

# Terahertz Materials Characterization in Extreme Environments

Emerging Tools for Materials Research

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# Materials Characterization Systems

Systems

## Magnetometers (VSM/AGM)



Measure magnetic properties:

- Hysteresis  $M(H)$  loops
- FORC curves
- Temperature dependencies

**Permanent Magnets**  
**High Density Recording Media**  
**Mineral Magnetism**

## Hall Effect Measurement Systems (HMS)

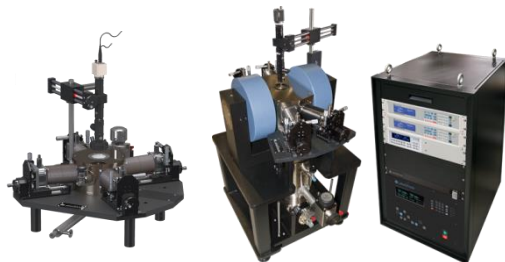


AC/DC measurements:

- Hall mobilities
- Carrier concentration
- Carrier type

**Thermoelectrics**  
**Solar Cells**  
**HEMTs**

## Probe Stations



Measure electrical  
properties of devices and  
materials in temperature-  
controlled environment

**Nanowires**  
**Magnetoresistance**  
**Organic electronics**

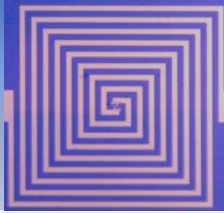
## THz Systems



Fully integrated for material properties:

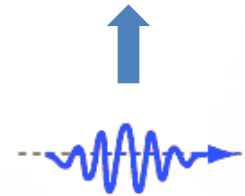
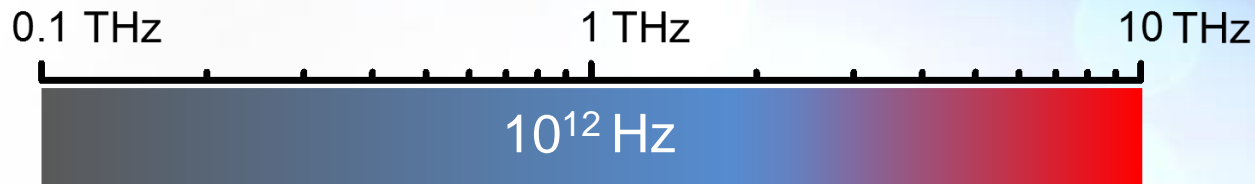
- Electronic
- Magnetic
- Chemical

**Electronic thin films**  
**Organic Electronics**  
**Spintronics**



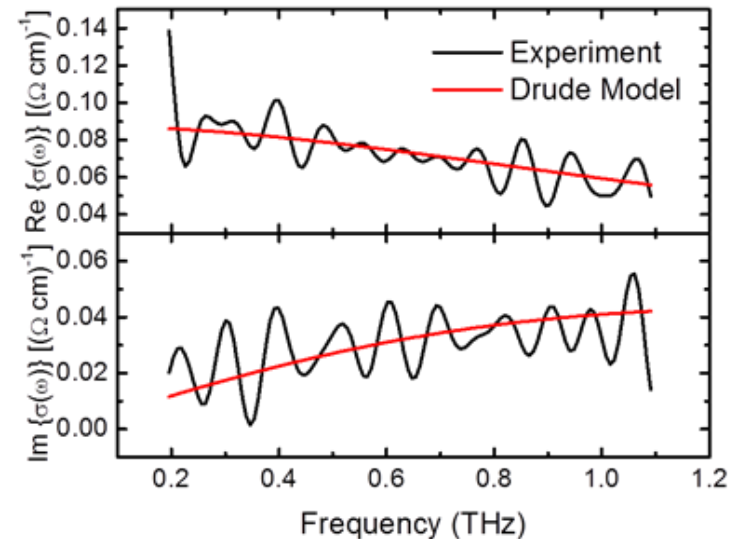
# Why THz?

# Equivalent Scales for THz



2.5 THz ~ 10 meV

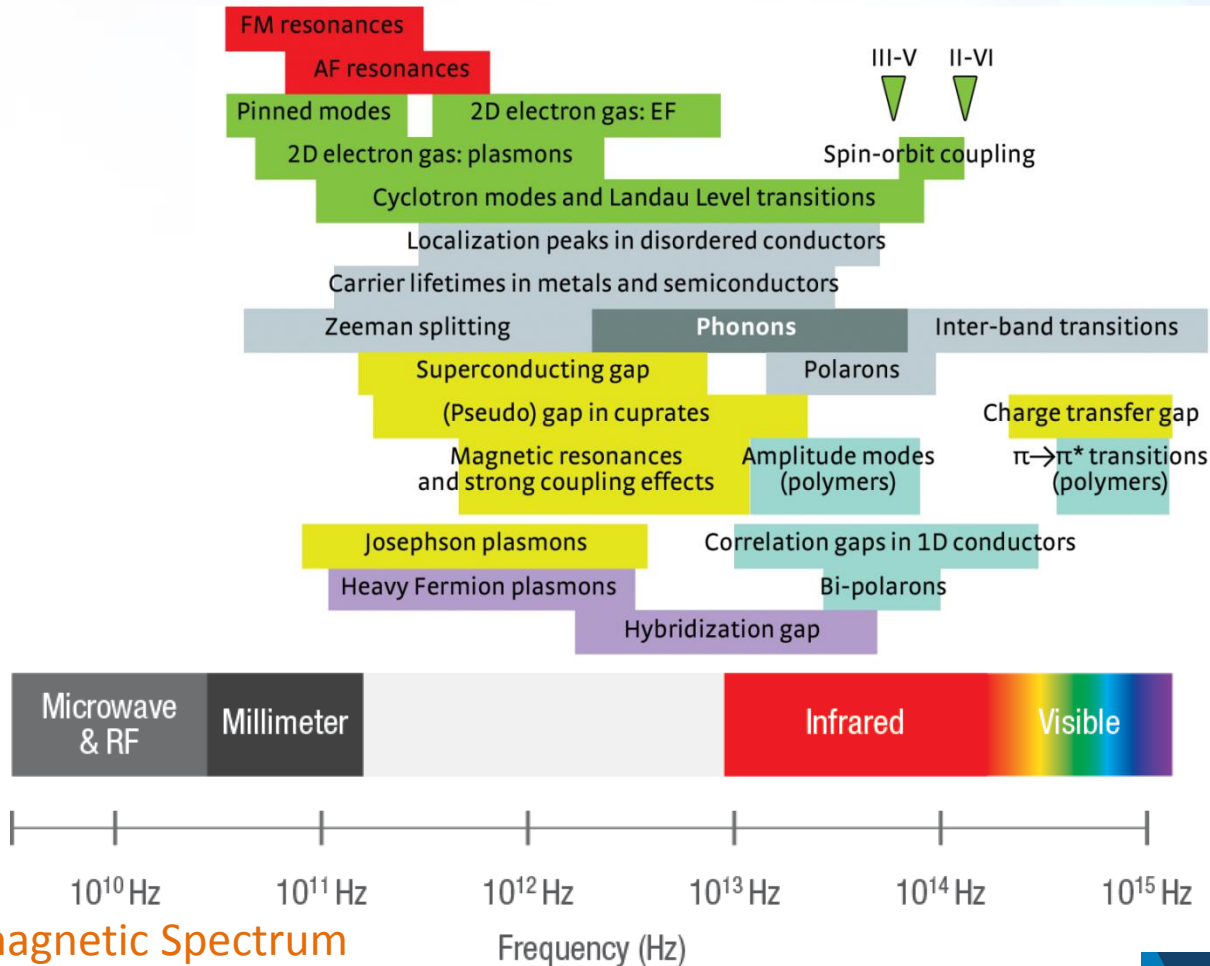
- Alignment of THz energy levels with phenomena of interest
  - Vibrational Resonances
  - Novel Spin Resonances
  - Free Carrier motion in semiconductors



# Materials Phenomena

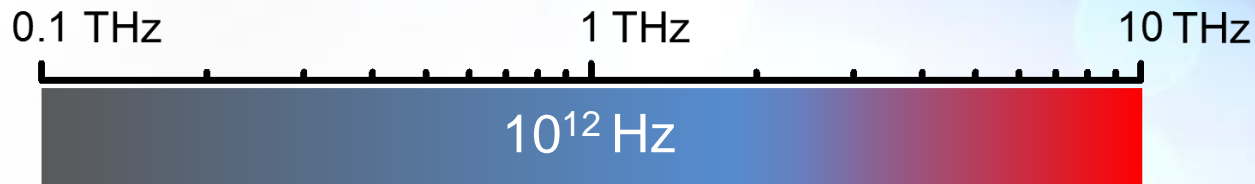


D. N. Basov, et al.  
 “Electrodynamics of correlated electron materials”  
 Rev. Mod. Phys. 85(2), 471 (2011).



## The Electromagnetic Spectrum

# Equivalent Scales for THz

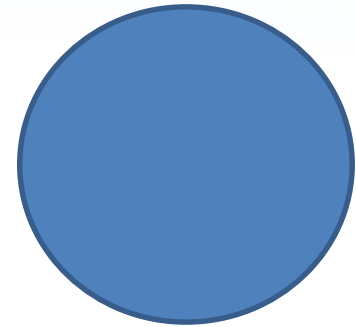


↑  
 $\lambda$

0.6 THz ~ 0.5 mm

- THz wavelengths match the feature sizes of development-grade electronic materials
  - Non-contact electronic characterization
  - Novel magnetic materials

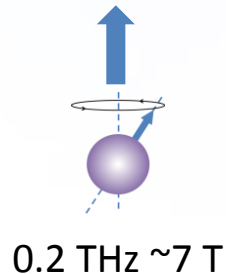
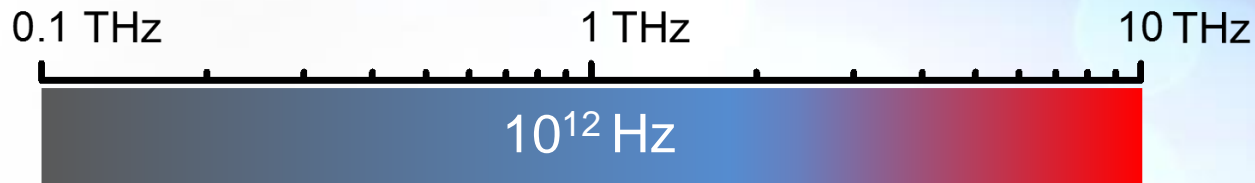
↔ 2" ↔



