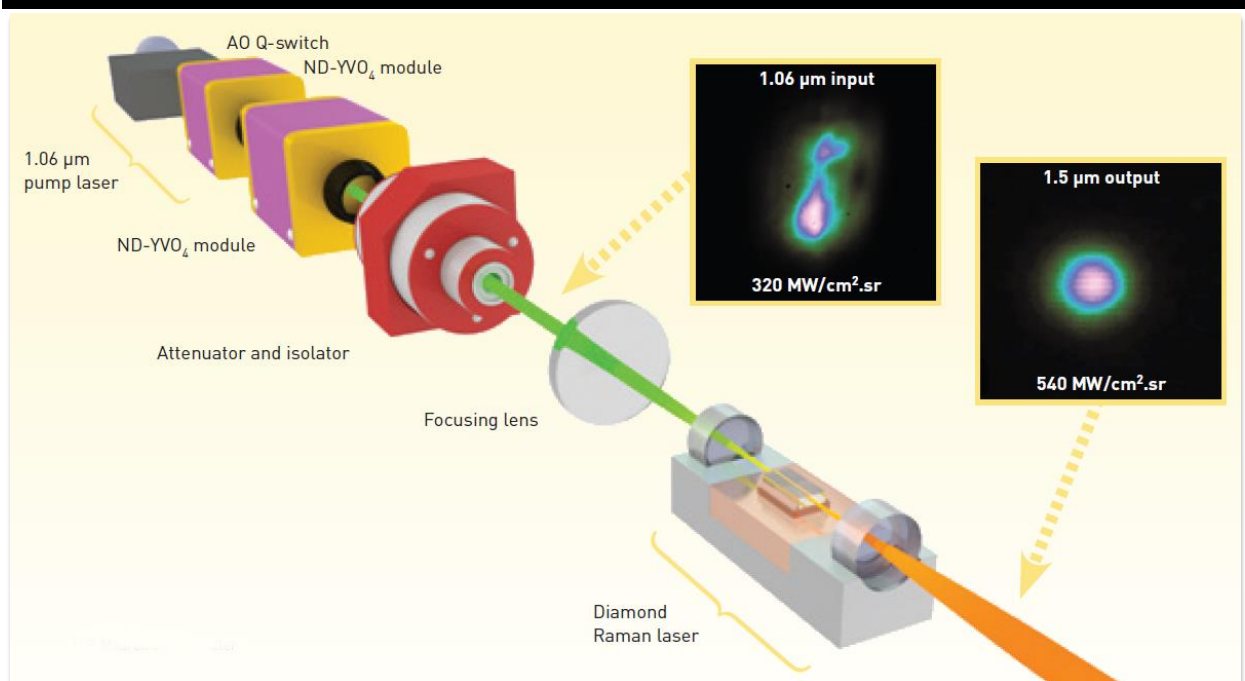


### Single longitudinal mode

- Exploits lack of the spatial hole burning in a Raman gain medium
- 3 W, tunable across 1235-1245 nm

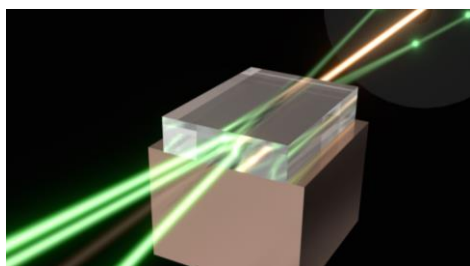
Lux et al, *Optica*, vol. 3, 876, (2016).

## Simultaneous brightness & $\lambda$ conversion



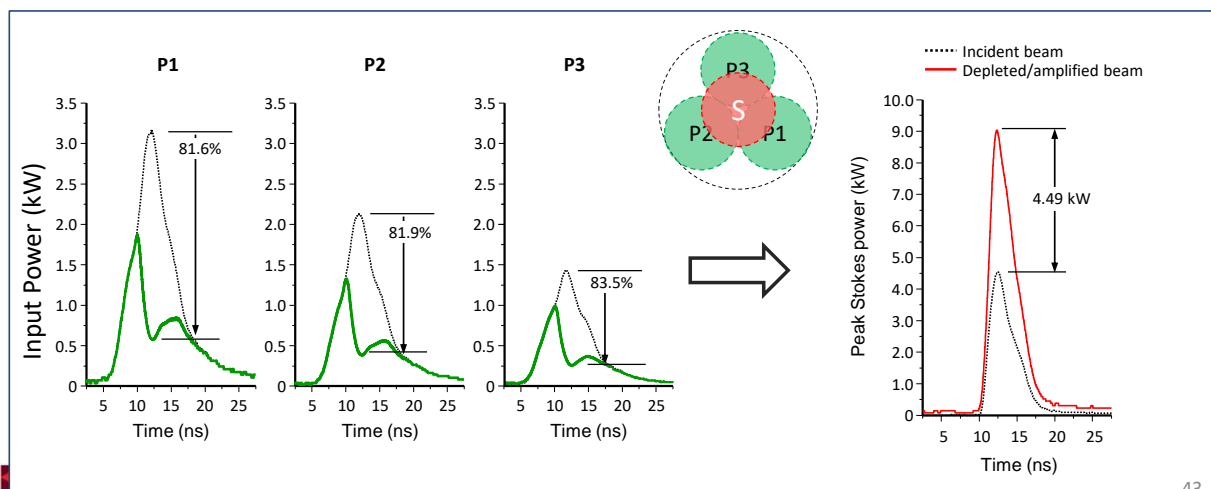
# Non-collinear Raman beam combination

In diamond:



