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Mid-infrared lasers based on transition metal doped II-VI semiconductors

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The work reported here partially involves intellectual property developed at the University of Alabama at Birmingham (UAB). This intellectual property has been licensed to the IPG Photonics Corporation. The UAB co-authors declare competing financial interests.

2nd Int. Conference on Lasers, Optics & Photonics 09/08/14, Philadelphia, USA

Outline

- **Overview, Introduction and Motivation**
- **What is special about Cr and Fe doped ZnSe/S for mid-IR?**
- **Spectroscopic properties of Cr and Fe-doped ZnSe/S Gain Media**
- **Progress in fiber-bulk hybrid mid-IR lasers**
 - CW**
 - Gain Switched**
 - Free-running oscillation**
 - Mode-Locked**
- **Practical applications**
- **Conclusions and Future Work**

Motivation for Cr²⁺, Fe²⁺ doped ZnSe/S lasers

For molecular time-resolved measurements, molecular spectroscopy, trace gas analysis, biomedical applications, etc. one should directly reach molecular fingerprint 2-20 μm region.

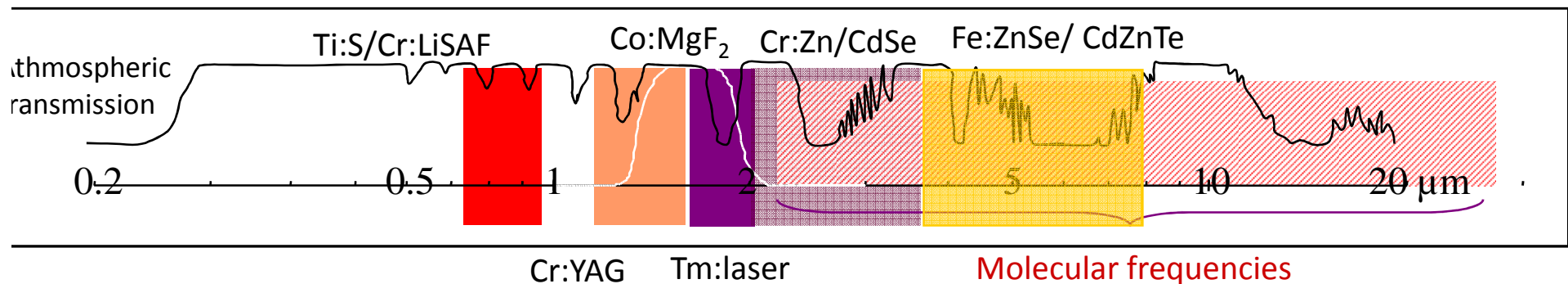
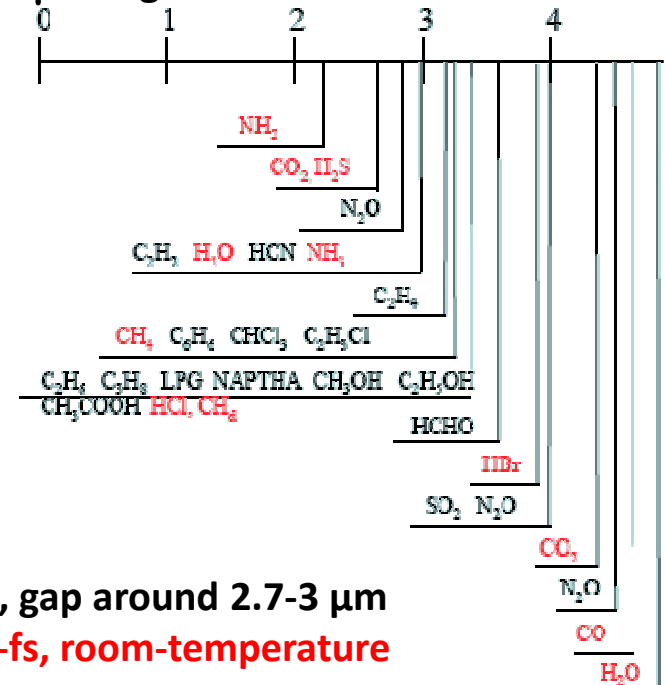
→ Mid-IR tunable sources are required

Requirements:

- Sufficient bandwidth
- Low cost, compact, convenient pumping low threshold
- High brightness, i.e. good spatial coherence (TEM₀₀).

Solutions:

- OPO (bulk ZGP, PPLN, orientation-patterned GaAs): almost ideal solutions, but rather complex and costly
- QCL: nice solution for λ > 3.4 μm, not as broadband
- Semiconductor InGaAsSb/GaSb lasers: narrow tuning, no fs, gap around 2.7-3 μm
- **Crystalline vibronic lasers: ultrabroadband up to 50 % λ, cw-fs, room-temperature**



What is special about $\text{TM}^{2+}:\text{II-VI}$?

TM (Cr^{2+} , Co^{2+} , V^{2+} , Mn^{2+} , Fe^{2+} , Ni^{2+}) doped II-VI (II-Cd, Zn) (VI- S, Se, Te) compounds have a wide bandgap and possess several important features that distinguish them from other oxide and fluoride laser crystals.

Chemically stable divalent TM dopant ions, **no need for charge compensation.**

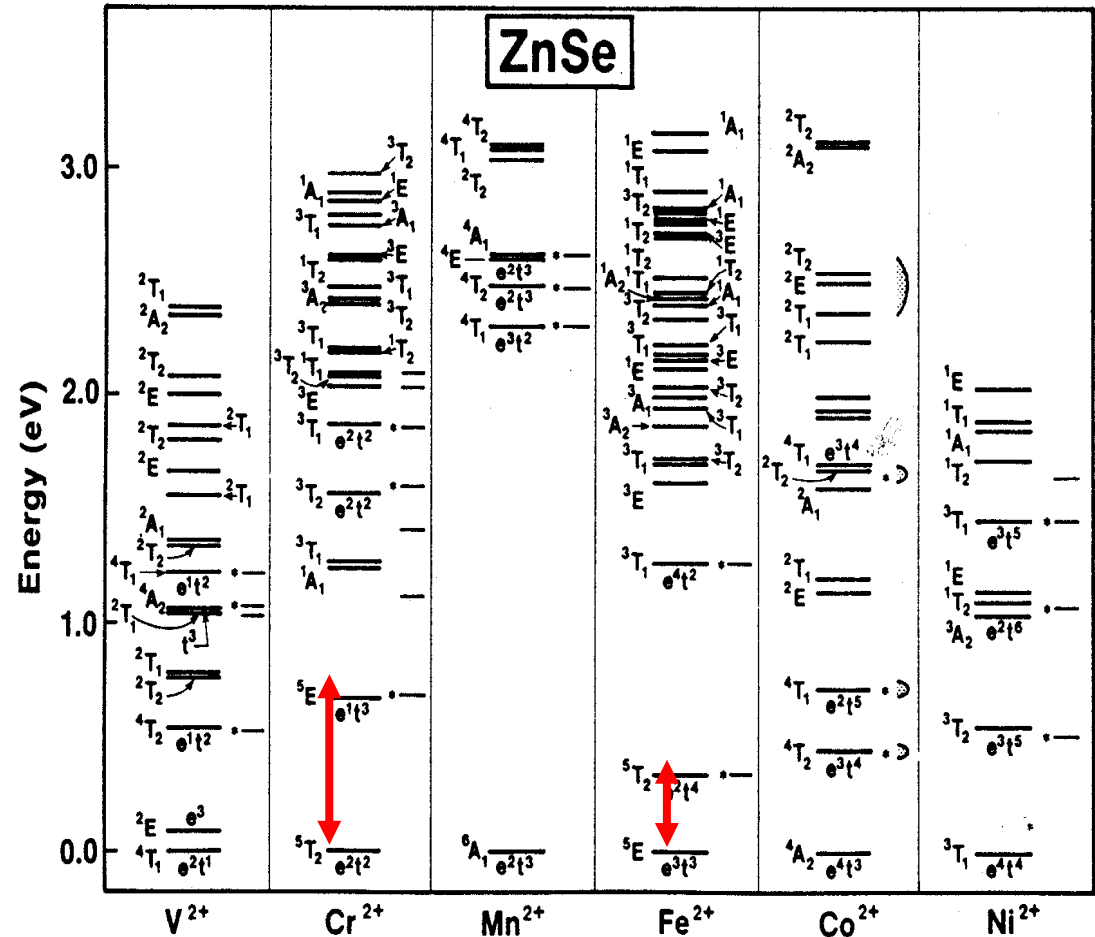
Crystallization as tetrahedrally coordinated structures, Tetrahedral coordination (T_d) gives small crystal field splitting, placing the dopant transitions into the IR.

Optical phonon cutoff occurs at very low energy, maximizing the prospects for radiative decay of mid-IR luminescence in these crystals.

Host	Phonon cut-off $\nu_{\text{max}}, \text{cm}^{-1}$
ZnTe	210
ZnSe	250
ZnS	350
YAG	850
YLF	560

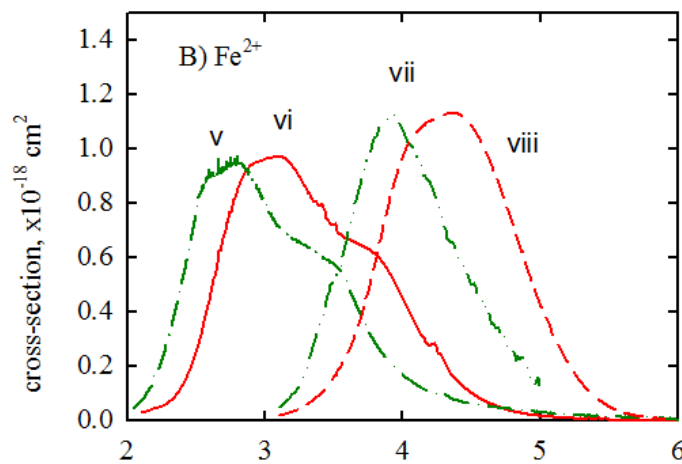
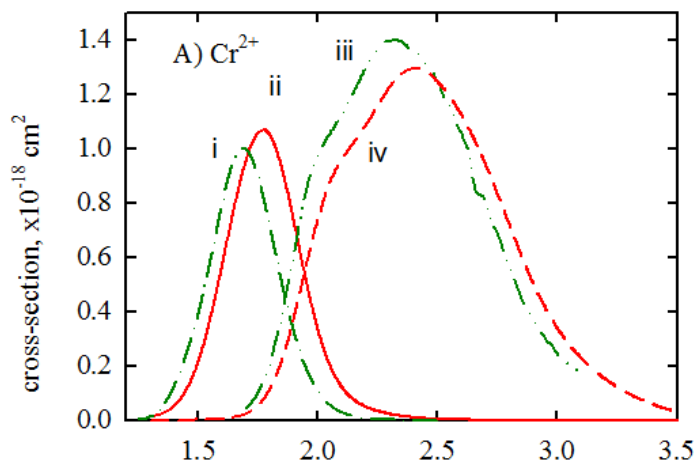
Why Cr²⁺ & Fe²⁺?

- ❑ First excited levels lie at the right energy to generate 2-3 (Cr) & 3.5-5 μm (Fe) mid-IR emission.
- ❑ The ground and first excited levels have the same spin, and therefore will have a relatively high cross-section of emission.
- ❑ Higher lying levels have spins that are lower than the ground and first excited levels, greatly mitigating the potential for significant excited state absorption at the pump or laser transition wavelengths.
- ❑ The orbital characteristics of the ground and first excited levels are different, and will experience a significant Franck-Condon shift between absorption and emission, resulting in broadband “dye-like” absorption and emission characteristics, suitable for a broadly tunable laser.

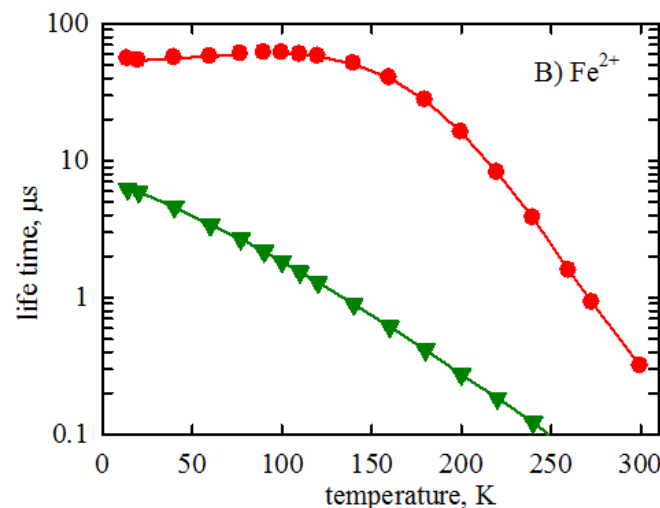
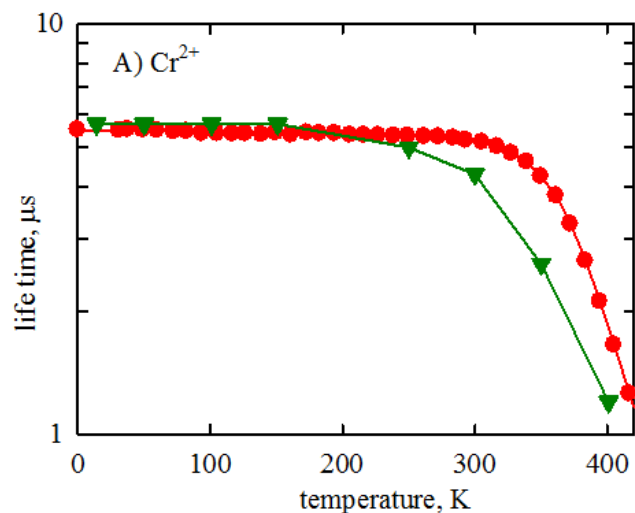


Calculated Multiplet Structure for 3d impurities in ZnSe (after A Fazio, et al., Phys. Rev. B, 30, 3430 (1984))

Spectroscopic properties of Cr and Fe doped ZnSe/S compounds



A) Absorption (Cr:ZnS- curve i; Cr:ZnSe-curve ii) and emission (Cr:ZnS- curve iii; Cr:ZnSe-curve iv) cross-sections of Cr^{2+} ions in ZnS and ZnSe; B) Absorption (Fe:ZnS- curve v; Fe:ZnSe-curve vi) and emission (Fe:ZnS- curve vii; Fe:ZnSe-curve viii) cross-sections of Fe^{2+} ions in ZnS and ZnSe.