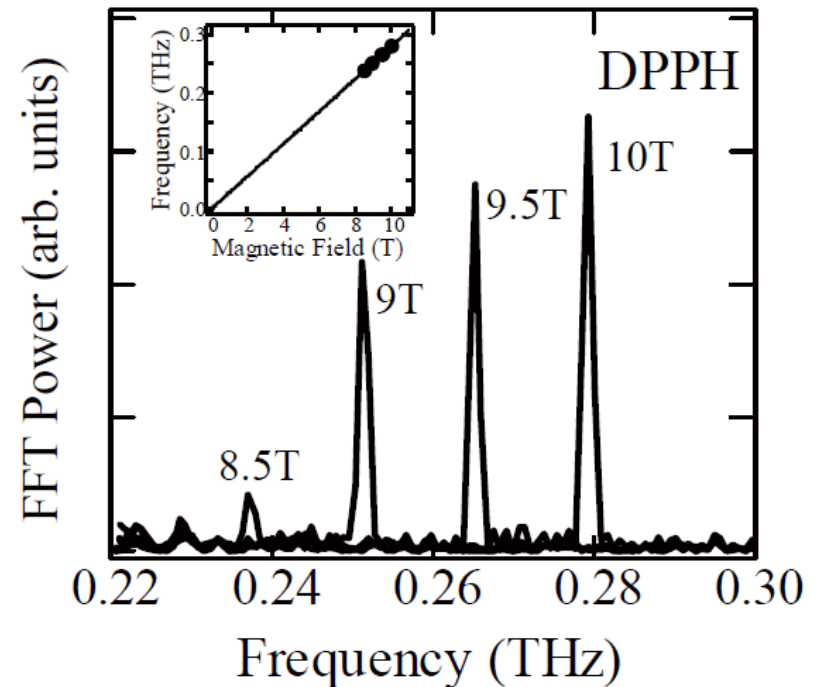
↔ 10 mm ↔



# Equivalent Scales for THz



- Coupling to spin-based materials at THz frequencies
  - High speed computing paradigms
  - May require large fields

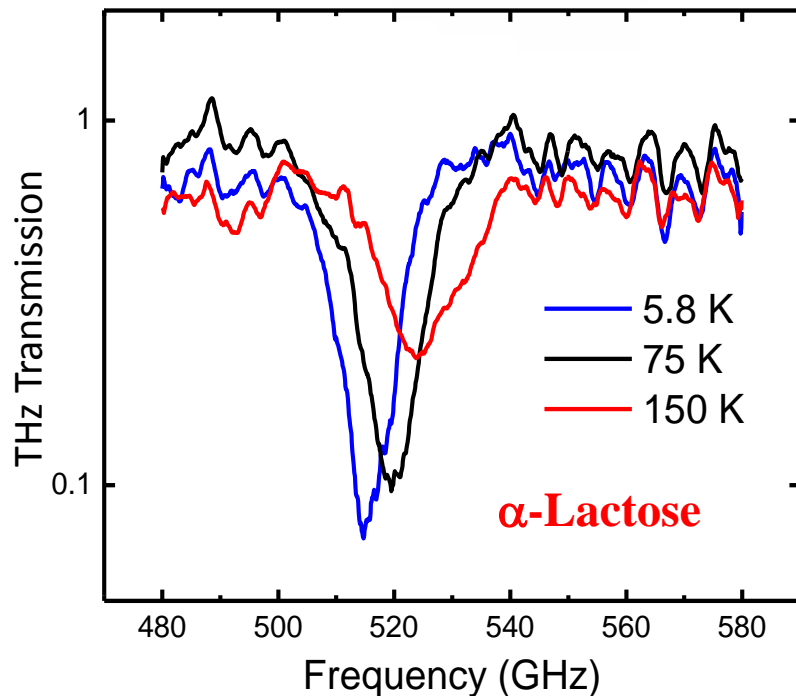


Nagashima, T.; Nishitani, J.; Kozuki, Kohei, "Lasers & Electro Optics & The Pacific Rim Conference on Lasers and Electro-Optics, 2009. CLEO/PACIFIC RIM '09. Conference on , vol., no., pp.1,2, 30-3 Aug. 2009

# Equivalent Scales for THz



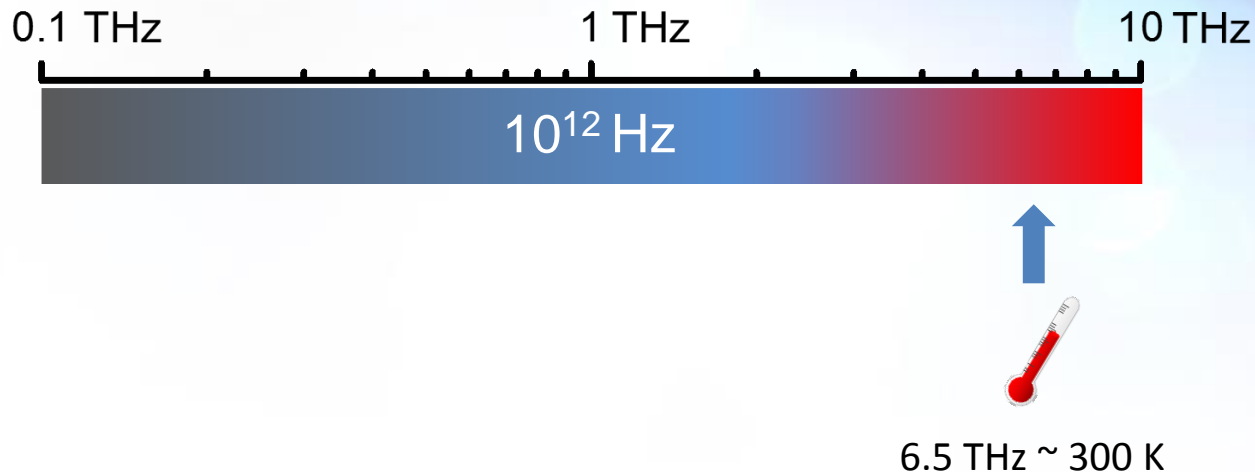
6.5 THz ~ 300 K



- Cryogenic temperatures
- Homogeneous broadening of vibrational resonances

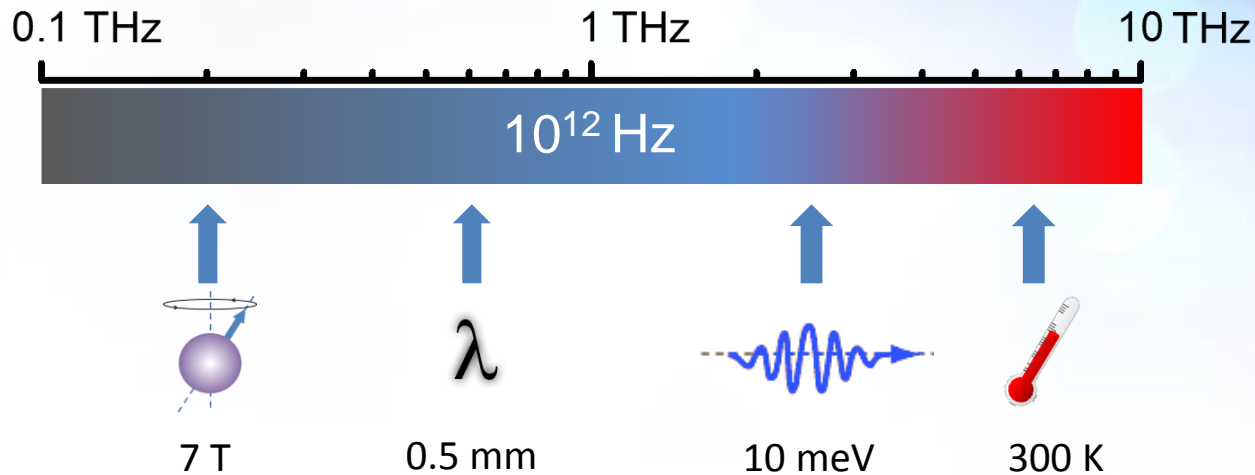


# Equivalent Scales for THz



- Cryogenic temperatures
  - Homogeneous broadening of vibrational resonances
  - Temperature dependent carrier concentration and mobility

# Equivalent Scales for THz



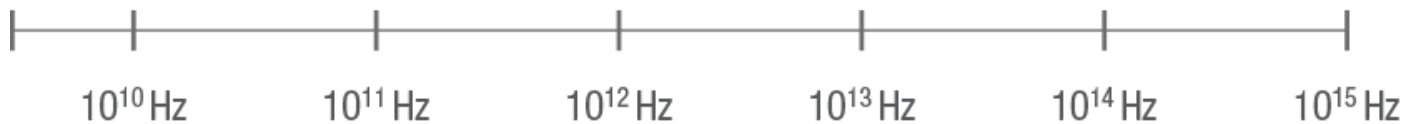
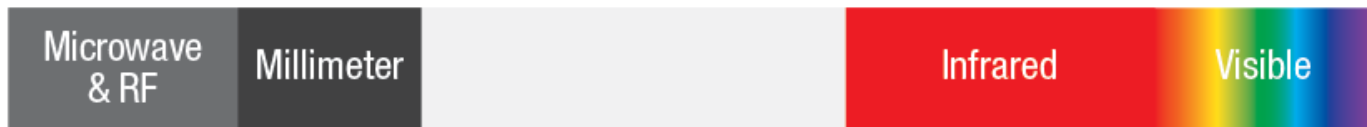
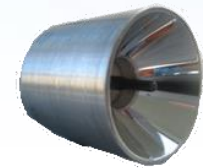
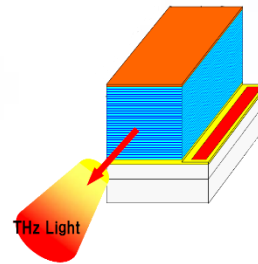
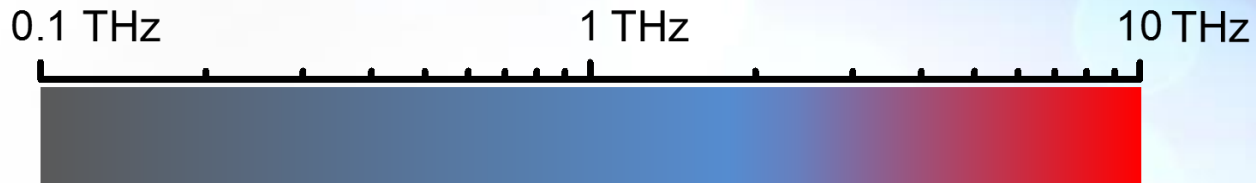
- Alignment of THz energy levels with phenomena of interest
- THz wavelengths match the feature sizes of development-grade electronic materials
- Coupling to spin-based materials at THz frequencies
- Cryogenic temperatures