$\Delta\omega_R = 30 \text{ GHz}$   
 $g_R \sim 10 \text{ cm/GW}$   
 $\kappa = 2000 \text{ W/m.K}$   
 $\Rightarrow$

- Multimode, uncorrelated, pumps
- Short interaction lengths
- High average power

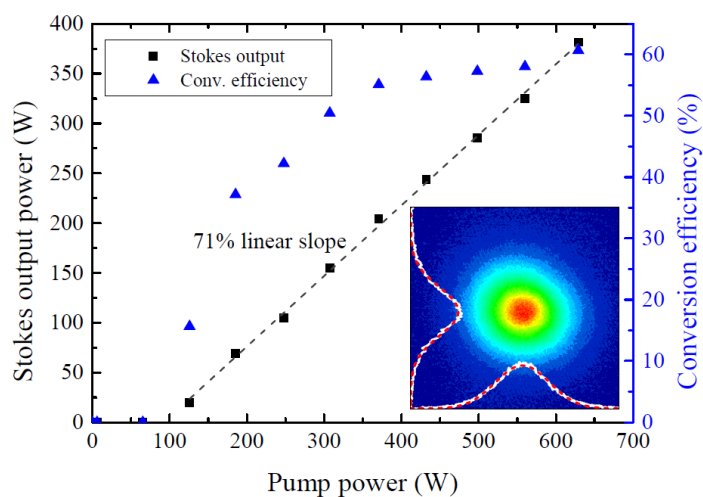
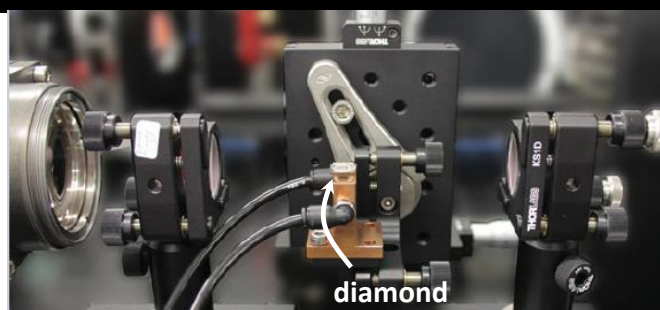


IMQ PHOTONICS

McKay, Coutts, Spence, Mildren, *Laser and Photonics Reviews* (2016)

# Fiber laser pumped

**Input**  
1 kW CW  
1060 nm  
Linewidth 30 GHz  
Circular polarization



- 150 W for 5s
- 380 W for 10 ms  
(*steady-state = 10 μs*)

**Collaborators:** J. Nold, M. Strecker and T. Schreiber at the Fraunhofer IOF (Institute of Applied Optics and Precision Engineering), Jena, Germany

R. J. Williams, J. Nold, M. Strecker, O. Kitzler, A. McKay, T. Schreiber, R. P. Mildren, *Laser Photon. Rev.*, **9** p405 (2015)

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# Review Sources

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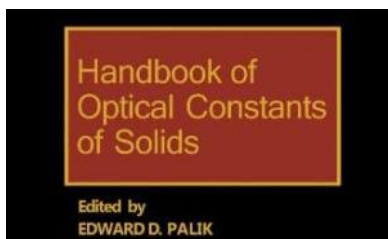
## Optical Study of the Electronic Structure of Diamond\*

R. A. ROBERTS† AND W. C. WALKER

Rep. Prog. Phys., Vol. 42, 1979. Printed in Great Britain

## Optical absorption and luminescence in diamond

JOHN WALKER†



## Optical properties of diamond

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Michael E. Thomas and William J. Tropf

## Light Scattering in Solids II

by M. Cardona

Max-Planck-Institut für Festkörperforschung

OSA Webi

Edited by S. Koizumi, C. E. Nebel, and M. Nesladek

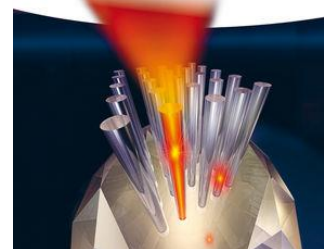
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Edited by Rich Mildren, James Rabeau

## Optical Engineering of Diamond



ALEXANDER M. ZAITSEV

## Optical Properties of Diamond

A Data Handbook

Springer

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