Luminescence lifetime versus temperature for chromium (A) and iron (B) doped ZnS (triangle) and ZnSe (circle) crystals.

S. Mirov, V. Fedorov, D. Martyshkin, I. Moskalev, M. Mirov and S. Vasilyev, "Progress in Mid-IR Lasers Based on Cr and Fe Doped II-VI Chalcogenides,"
J. Special Topics in Quantum Electronics, submitted April 2014.

Challenges of Bulk Crystal Fabrication

- Bridgman technique (sublimation of chemicals requires simultaneous use of high temperature and pressure, (1550C & 75 atm – economical viability?)
- Chemical vapor transport (CVT) (doping is very difficult)
- Physical vapor transport methods (PVT) (doping is very difficult)
- Hot-pressed ceramics
- The post-growth thermal diffusion doping of ZnSe/S ceramics
- Pulsed laser deposition
- Nano & micropowders, composite II-VI-liquid, II-VI-polymer and II-VI-glass gain media

Key challenges:

Hard to get high Cr concentration

Hard to get uniform Cr distribution

Hard to make large Cr²⁺:ZnSe crystals

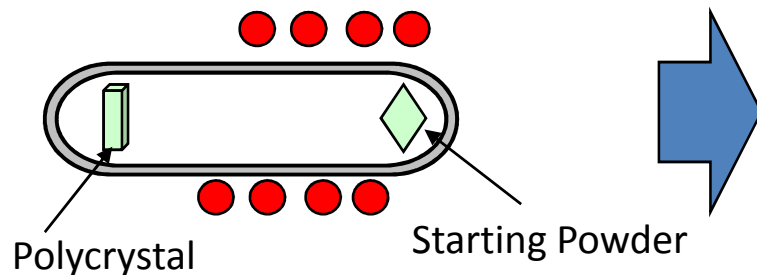
High scattering losses

Low damage threshold

Strong thermal lensing effects

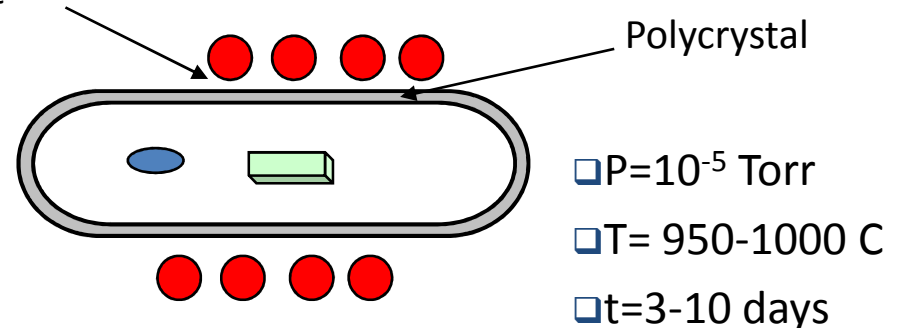
Bulk Crystal Preparation by a Quantitative Post-growth Thermal Diffusion

Chemical vapor transport
polycrystal growth or IR
window purchase



Post-growth thermal
diffusion

Dopant



Fast diffusion of dopant with suppressed sublimation in Zn and Se sub-lattices

Low scattering loss (1-2 % per cm) in thermally diffusion doped crystals

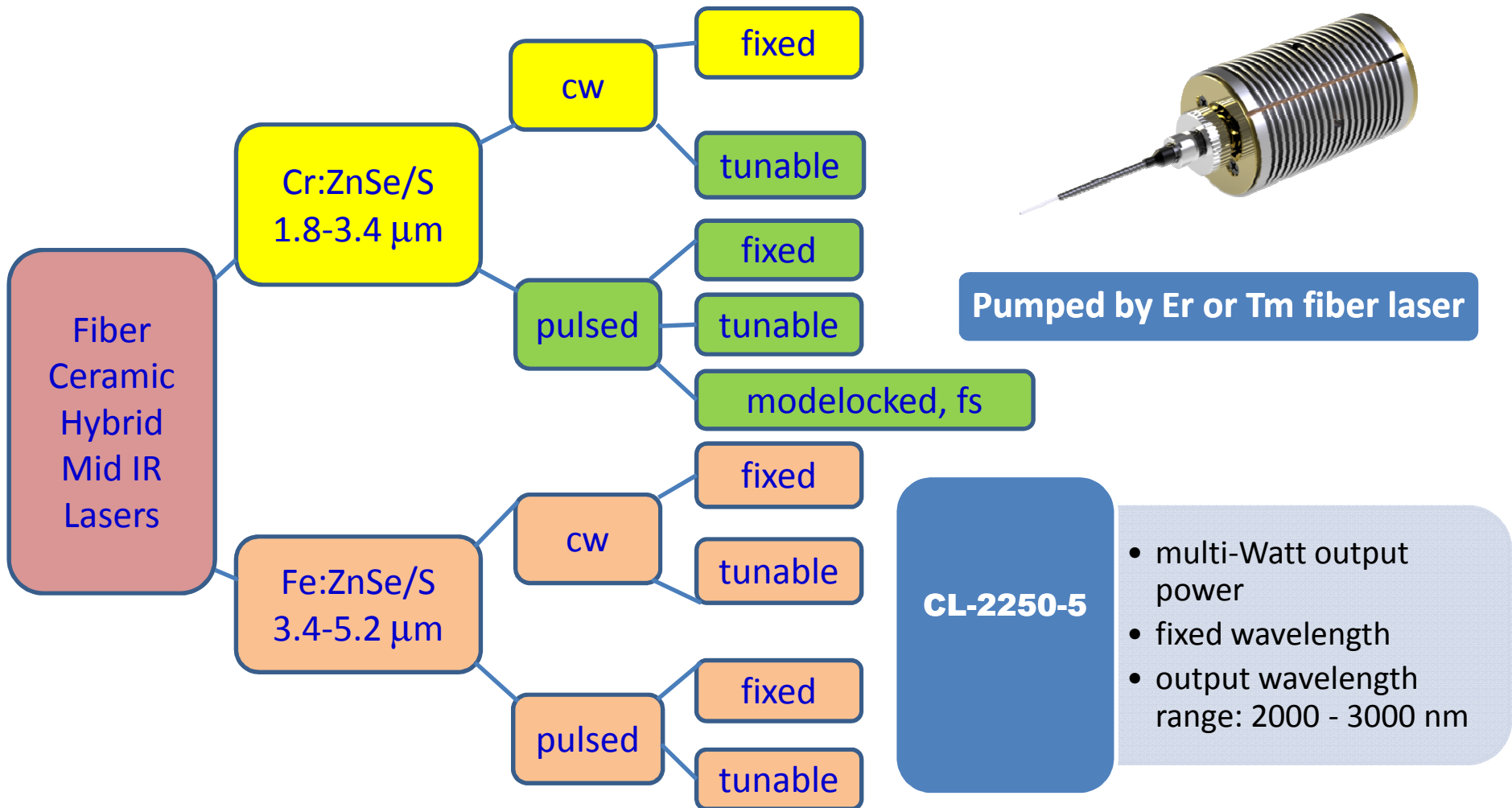
Uniformly-doped, reasonably large samples up to 7 mm thickness

Quantitative technology enabling pre-assigned concentration of dopant with accuracy better than 3%

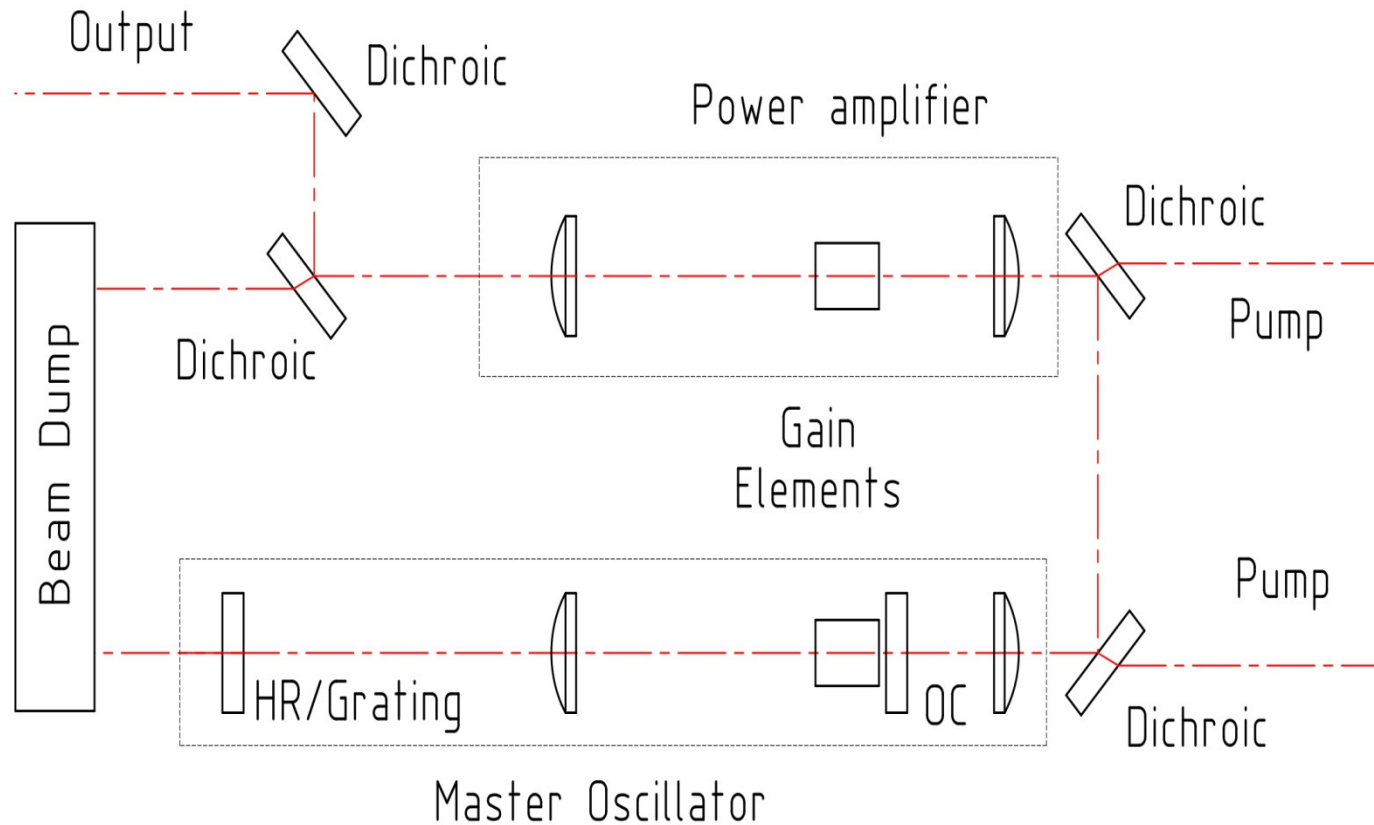
Good for High-Power (tunable) Lasers

S.B. Mirov, V.V. Fedorov, (November 1, 2005) Mid-IR microchip laser: ZnS:Cr²⁺ laser and saturation absorption material", *US Patent* No 6,960,486.