# THz System Product Challenges



- THz performance suitable for materials characterization
  - Continuous tuning over bandwidth
  - Spectral resolution
- Ability to characterize samples while exposed to variable cryogenic temperatures and magnetic fields
- Affordable THz-based measurement platform
- Robust Design
- Proceduralized experimental methods and reliable analysis of the spectral results
  - Convenient sample insertion without complicated alignment
  - Enable materials developers to rapidly begin productive research

# Filling the THz Gap



## The Electromagnetic Spectrum

Frequency (Hz)

# THz with cryogenics & magnetics



- Many materials of interest for THz require variable temperature and/or high magnetic fields
- Optical cryostats are the standard approach
  - THz generation outside the environment
  - Must pass optical energy through windows, cutting signal power and spectral distortion
- **Not ideal for THz spectroscopies**



# Variable Temperature and High Magnetic Field THz Platform

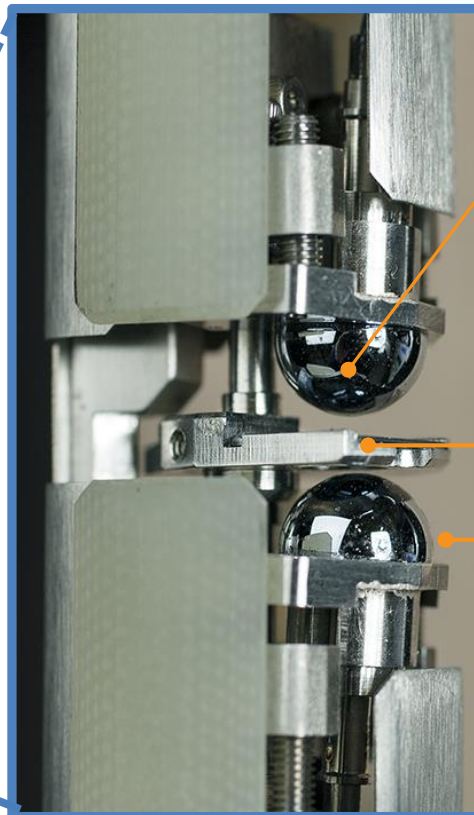
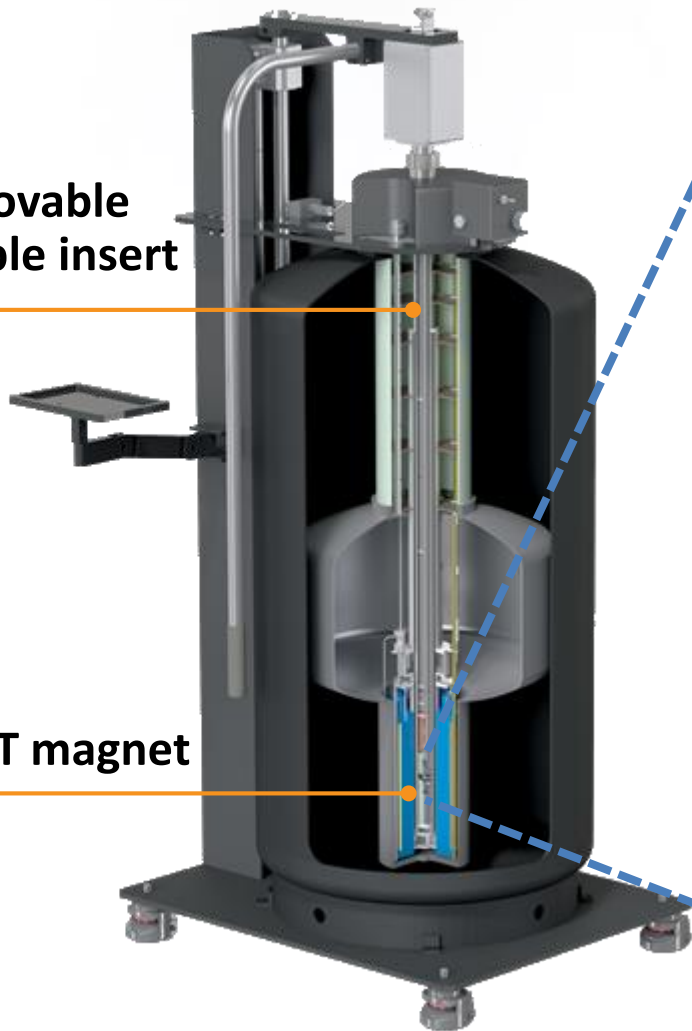


## Cryostat

## Sample Space

Removable sample insert

$\pm 9$  T magnet



Cryo-compatible THz emitter with collimating optics

Rotatable sample stage (10 mm  $\times$  10 mm)

Thermally stable optical alignment stages

# Integrated THz System Details



## Application software for turnkey operation

- Experiment setup/run
- Analysis of spectral data
- Calculation of material properties

## Fiber-based optical platform for CW THz spectroscopy

- CW THz spectrometer instrument
- Amplitude and phase detection from 200 GHz to 1.5 THz
- Spectral resolution under 500 MHz
- Circularly polarized light

## Integrated controls

- Model 336 cryogenic temperature controller
- Model 625 superconducting magnet power supply



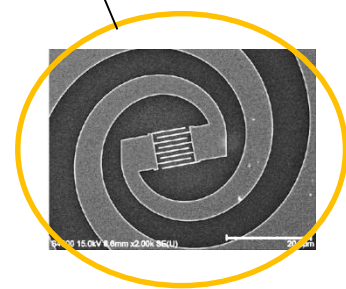
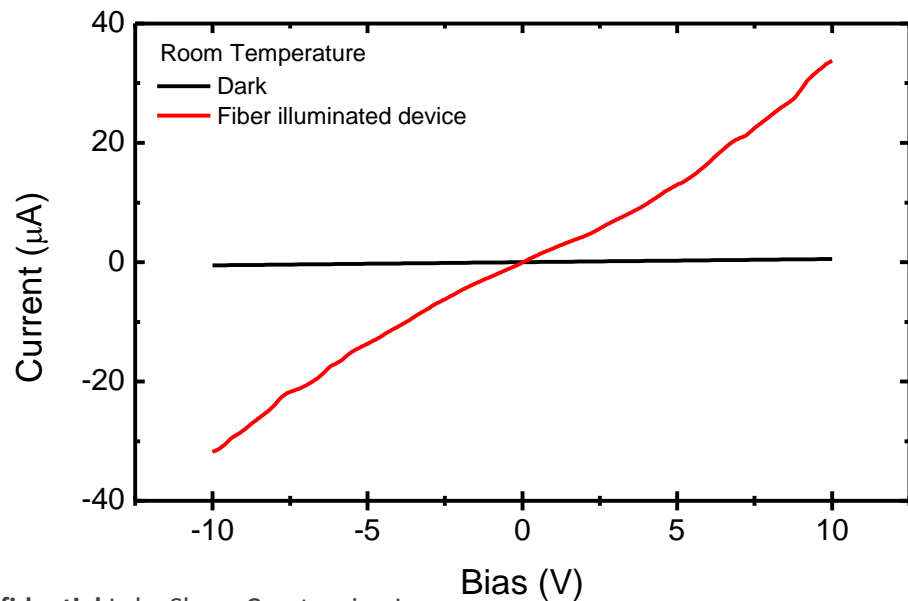
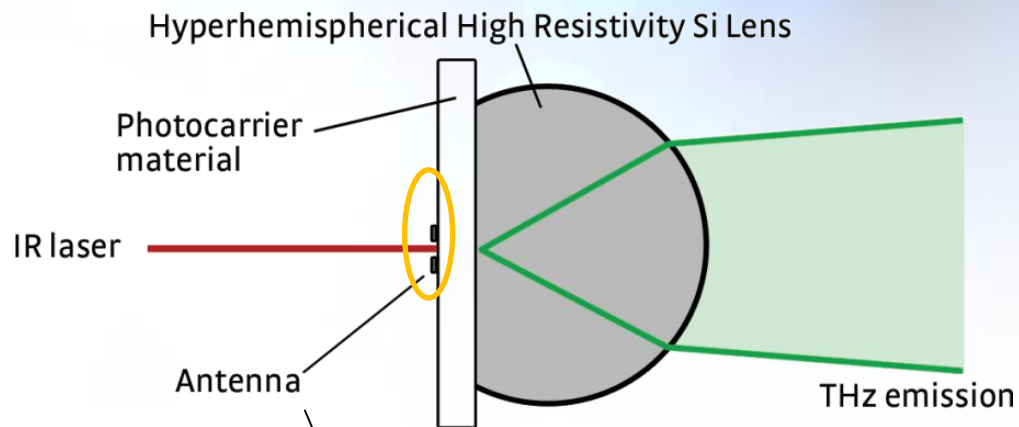
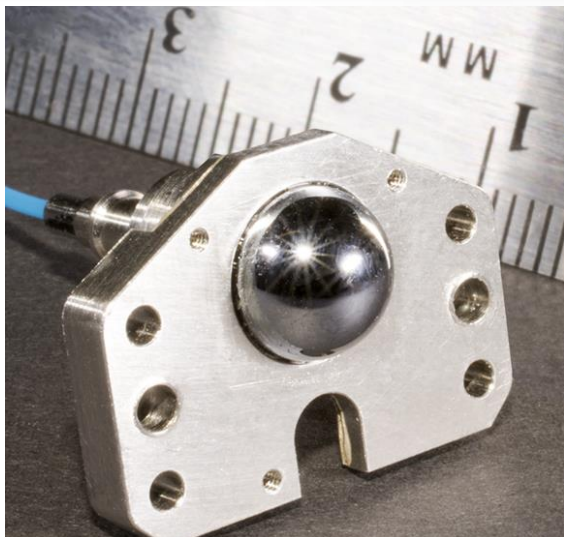
## Integrated cryostat & insert