Fixed Frequency Wavelength Converter



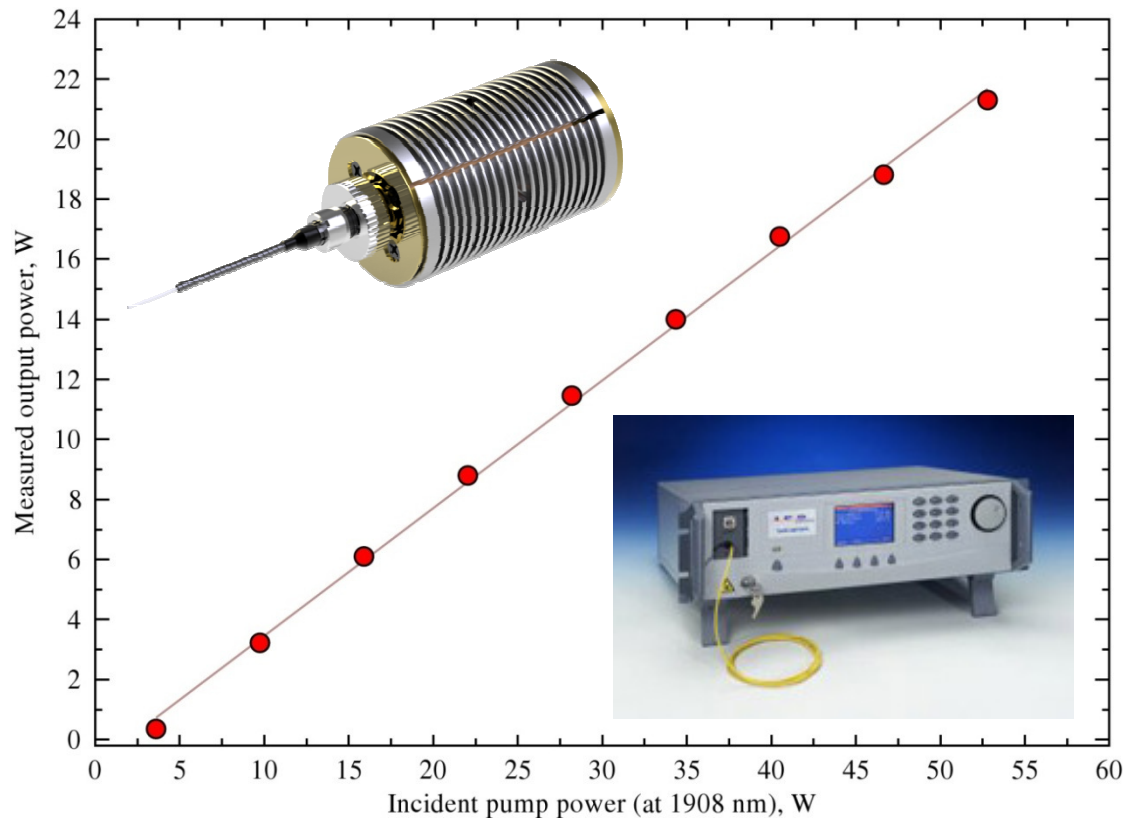
Optical scheme of high-power tunable $\text{Cr}^{2+}:\text{ZnS}$ and $\text{Cr}^{2+}:\text{ZnSe}$ CW MOPA systems based on linear cavity design



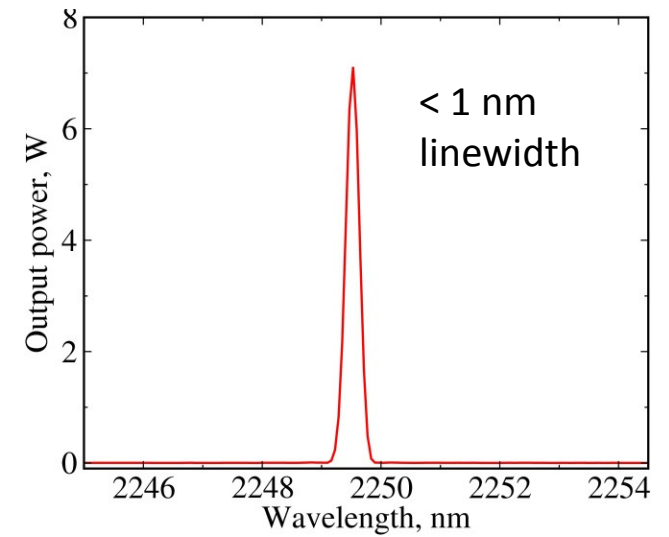
CL-2250-20

Fixed Frequency Mid IR Wavelength Converter

- multi-Watt output power
- fixed wavelength
- output wavelength range: 2000 - 3000 nm



Typical Output Spectrum



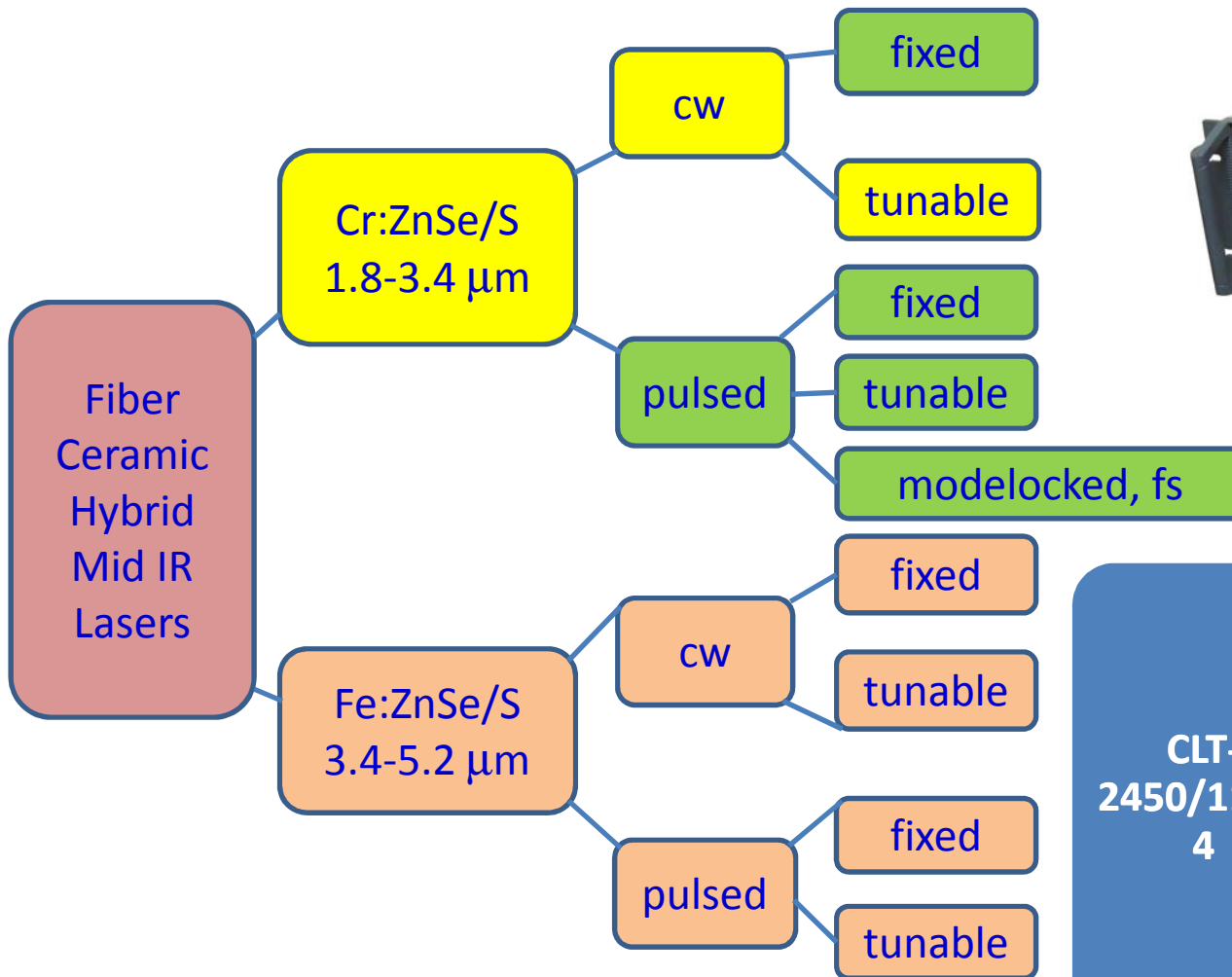
Plastics Cutting, Welding, Marking, Drilling

Tissue & Bone Cutting

Dental Applications

Skin Rejuvenation

CW Tunable Cr:ZnSe/S Laser



CLT-2450/1100-4

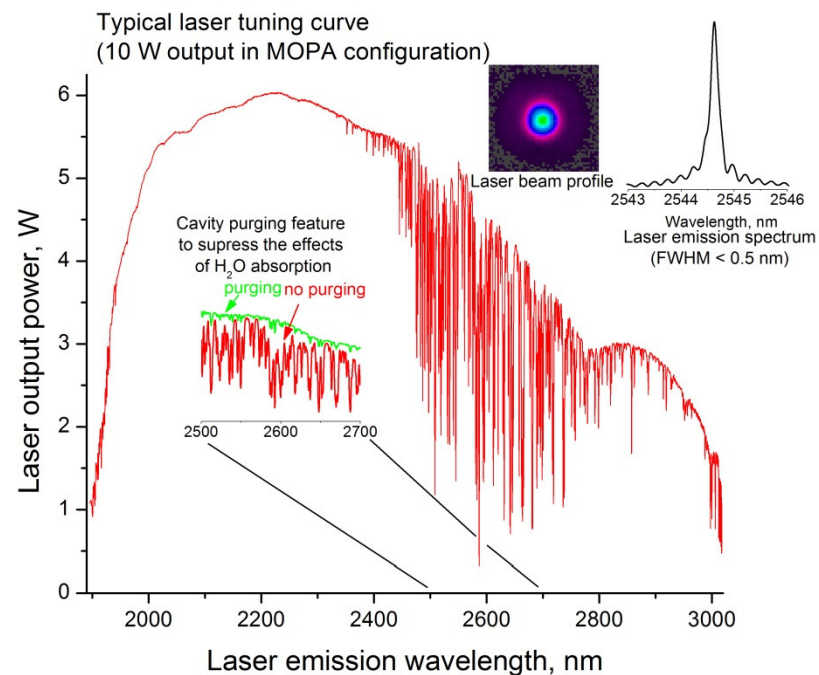
- cw output, 20 mW to >10 Watt
- typical linewidth < 0.5 nm, < 0.1 nm available
- up to 1100 nm tunable range
- any central wavelength within 2-3μm
- TEM₀₀, M² < 1.2



CW Tunable Cr:ZnSe/S Laser

CLT-2450/1100-4

- cw output, 20 mW to >10 Watt
- typical linewidth < 0.5 nm, < 0.1 nm available
- up to 1100 nm tunable range
- any central wavelength within 2-3 μ m
- TEM₀₀, M² < 1.2