- Variable temperatures from 5 K to 300 K
- Sample size – 10 mm
- Measurement – THz transmission

## Superconducting magnet

- High magnetic fields to  $\pm 9$  Tesla

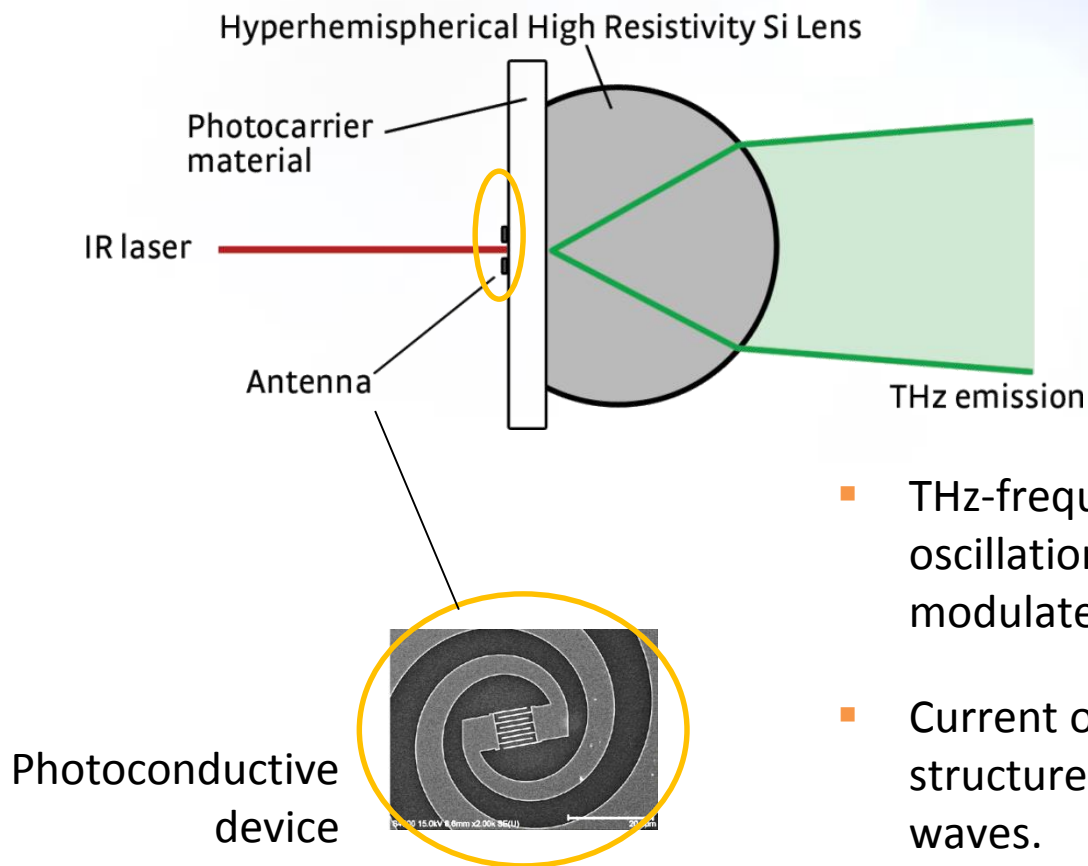
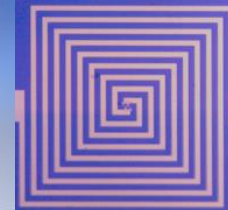
# Photoconductive THz Emitter



Photoconductive device

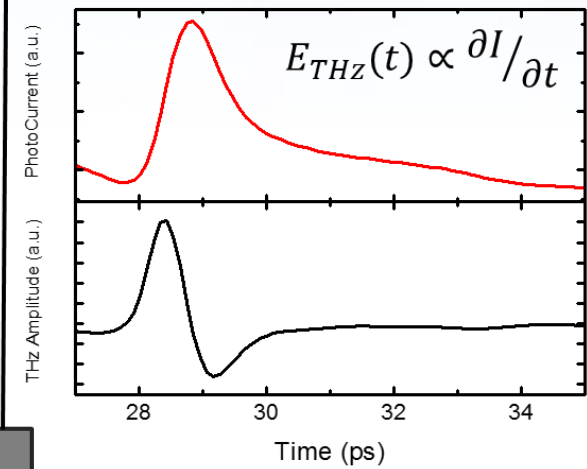
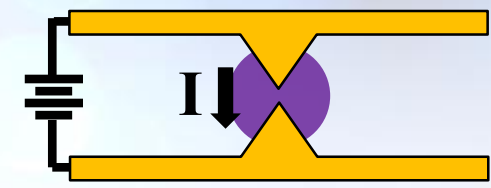
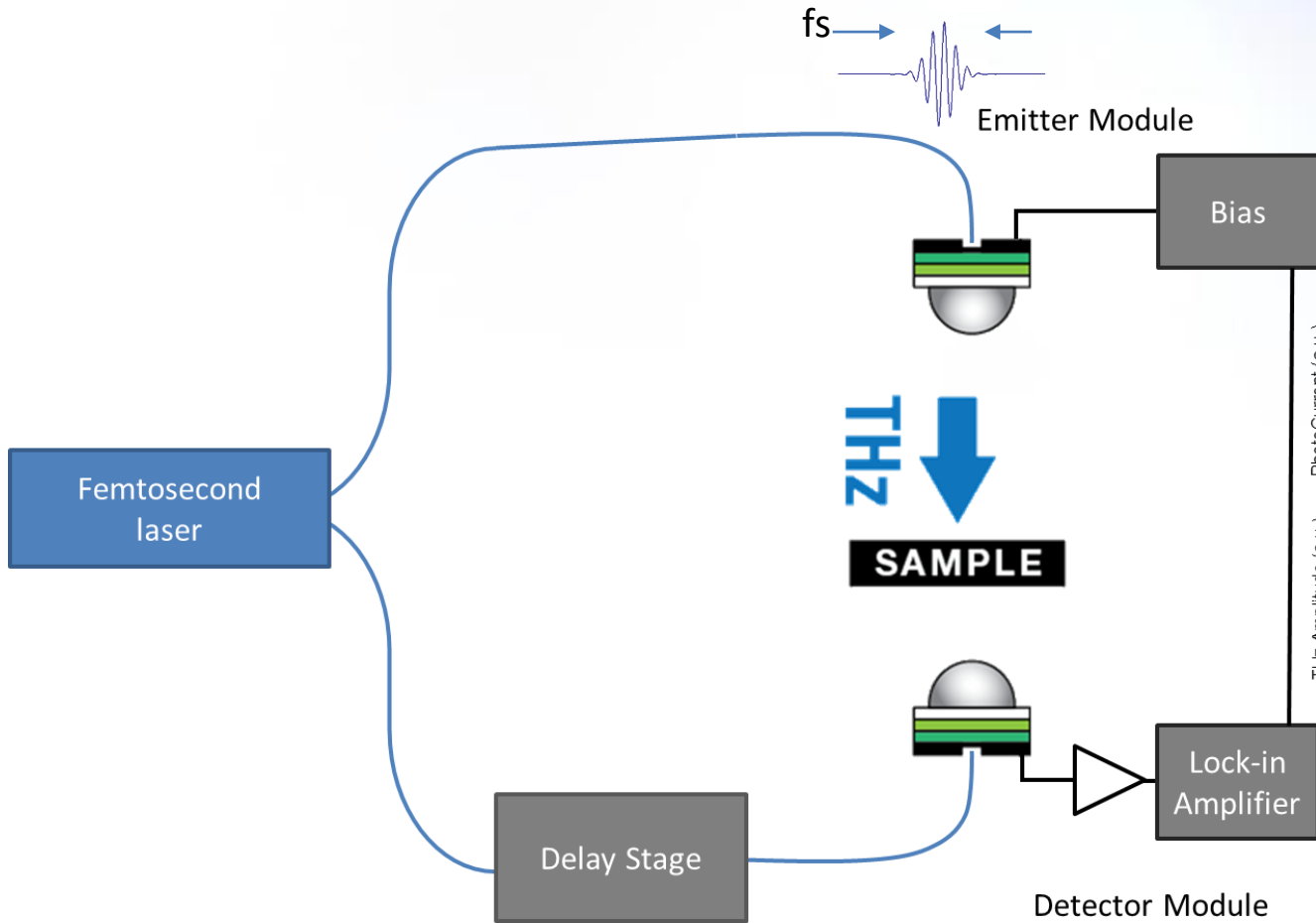


# Photoconductive THz Emitter

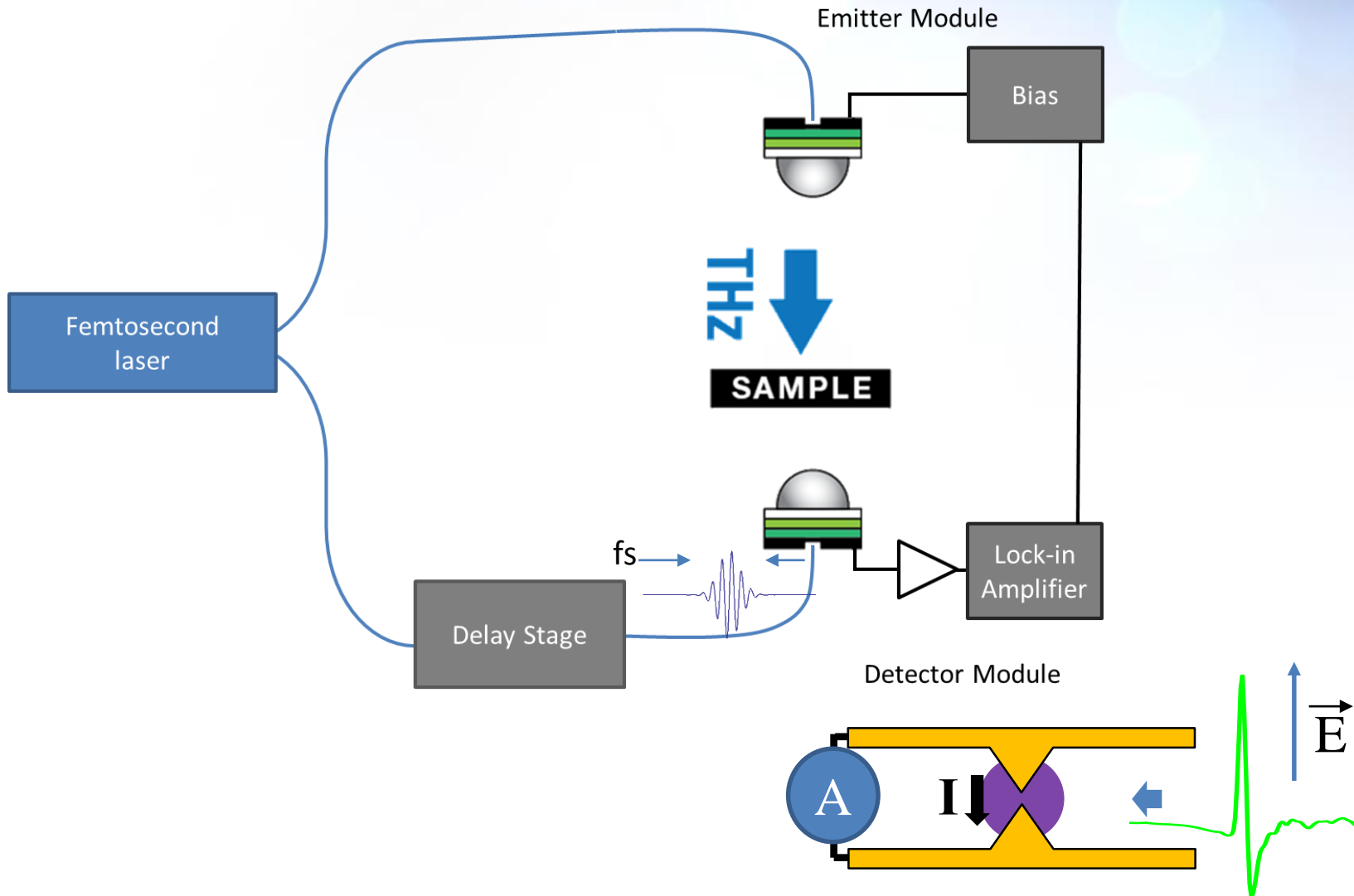


- THz-frequency transient current oscillations induced by amplitude modulated IR laser light
- Current oscillations in the antenna structure radiate propagating EM waves.
- A high resistivity Si lens is attached to the emitter device to couple the THz emission to free space.

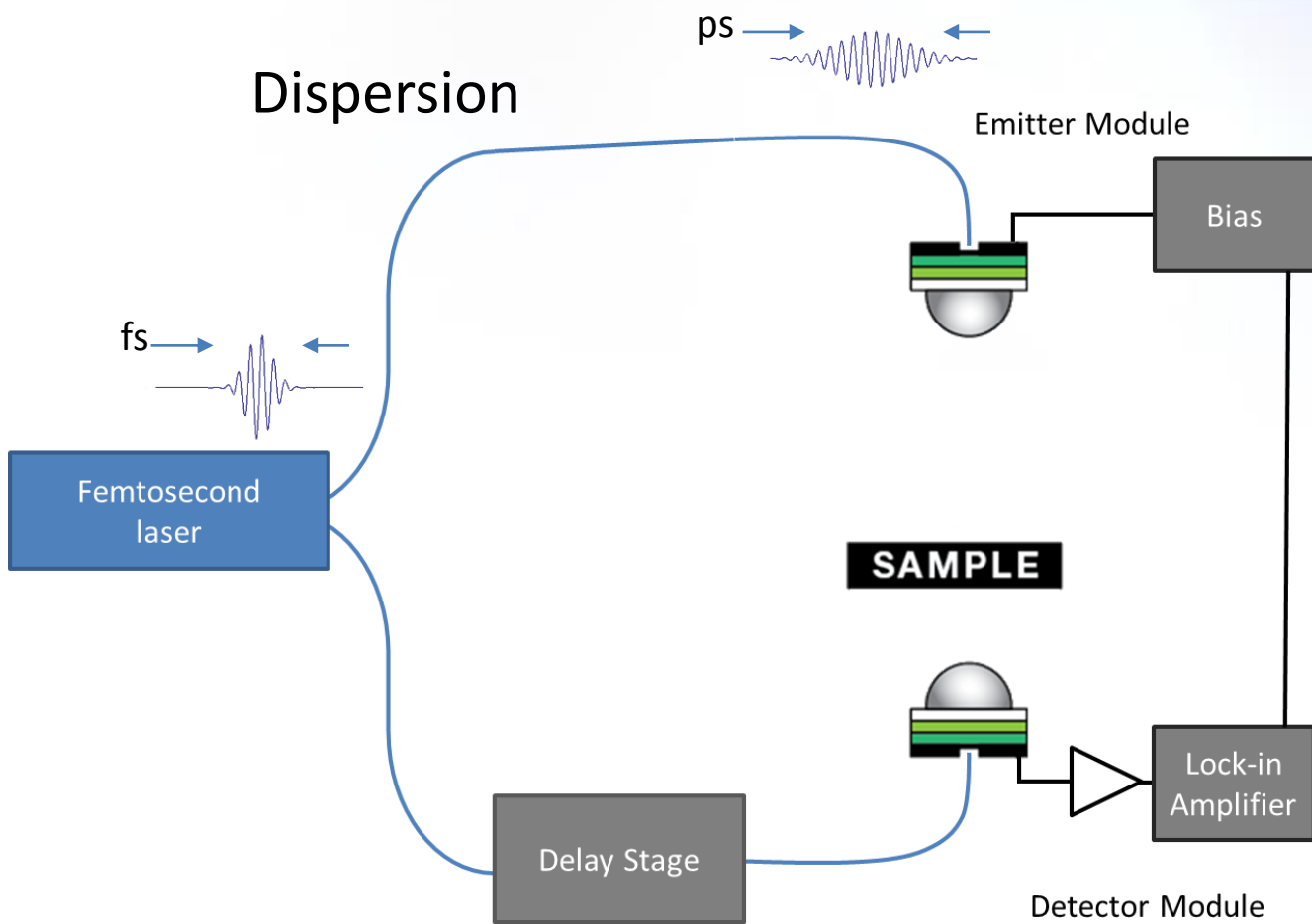
# THz Time Domain Spectroscopy



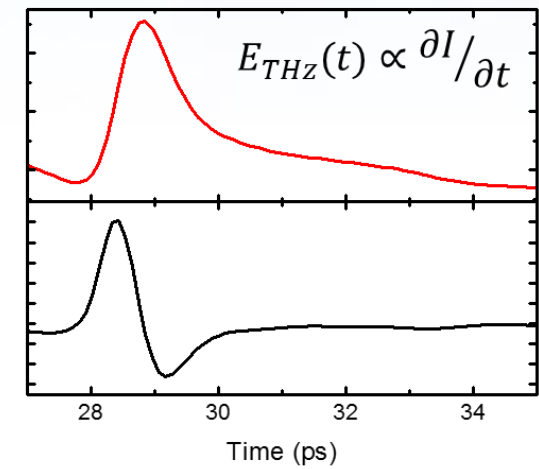
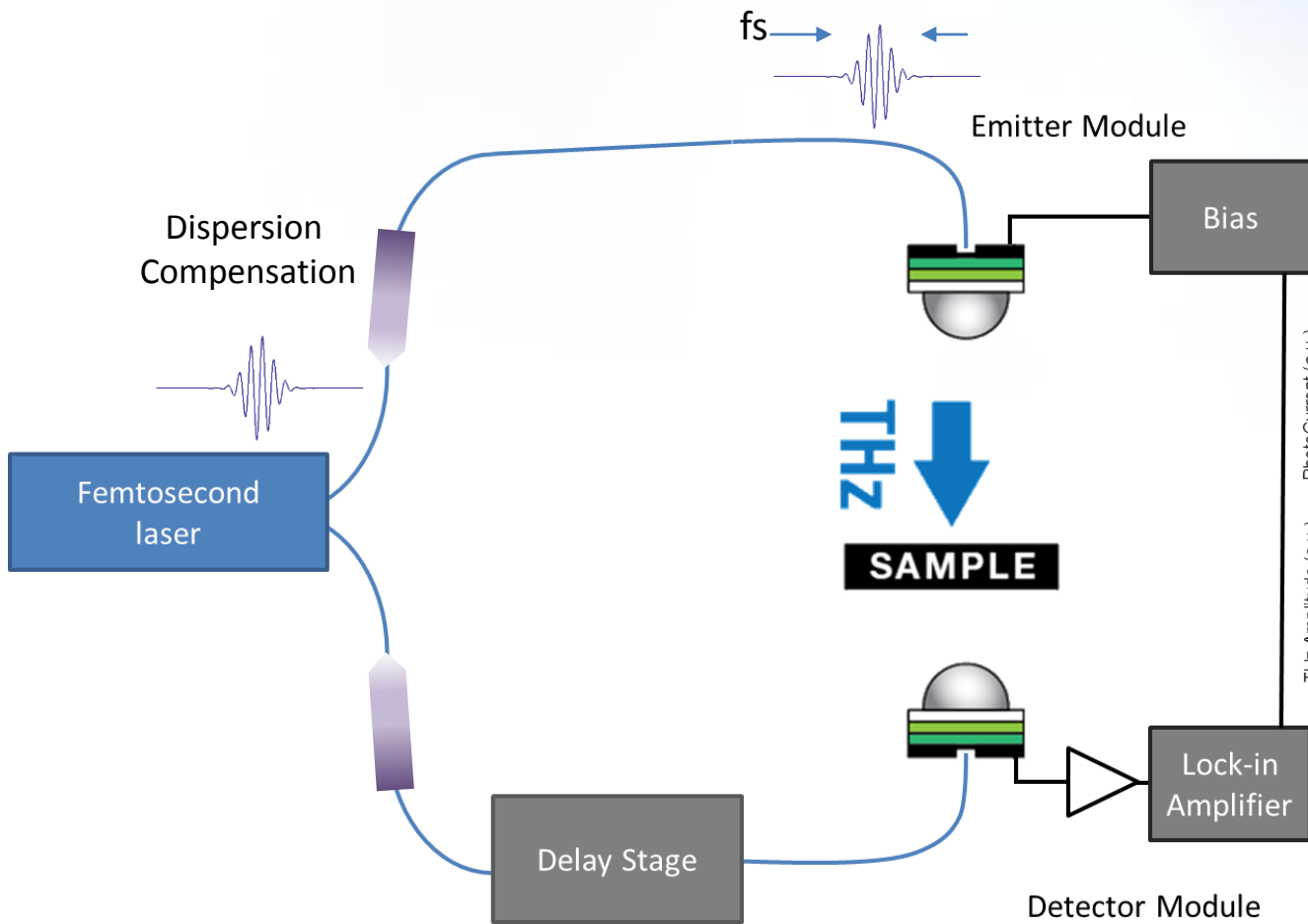
# THz Time Domain Spectroscopy