Spectroscopy

OPO Pump Source

Medical Applications

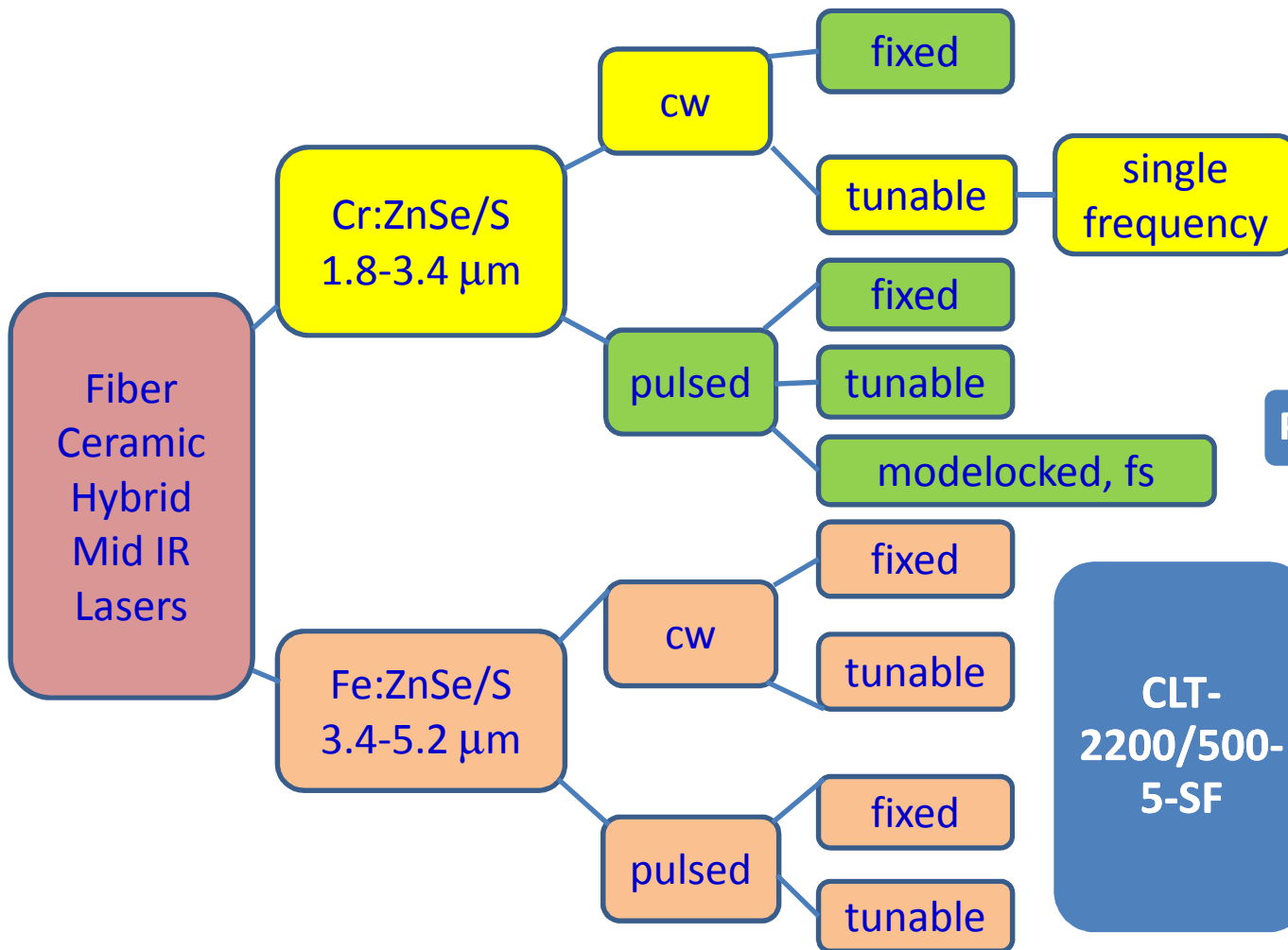
Environmental Monitoring

Industrial Process Control

Material processing



Single-Frequency CW Tunable Laser



Pumped by Er fiber laser

CLT-2200/500-5-SF

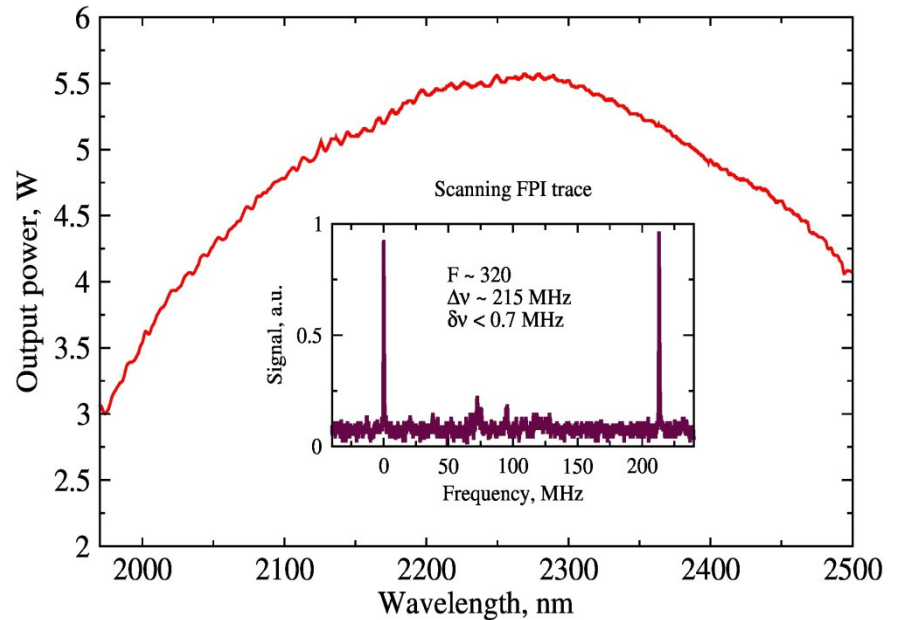
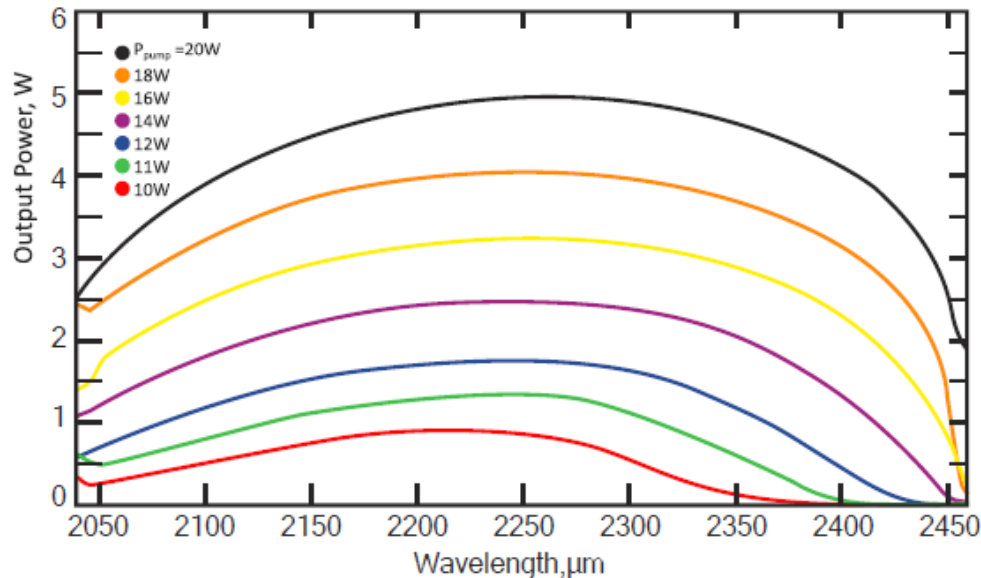
- multi Watt output power
- narrow linewidth, <1 MHz
- large tuning range with a single set of optics (any wavelength within 2000 - 3000 nm tuning range)

CLT-2200/500-5-SF

Single-Frequency CW Tunable Laser

- multi Watt output power
- narrow linewidth, <1 MHz
- large tuning range with a single set of optics (any wavelength within 2000 - 3000 nm tuning range)

Output Power vs Wavelength for Different Levels of Pump Power



Output power vs wavelength of a tunable single-frequency $\text{Cr}^{2+}:\text{ZnS}/\text{Se}$ laser system. The insert shows an interferogram obtained with high-resolution ring interferometer

High Resolution Spectroscopy

OPO Pump Source

Industrial Process Control

Medical Applications

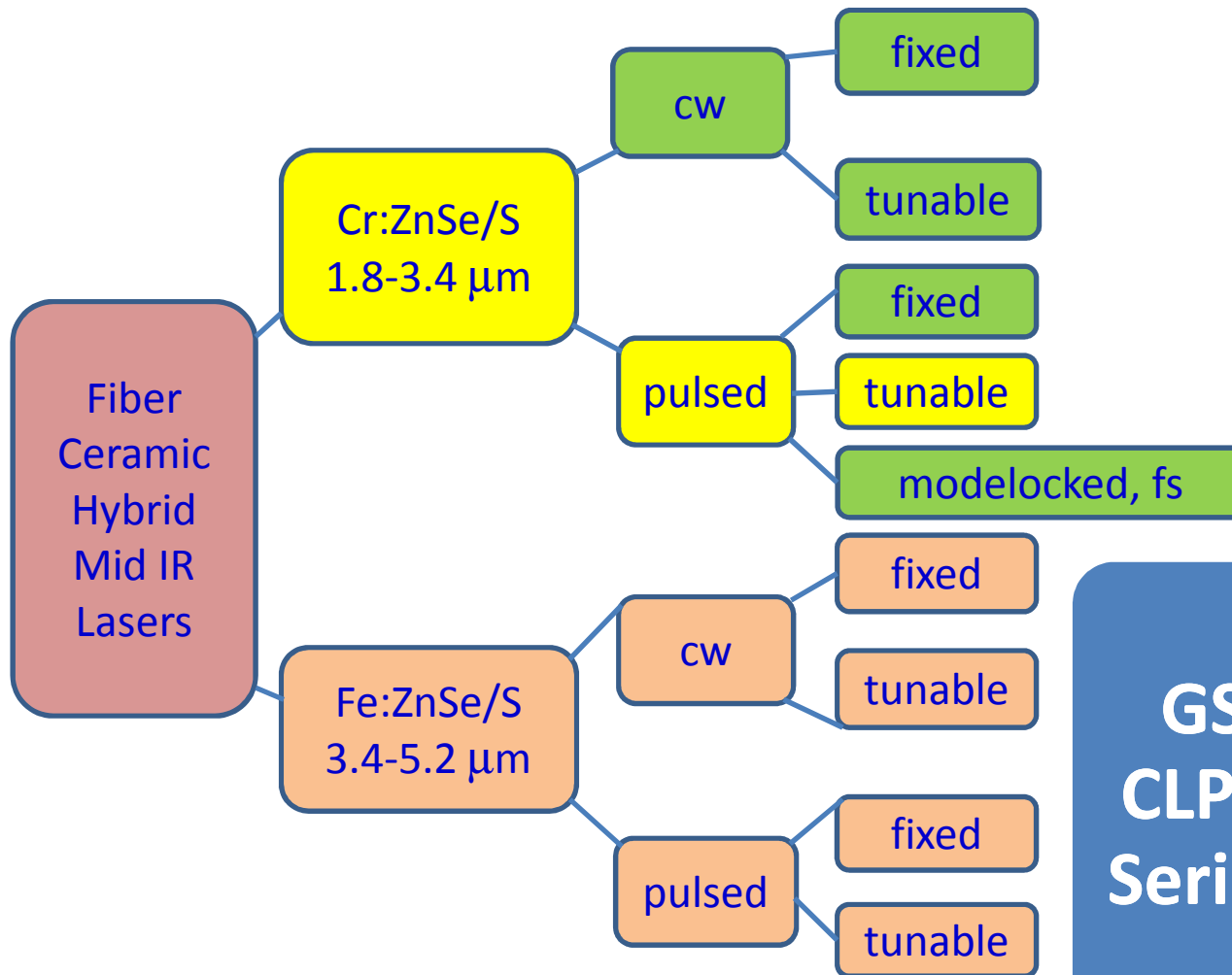
Material processing

Environmental Monitoring

Free Space Communications



Gain-Switched Pulsed Tunable Cr:ZnSe/S Lasers



**GS
CLPN
Series**

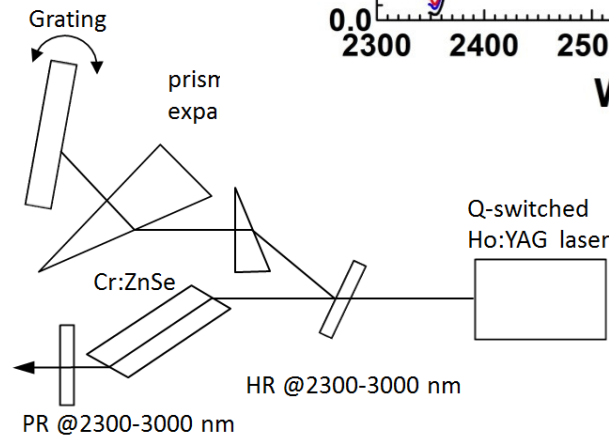
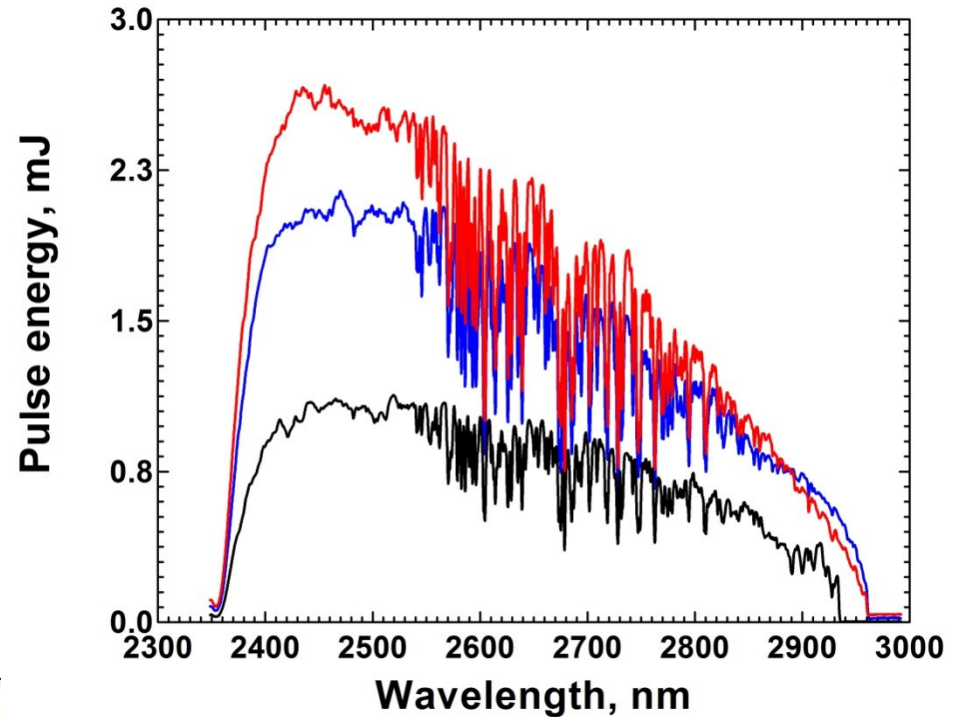
- Tunability 2300-3000 nm
- Pulsed output energy up to 10 mJ
- Pulse duration 2-20 ns
- Linewidth <0.5nm
- Repetition rate 0.1-1 kHz
- TEM₀₀



Gain-Switched Cr:ZnSe/S Lasers. CLPN Series

Gain-switched Tunable Cr:ZnSe Laser

- Tunability 2300-3000 nm
- Pulsed output energy up to 10 mJ
- Pulse duration 2-20 ns
- Linewidth <0.5 nm
- Repetition rate 0.1-1 kHz
- TEM₀₀