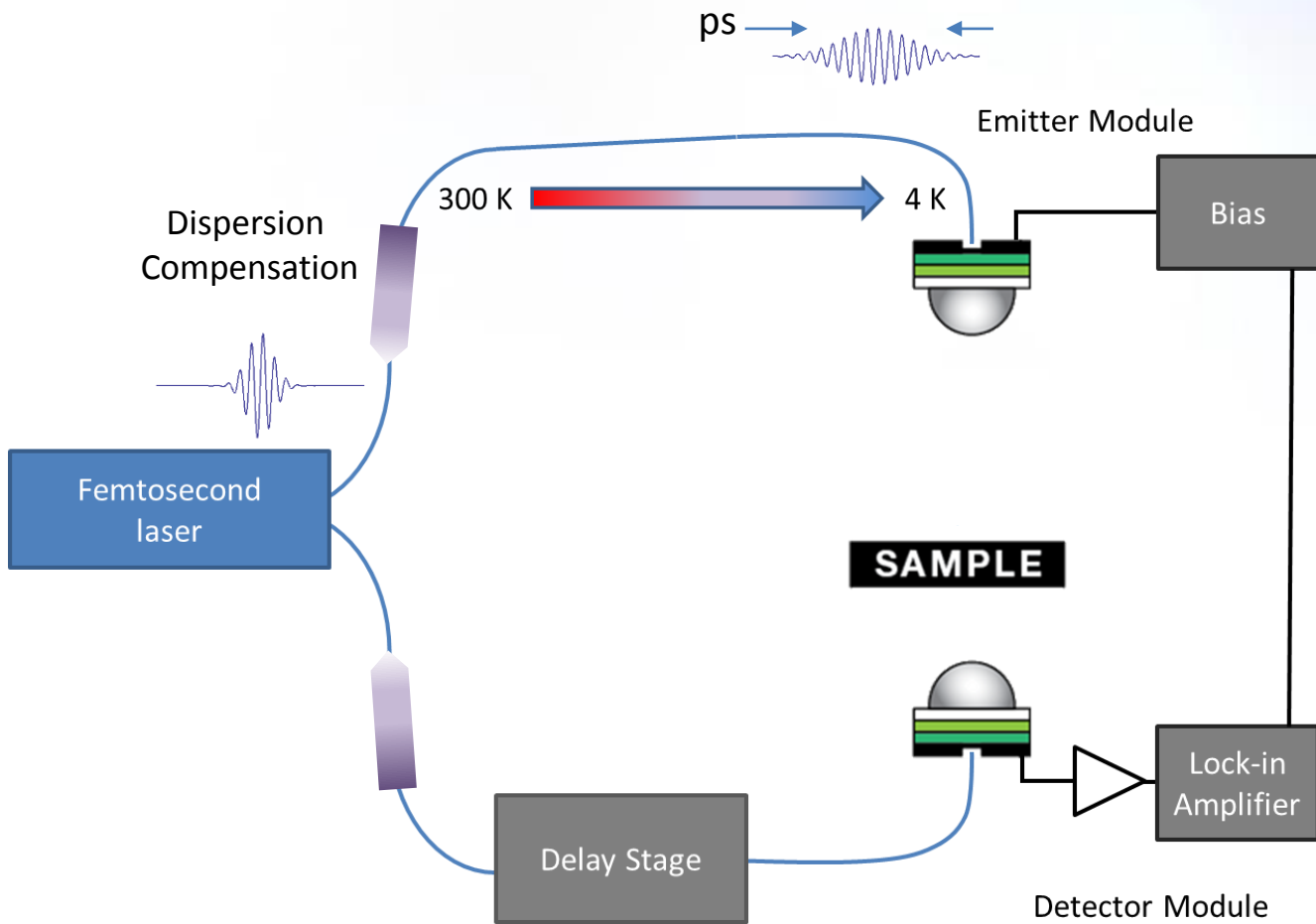
# THz Time Domain Spectroscopy



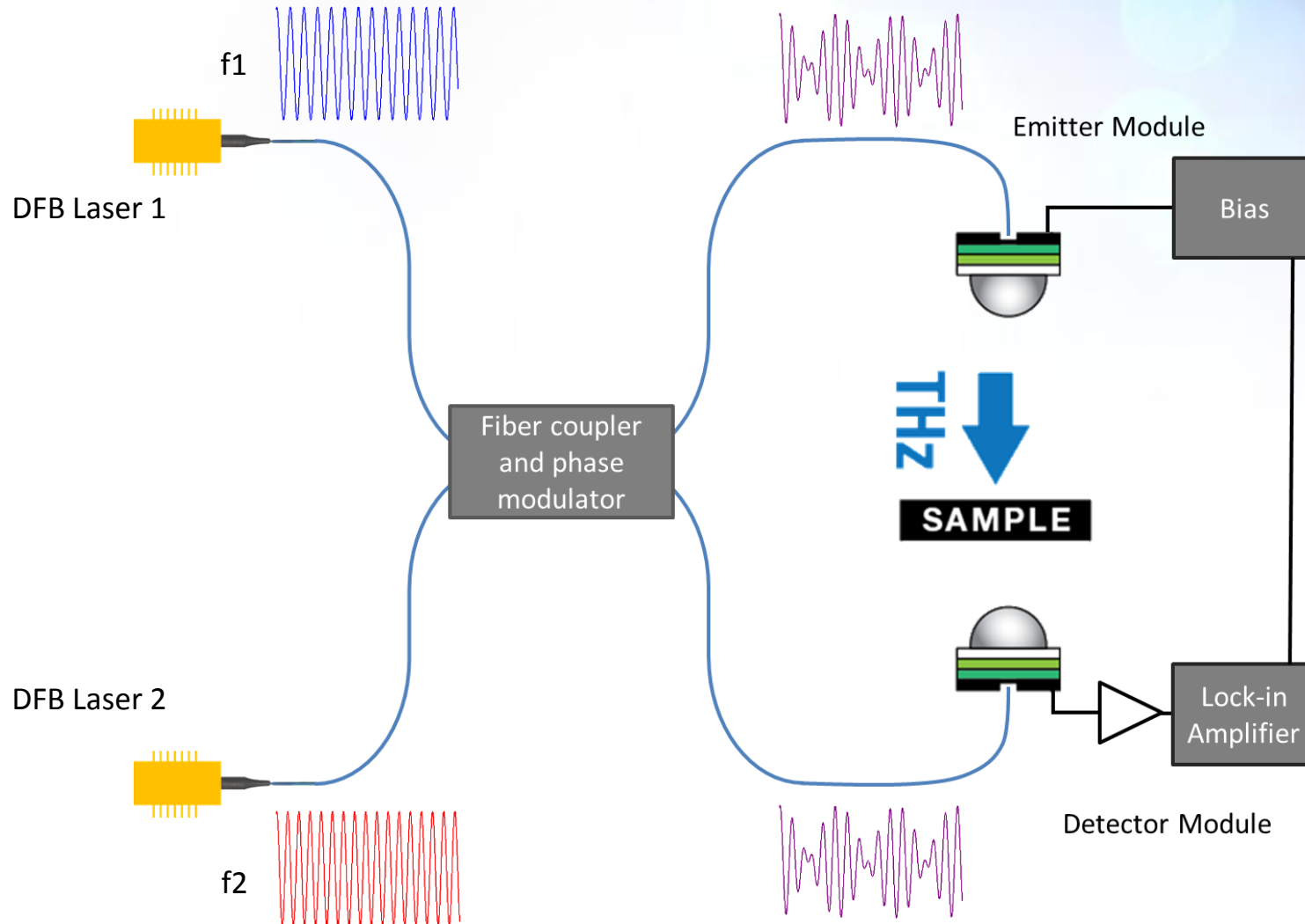
# THz Time Domain Spectroscopy



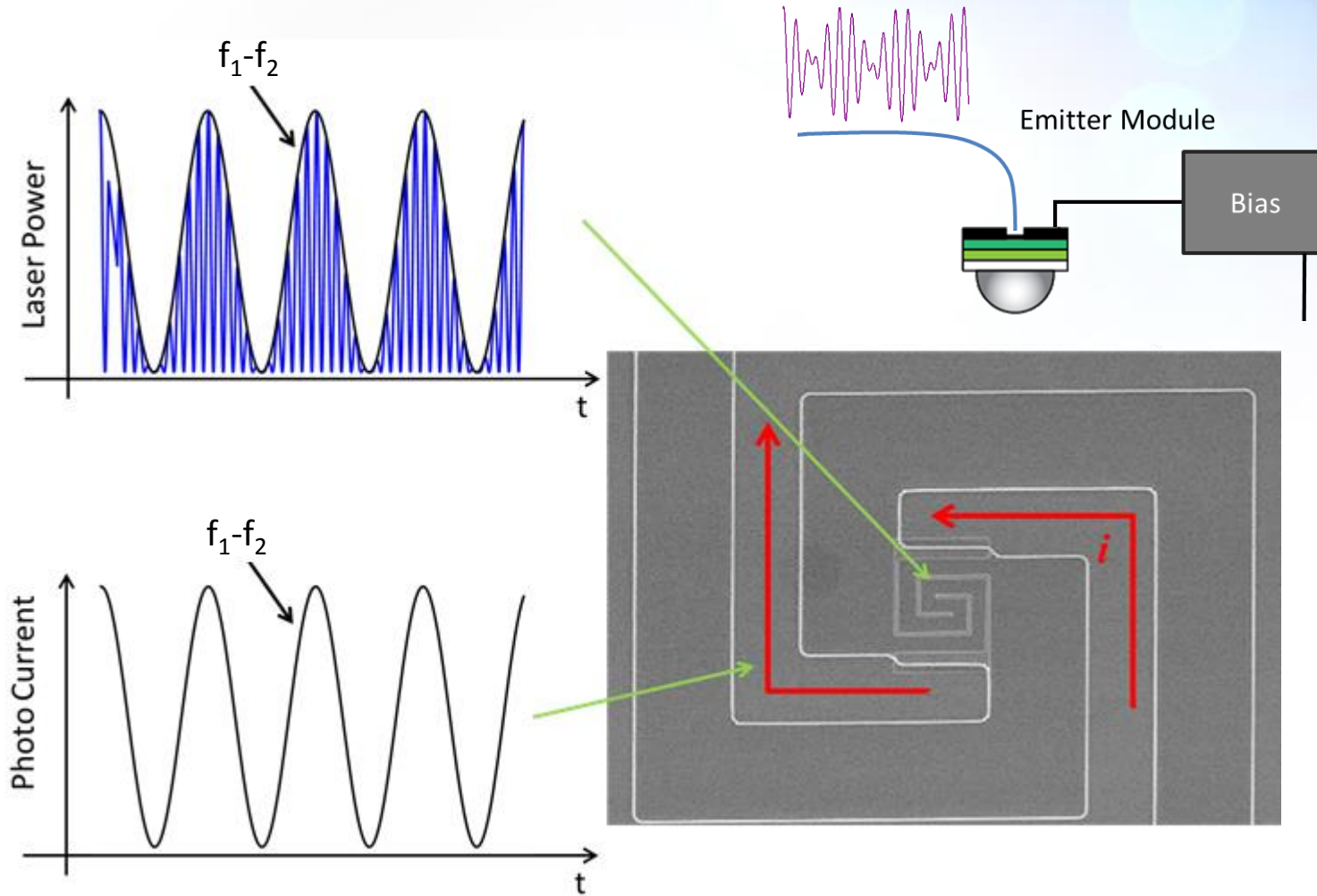
# THz Time Domain Spectroscopy



# Continuous-Wave THz Spectroscopy

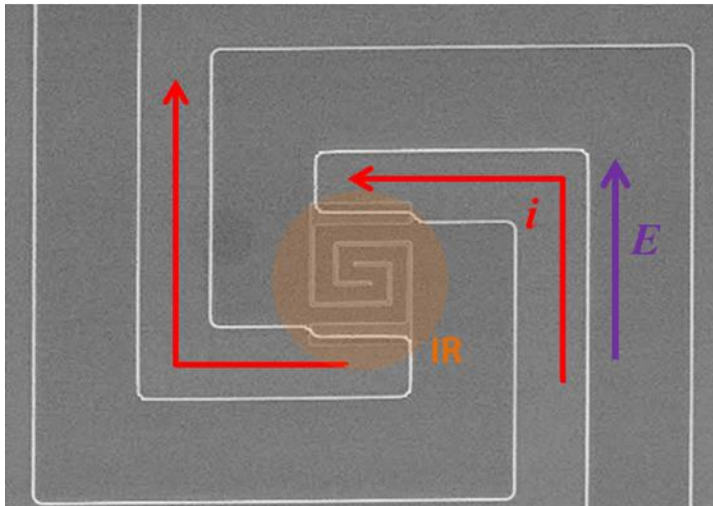
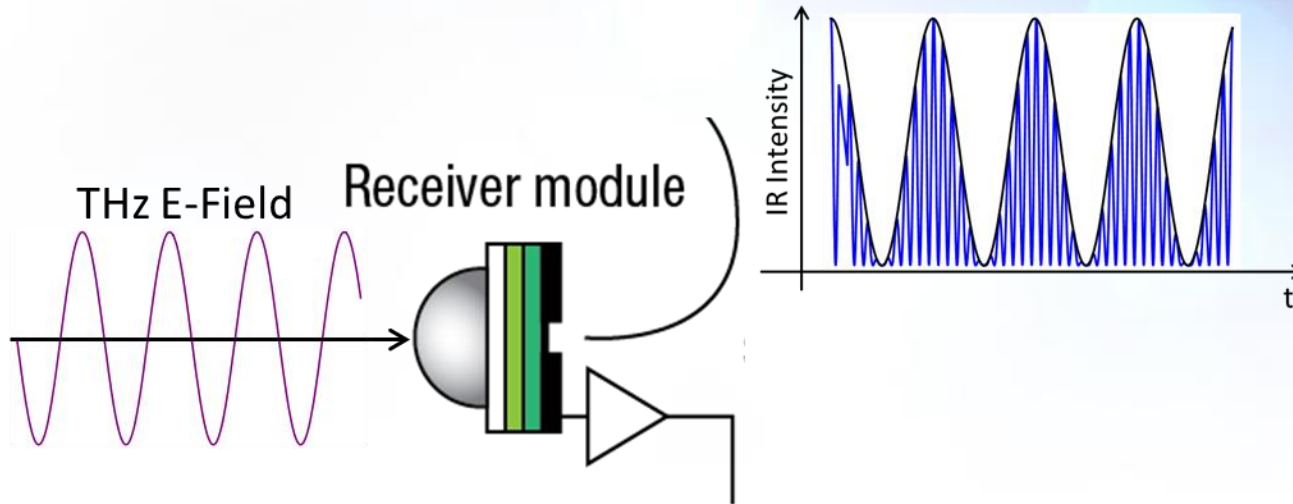