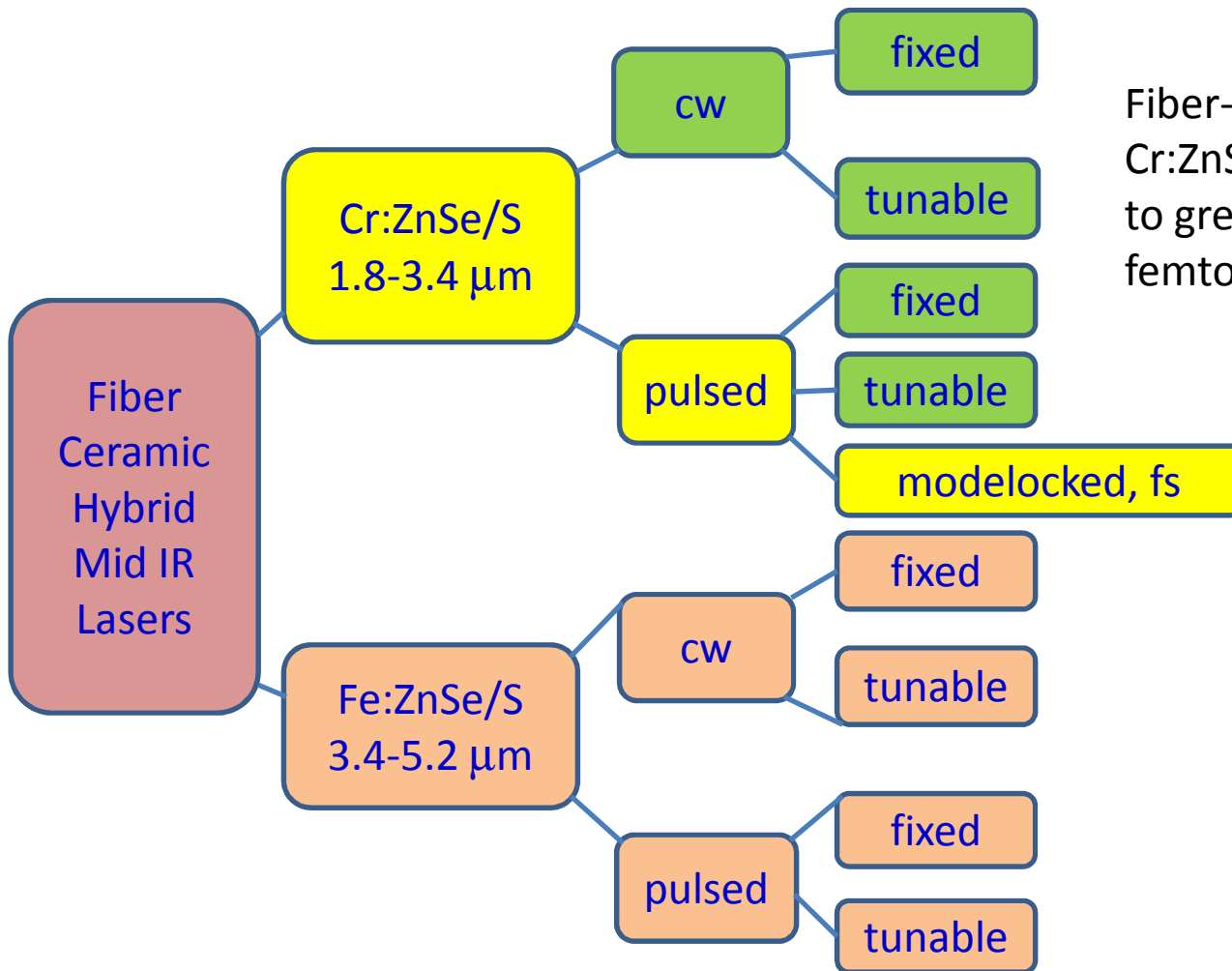


Laser skin treatment

Laser scalpel

Material processing

Ultrafast Mid IR Lasers



Fiber-pumped mode-locked Cr:ZnS/Se lasers are very similar to green-laser pumped femtosecond Ti:sapphire lasers.

transient absorption spectroscopy

2D IR spectroscopy

2 and 3 photon bioimaging

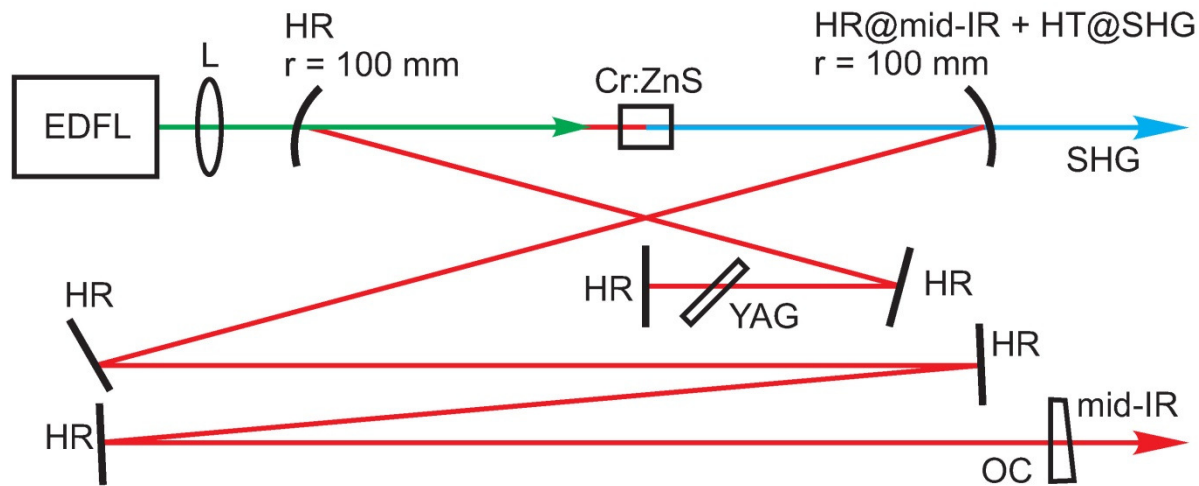
Kerr-lens mode-locked polycrystalline Cr²⁺:ZnSe/ZnS lasers

Motivation

- Power scaling of mid-IR fs oscillators
- Investigation of three-wave mixing in polycrystalline Cr:ZnS/ZnSe in femtosecond regime

Approach

- Mounting of polycrystalline Cr:ZnS/ZnSe at normal incidence
 - ✓ Reduced thermal optical effects
 - ✓ Increased pump, laser intensity
 - ✓ Enables use of long gain element with high pump absorption



Schematic of Kerr-lens mode-locked Cr:ZnS laser setup

Features:

- Standard EDFL pump at 1567 nm
- 5-mm long AR coated Cr:ZnS with 89% pump absorption
- Asymmetric 1.9 m long resonator (79 MHz mode spacing)
- 2.3 μm central wavelength
- Dispersion management by YAG plate, dispersive mirrors
- $\text{GDD} = -1500 \pm 400 \text{ fs}^2$



Kerr-lens mode-locked polycrystalline Cr²⁺:ZnSe/ZnS lasers

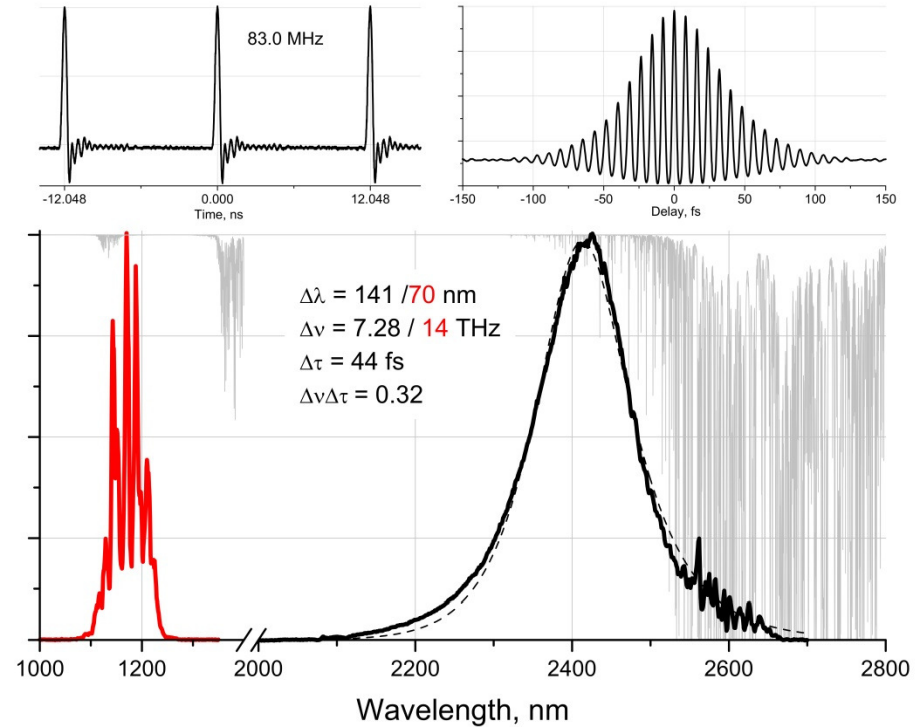
Main results

(for different laser configurations)

- Up to 2.0 W mid-IR average power
67 fs pulses, 95 MHz rep. rate
- 20% pump efficiency
- Up to 44 fs mid-IR pulse duration
at 0.5-0.6 W average power, 79 MHz rep. rate
- Up to 340 kW peak power
22 nJ, 55 fs pulses, 79 MHz rep. rate
- Up to 0.3 W SHG output
- Up to 14 THz SHG spectral bandwidth

Outlook

- 4 W Cr²⁺:ZnS/Se oscillators
- GW peak power Cr²⁺:ZnS/Se CPAs
- Single-optical-cycle Cr²⁺:ZnS/Se oscillators

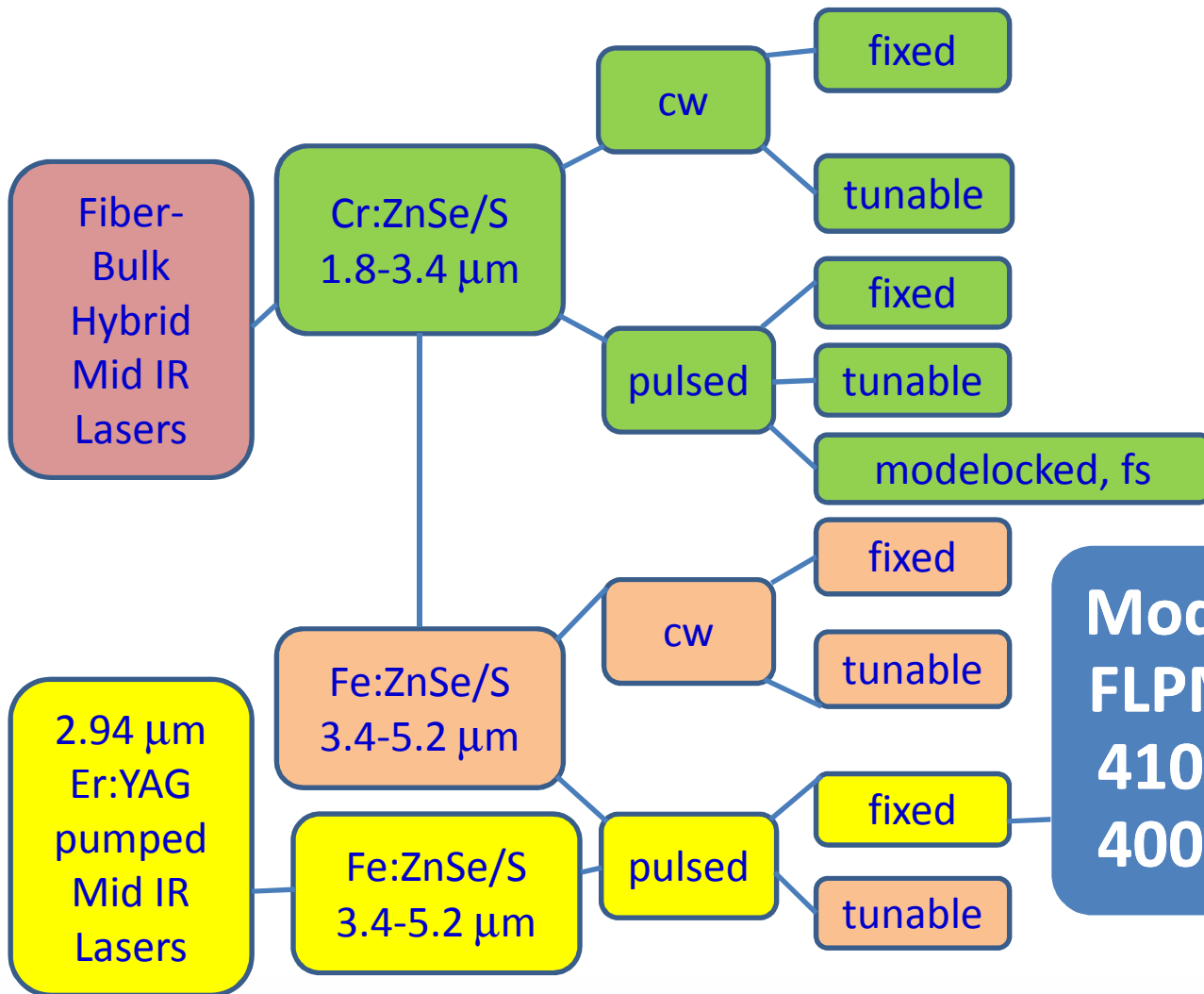


Typical fs pulse train, autocorrelation trace, emission spectrum of mode-locked Zr:ZnS/ZnSe lasers (dashed line shows sech fit)