# Continuous-Wave THz Spectroscopy - Generation

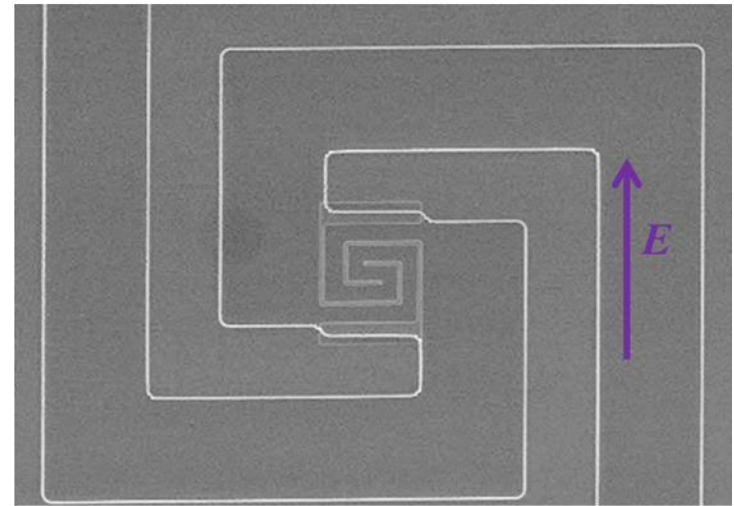




# Continuous-Wave THz Spectroscopy - Detection

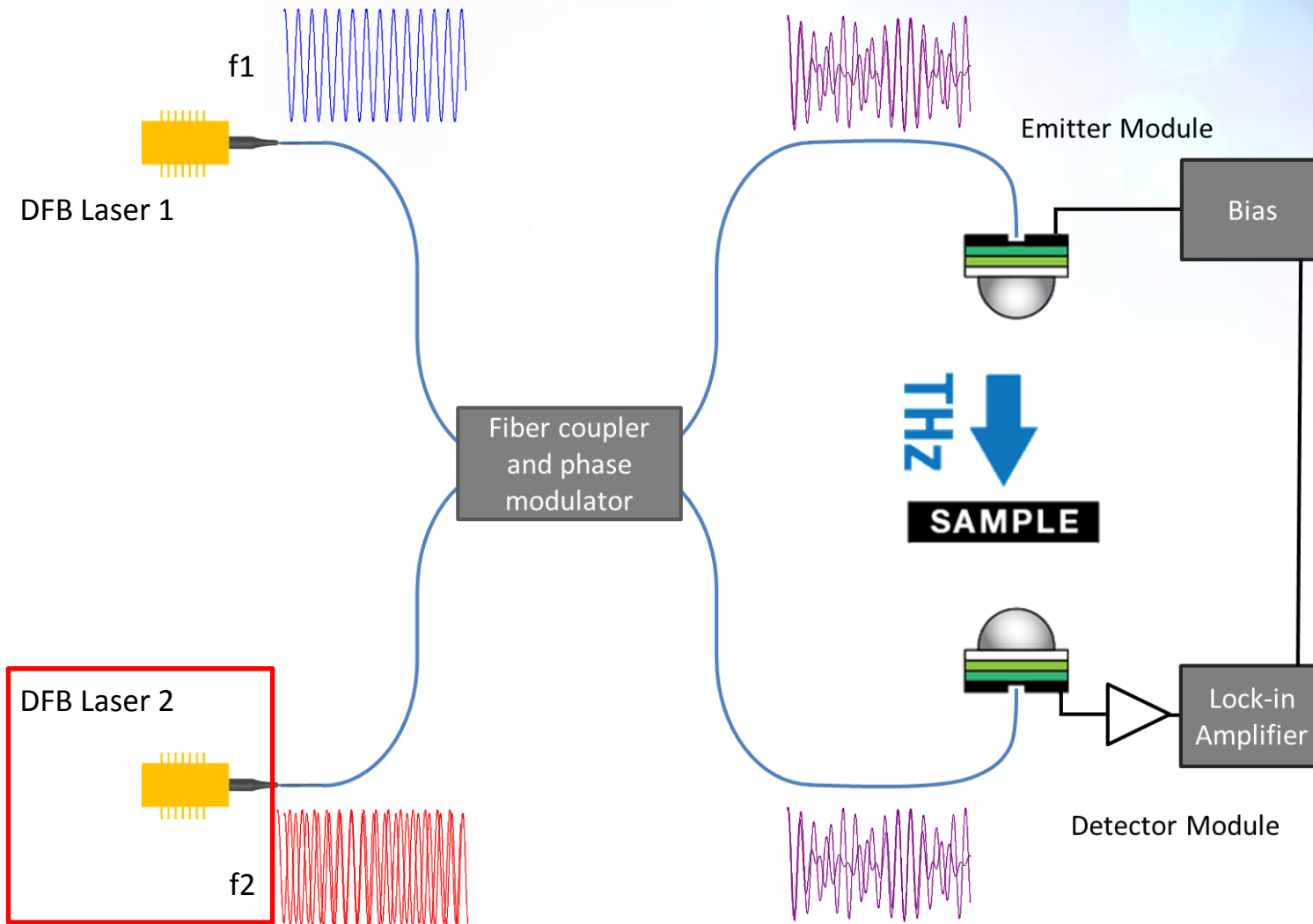


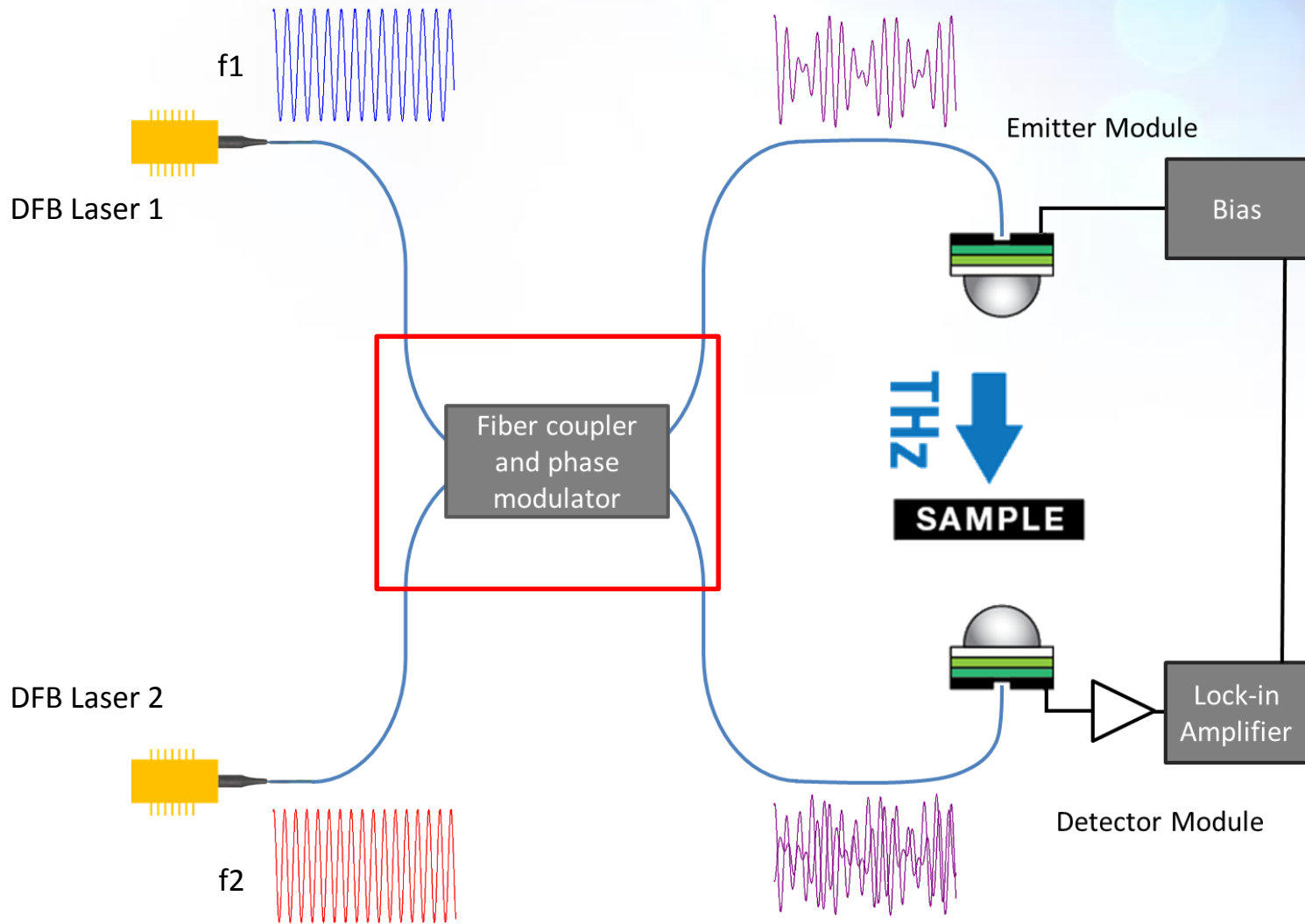
Detected current



No detected current

# Continuous-Wave THz Spectroscopy - Frequency Tuning





# Integrated Software for Turn-key Operation



The screenshot displays the software interface for the Lake Shore Model 8500 Series THz System. The main window is titled "Lake Shore Model 8500 Series THz System" and features a standard Windows-style menu bar with "File" and "Home" options. A toolbar on the left includes "Restore Defaults" and "Setup". The interface is organized into several functional areas:

- Measure Panel:** Shows the current operator as "Joe User" and the selected sample as "Lactose .5mm sample #5". It also lists available activities, with "Spectrum Measurement" currently selected.
- Sample Panel:** Contains the sample ID "Lactose .5mm sample #5" and a section for "Materials and dimensions". This section includes fields for "Sample material ID" (set to "Lactose"), "Sample film thickness" (set to ".5 mm"), "Sample substrate material ID", "Sample substrate thickness", "Reference substrate material ID", and "Reference substrate thickness". Below this is a "Limits" section with fields for "Min temperature" (0 [K]), "Max temperature" (400 [K]), "Max Field" (9 T), and "Max Bias Voltage" (20 [V]).