Mid-IR, SHG, THG, FHG beams on IR-sensitive card

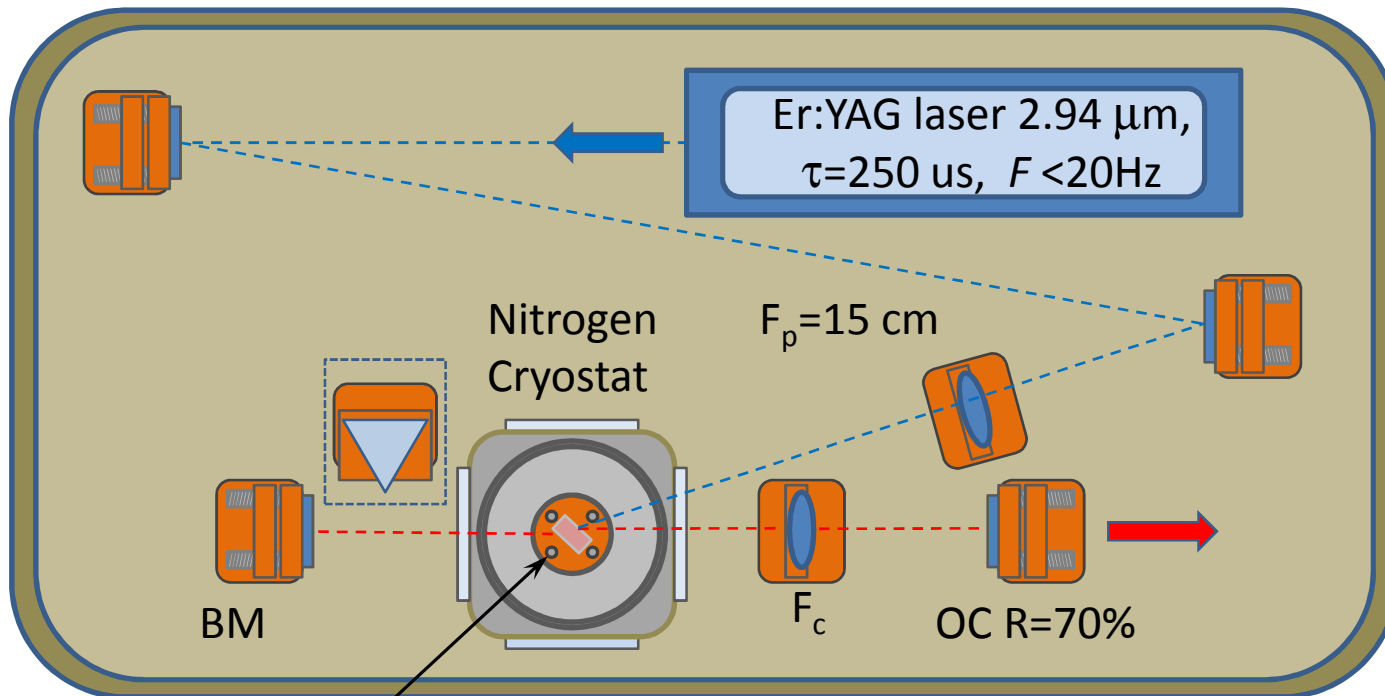
High Energy Free-Running Pulsed Fixed Frequency Fe:ZnSe Laser



**Model
FLPM-
4100-
400-4**

- Any wavelength within 3.9-5.2μm
- High Output Energy >400 mJ
- Pulse Duration 200 μs
- Max. Repetition Rate >20 Hz

Optical Scheme of Fe:ZnSe pulsed Laser

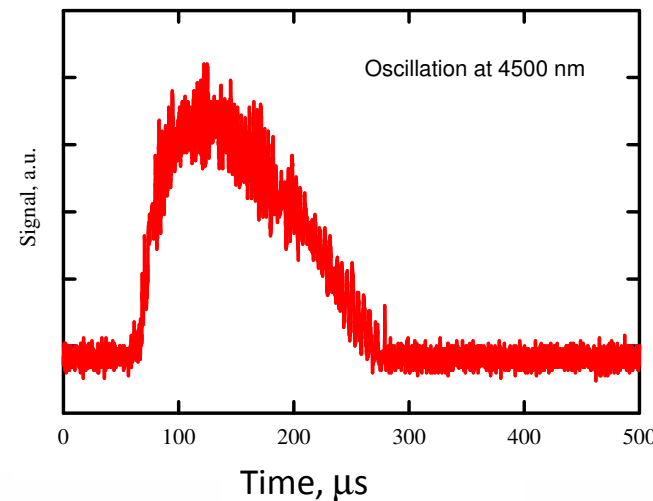
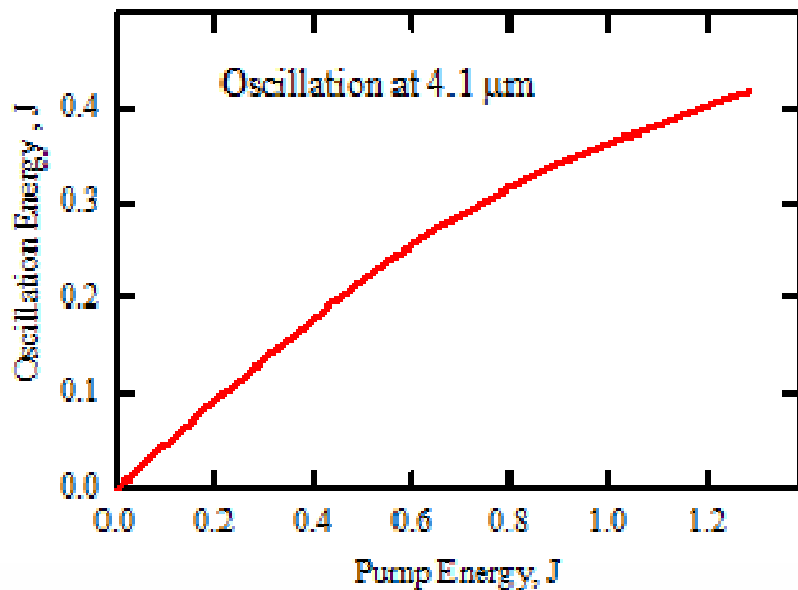
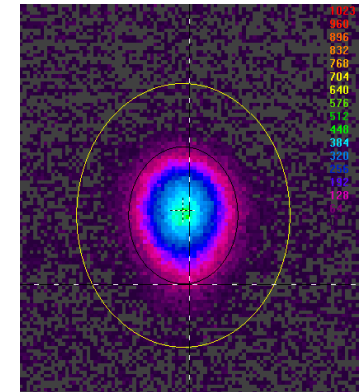
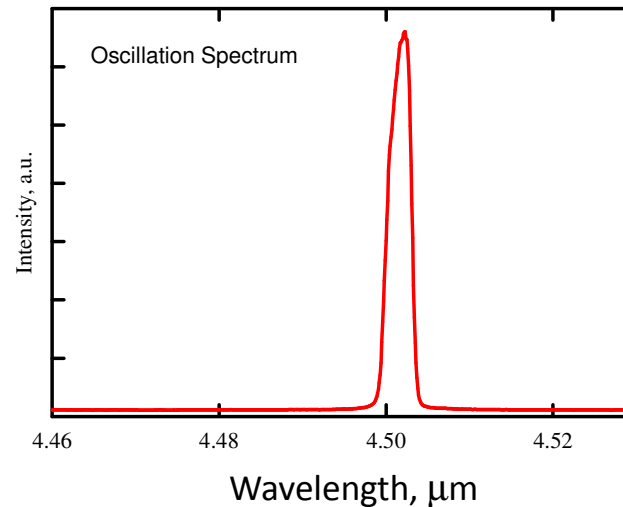


Fe^{2+} doped ZnSe polycrystalline with iron concentration of $1.5 \times 10^{19} \text{ cm}^{-3}$

High Energy Free-Running Pulsed Fixed Frequency Fe:ZnSe Laser

Model FLPM-4100-400-4

- Any wavelength within 3.9-5.1 μm
- High Output Energy >400 mJ
- Pulse Duration 200 μs
- Max. Repetition Rate >20 Hz



Spectroscopic

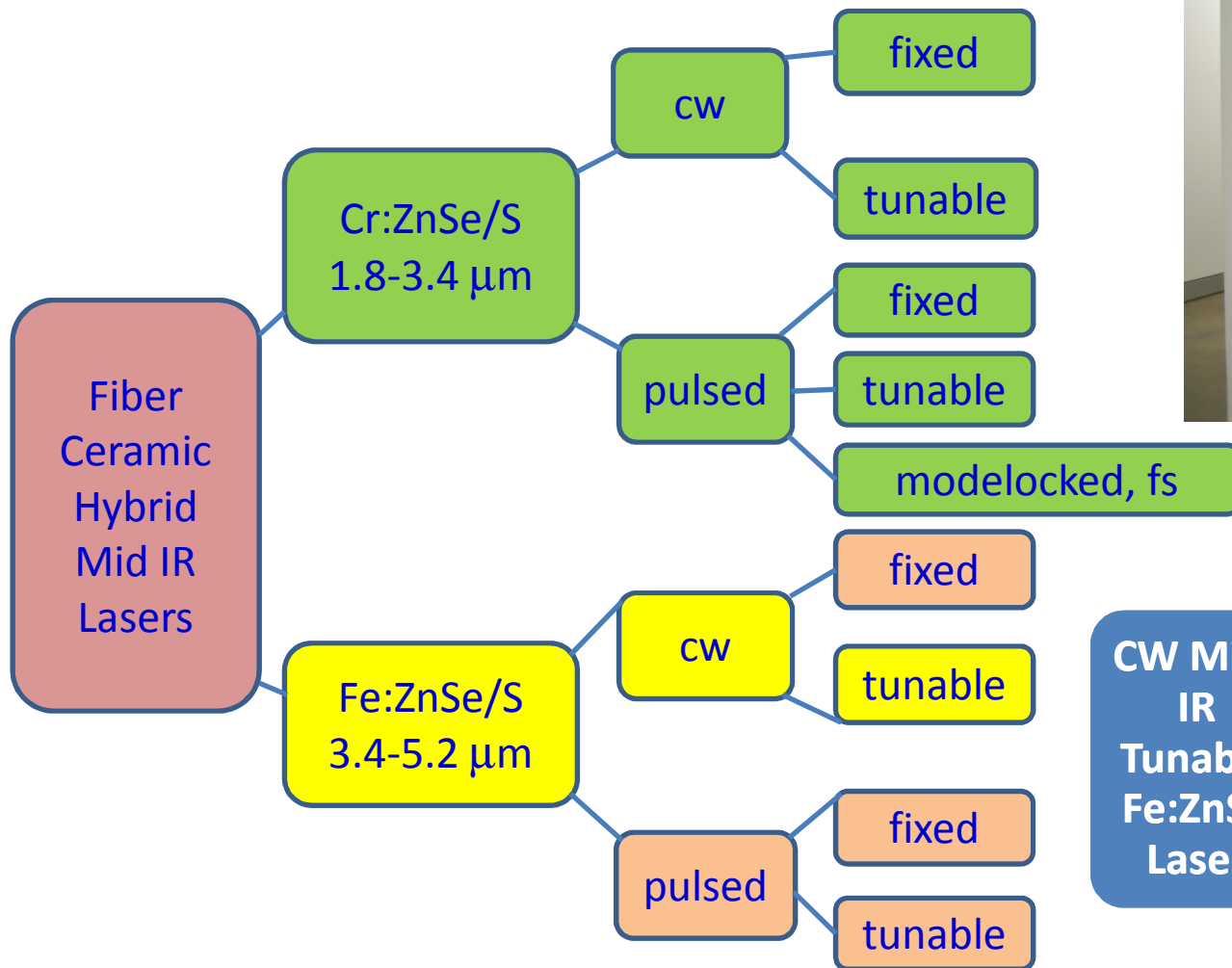
Sensing

Medical

Defense

OPO seeding and pumping

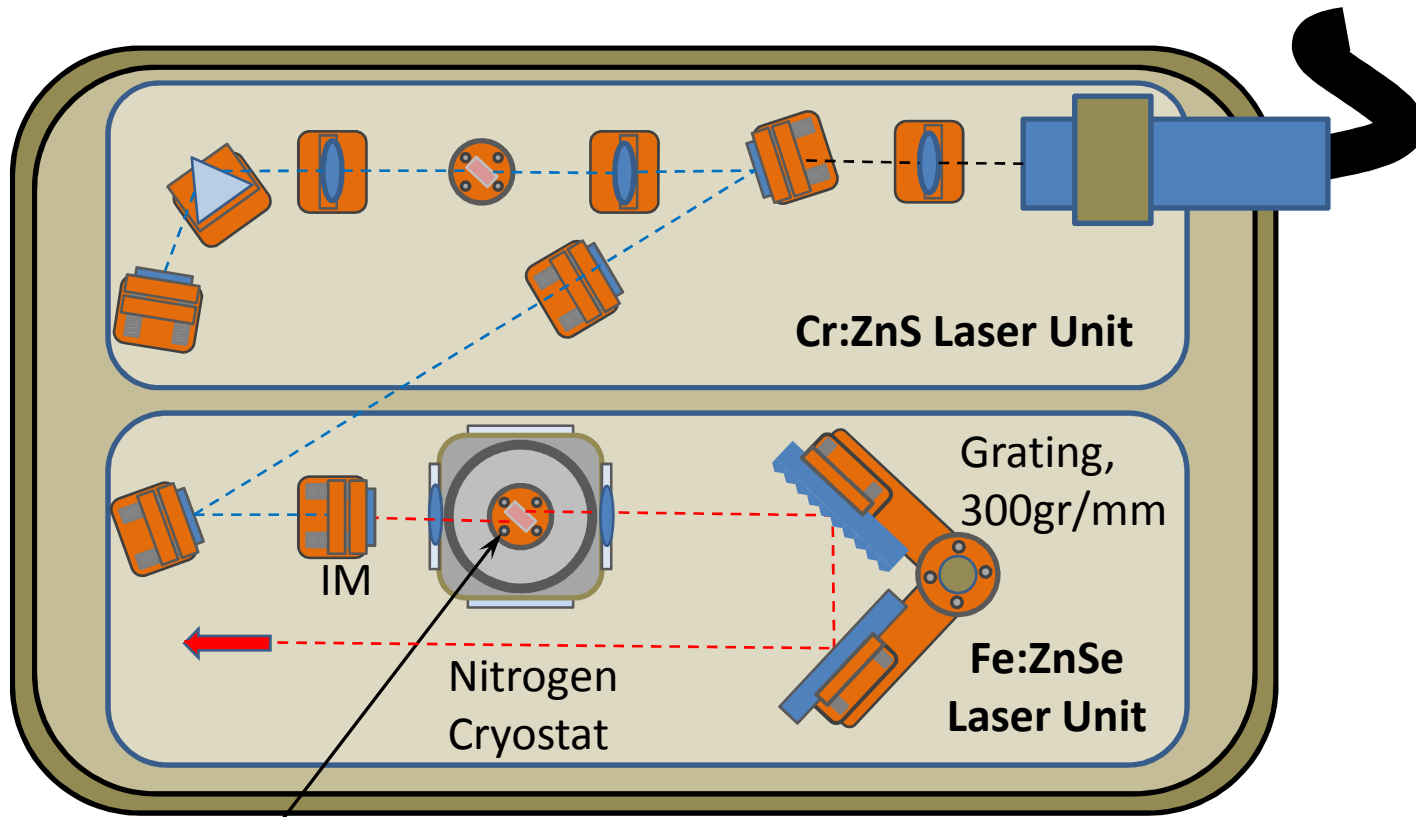
CW Tunable Fe:ZnSe Laser



CW Mid-IR Tunable Fe:ZnSe Laser

- Any wavelength within 3.7-4.8 μm
- Output Power > 300 mW

CW Tunable Fe:ZnSe Laser



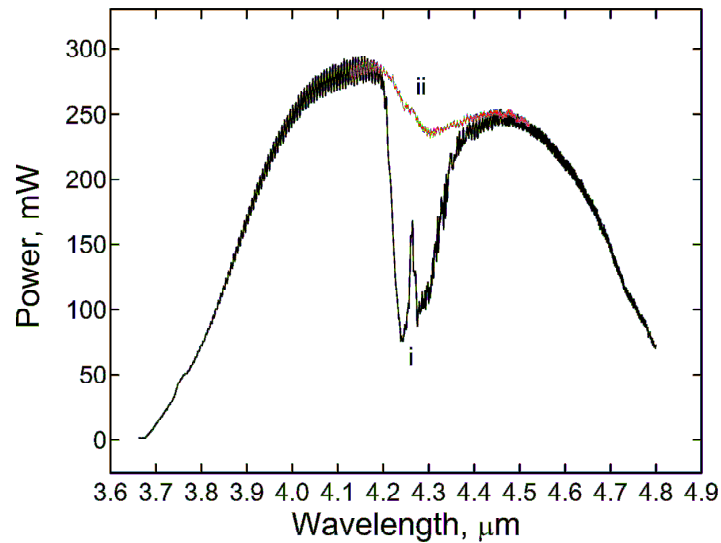
Fe²⁺ doped ZnSe
N=1.5x10¹⁹ cm⁻³



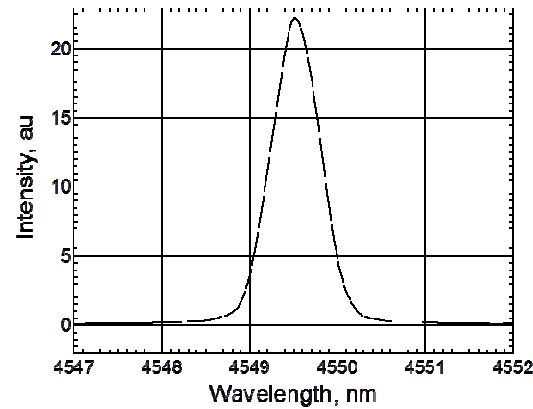
CW Tunable Fe:ZnSe Laser FLT-4200/100-0.3

Any wavelength within 3.7-4.8 μ m

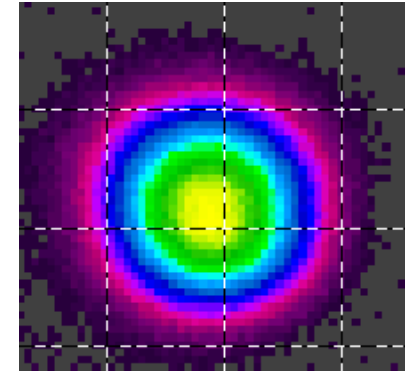
Output Power up to 1.5 W



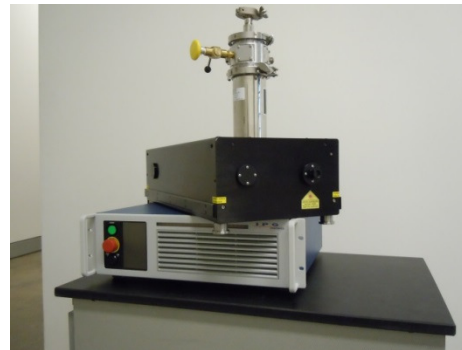
Typical tuning curve (i) in ambient atmosphere
(ii) purging with AR



Linewidth



Beam profile



Spectroscopic

Sensing

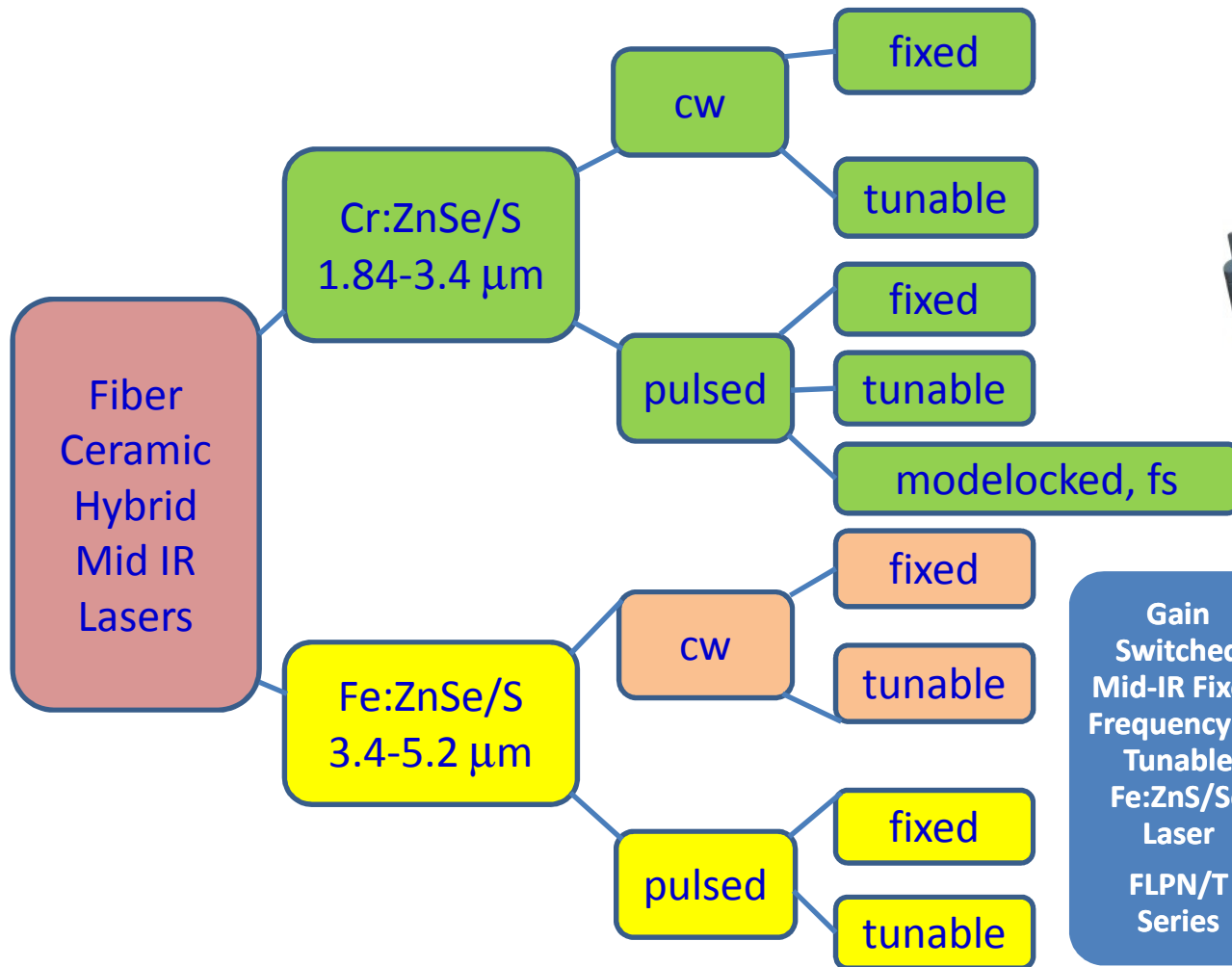
Medical

Defense

OPO seeding or pumping



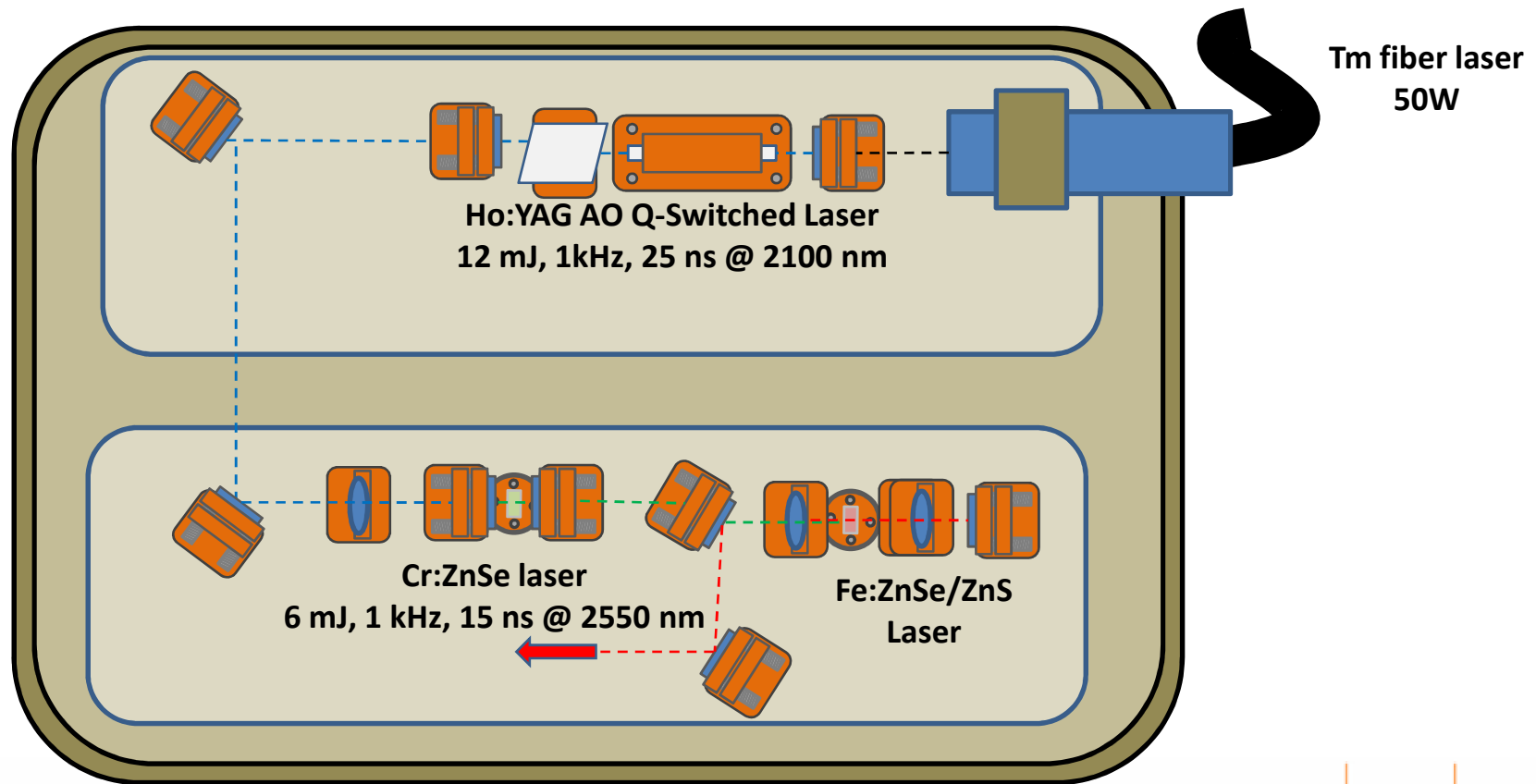
Gain Switched Mid-IR Fixed Frequency or Tunable Fe:ZnS/Se Lasers



Gain Switched Mid-IR Fixed Frequency or Tunable Fe:ZnS/Se Laser FLPN/T Series

- Output wavelength within 3.6-5.0μm
- Output Energy >0.5 mJ
- Pulse Duration 2-20 ns
- Repetition rate 0.1-1 kHz
- TEM₀₀

Gain Switched Mid-IR Fixed Frequency or Tunable Fe:ZnS/Se Lasers



Gain Switched Fixed Frequency or Tunable Fe:ZnS/Se Lasers FLPN-3950-1 Series

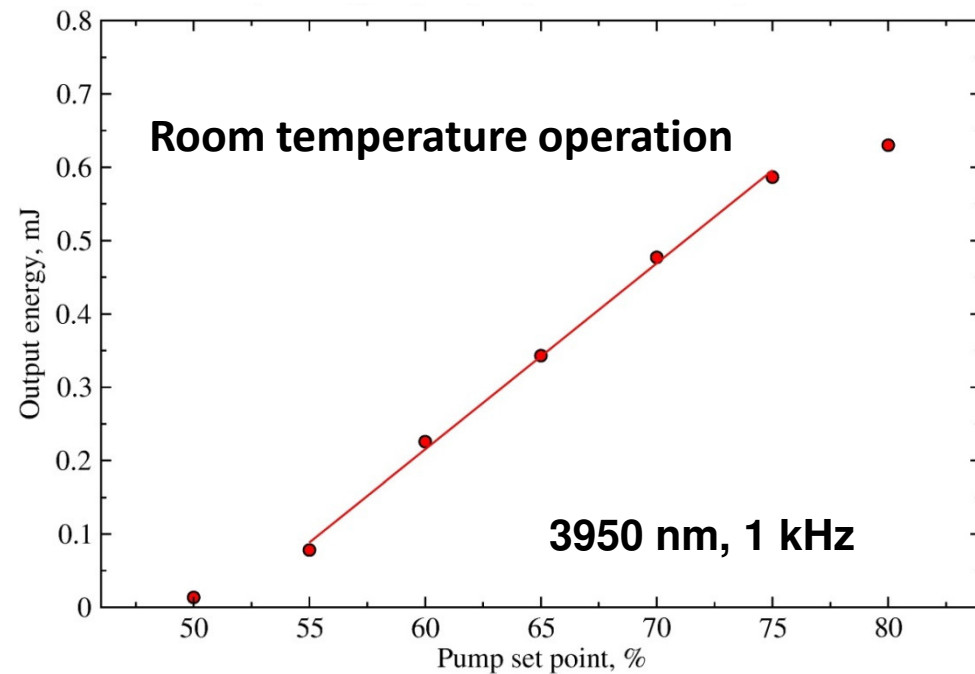
Output wavelength within 3.6-5.0 μ m

Output Energy >0.5 mJ

Pulse Duration 2-20 ns

Repetition rate 0.1-1 kHz

TEM₀₀



Medical

Materials Processing

Environmental Monitoring

Sensing

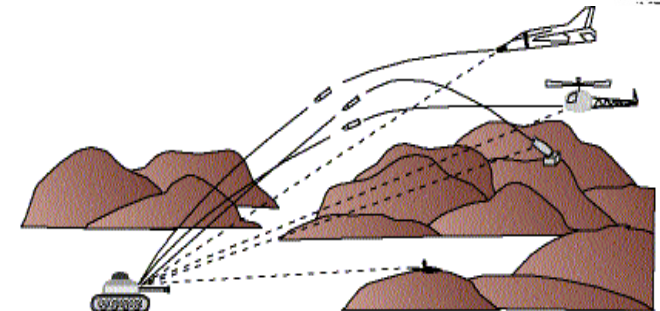
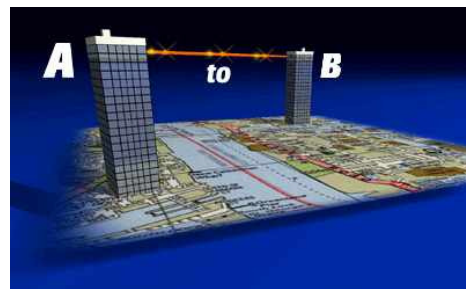
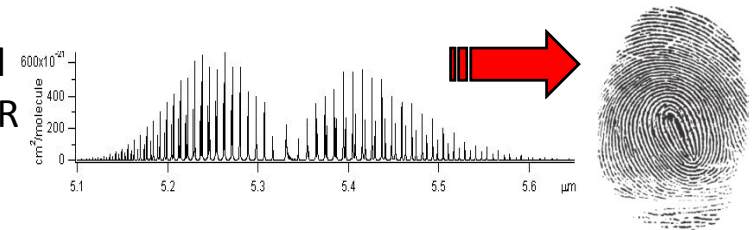
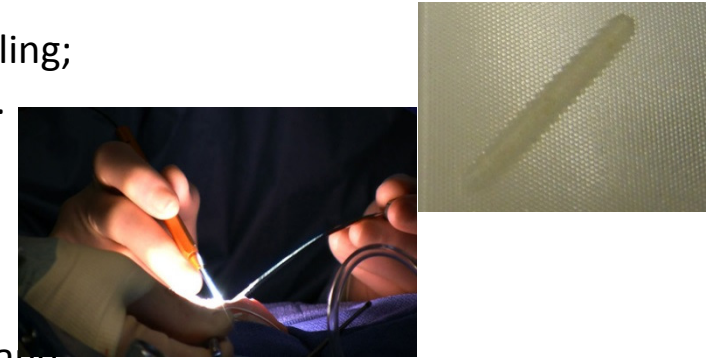
Industrial Process Control

MALDI Mass Spectroscopy



Mid IR Applications

- **Materials processing:** plastics cutting, welding, marking, drilling; forming of plastics, curing of coatings, ICs lift-off from Si sub.
- **Medical:** diagnostic, therapeutic, surgical
- **Spectroscopy:** molecular identification and dynamics; noninvasive nondestructive measurements; chemical agent and biomolecular sensing/ detection
- **Sensing and Imaging:** bioimaging, art imaging, hyperspectral imaging, thermography, tracking/ homing, night vision, LIDAR
- **Defense:** infrared countermeasures, target illumination and designation
- **Meteorology / Climatology / Astronomy**
- **Communications**
- **Pumping OPG/OPO**
- **X- ray Generation**



Conclusions and Outlook

- ❑ Chromium and Iron doped ZnSe and ZnS lasers have come of age, and, arguably, represent nowadays the most effective route for lasing over 1.9-5 μm spectral range with multi-watt average power, peak power up to 1 GW, multi-Joule energy and pulse durations up to 40 fs.
- ❑ Future progress of Cr- and Fe-doped II-VI lasers in terms of extending spectral coverage over 3-4 μm and 5-9 μm depends on a search of new, low-phonon-cutoff Cr- and Fe-doped binary and ternary II-VI semiconductors bulk materials.

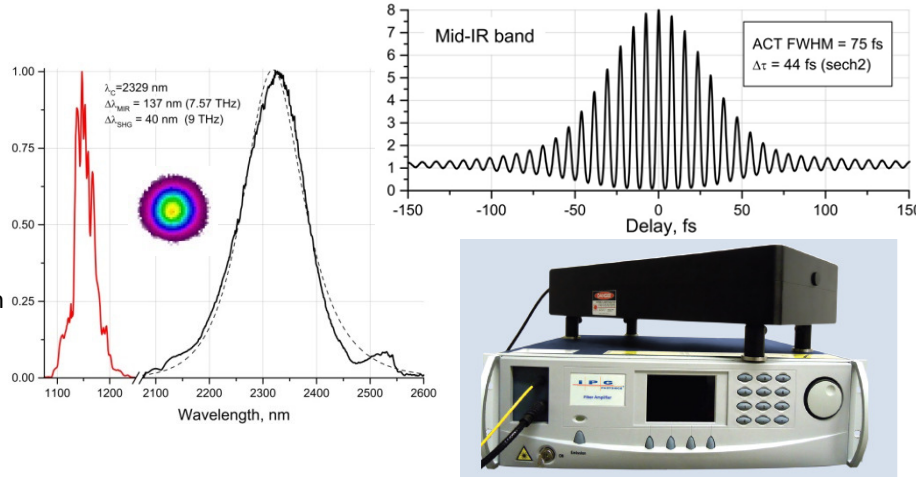
Conclusions and Outlook

- ❑ Future improvements in output power will depend on new schemes of thermal management of gain elements including utilization of fiber, waveguide and disk geometry.
- ❑ Future Improvements in technology of hot-pressed ceramic II-VI compounds can stimulate design flexibility of the laser elements with high optical quality (undoped ends, , gradient of dopant concentration, etc) important for development of efficient, high performance lasers with output power and energy scaled-up to hundreds of Watts and tens of Joules.

Thank you for your attention!

Femtosecond Mid-IR Laser Model CLPF-2400-20-60-2

- Output power up to 2W
- Pulse duration from 50 fs
- Custom fixed central wavelength
- Wavelength tuning
- SHG option (up to 0.3W)
- Power amplifier Option



High Power Narrow Line Cr²⁺:ZnS/Se Tunable Laser

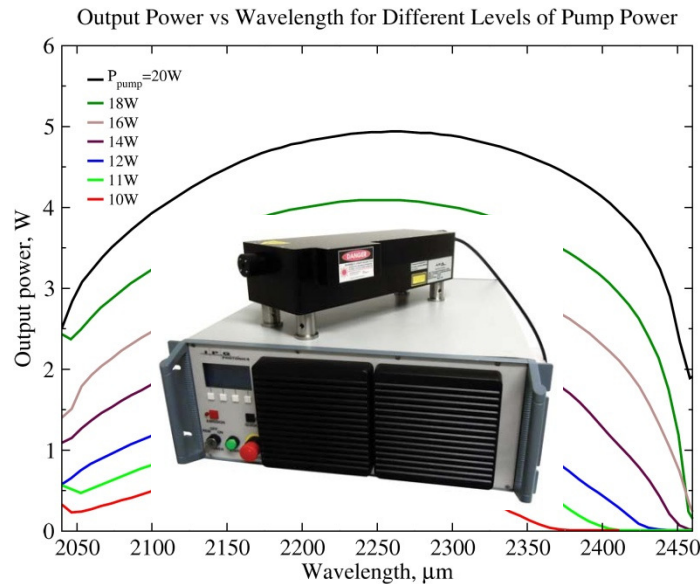
MODEL: CLT--2400/1000-4

- Multi-Watt (20W) Output Power
- Narrow Linewidth, < 0.1 nm available
- Large tuning range with a single set of optics (any wavelength within 2000 - 3000 nm tuning range is available)

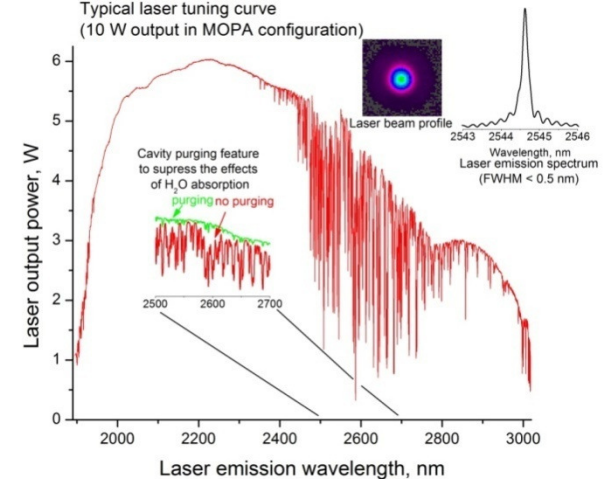
High Power, Tunable, Single Frequency Mid-IR Product Line

Model CLT-2200/500-5

- Multi-Watt Output Power
- Narrow Linewidth, <10 MHz available
- Large tuning range with a single set of optics (any wavelength within 2000 - 3000 nm tuning range is available upon request)



Typical laser tuning curve
(10 W output in MOPA configuration)



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