- Monitor Panel:** Displays real-time system status. It shows "Spectrometer" as "OK", "THz Off", and "Temperature" as "Ramping" with a current value of "131.248 K" and a "Setpoint" of "5.000 K". Other indicators include "Field" (0.000 T), "LHe Level" (65.0%), and "Sample Tray Position" (Unknown).

At the bottom of the window, there are "Setup" and "Comments" tabs.

# Integrated Software for Turn-key Operation



The screenshot displays the software interface for the Lake Shore Model 8500 Series THz System. The window title is "Lake Shore Model 8500 Series THz System". The interface is divided into several sections:

- File Menu:** Located at the top left, it includes options for "Start", "Stop", "Restore Defaults", and "Execute". The "File" menu itself is highlighted with a red box.
- Operator:** Shows the name "David Daughton".
- Sample:** Currently empty.
- Activities:** A list of activities with "Spectrum Measurement" selected and highlighted in orange.
- Measure Section:** Contains sub-sections for:
  - Spectrometer frequencies:** Start frequency (0.2 THz), End frequency (1.4 THz), and Frequency step (1000 MHz).
  - Spectrometer settings:** Number of scans (1), Time constant (30 ms), and Phase modulator value (Up: 14000, Down: 14000).
  - Processing settings:** Smoothing points (None).
  - Estimated running time:** 00:07:40.
- Monitor Section:** A vertical stack of status indicators:
  - Spectrometer: OK
  - THz Off
  - Temperature: OK, Setpoint: 290.002 K
  - Field: OK, Setpoint: 0.000 T
  - LHe Level: OK
  - 64.5% (with a timer of 0:46)
  - Sample Tray Position: OK, Setpoint: Unknown

# Integrated Software for Turn-key Operation



Sample

Reference

Background

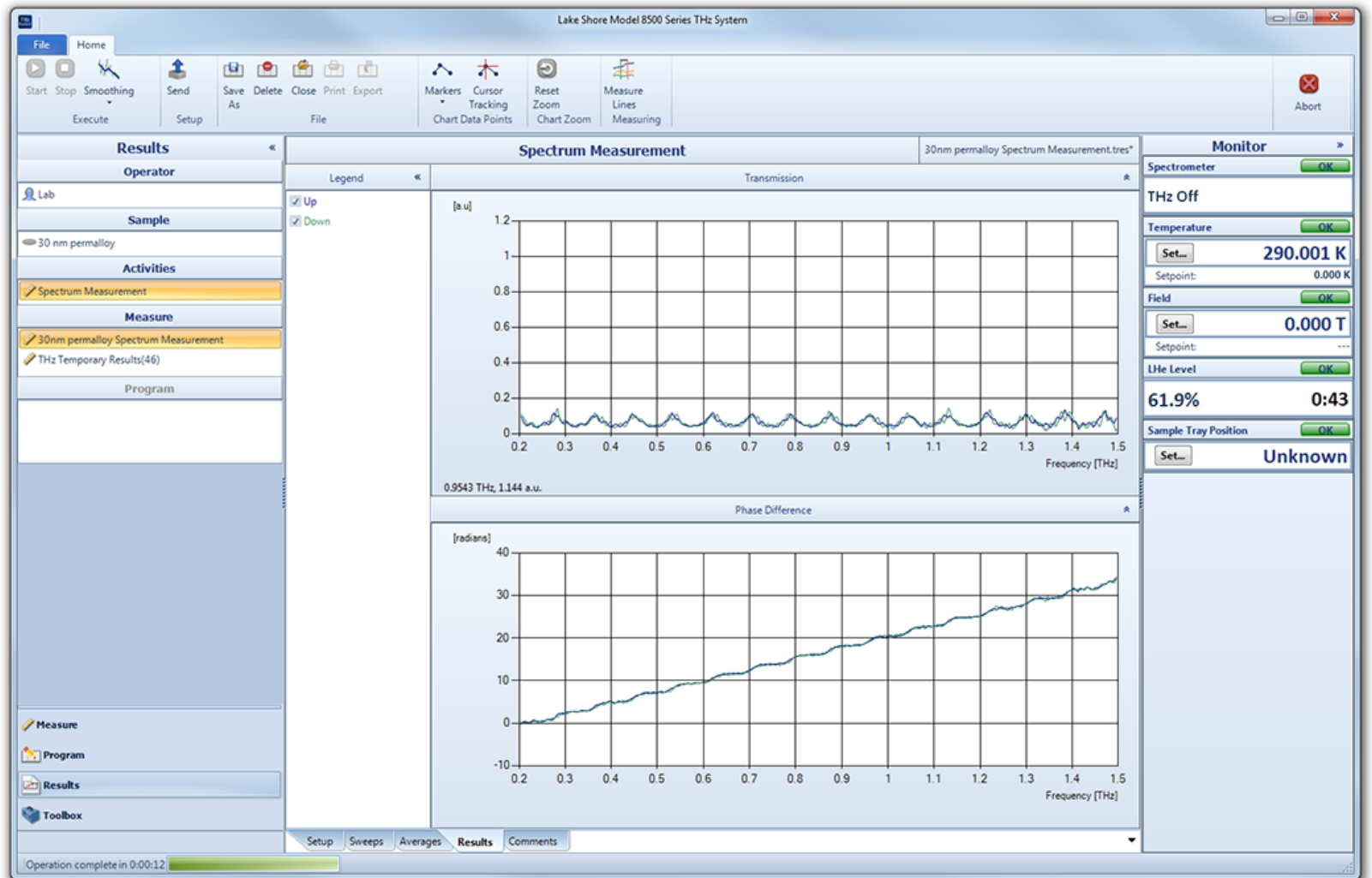


**SAMPLE**

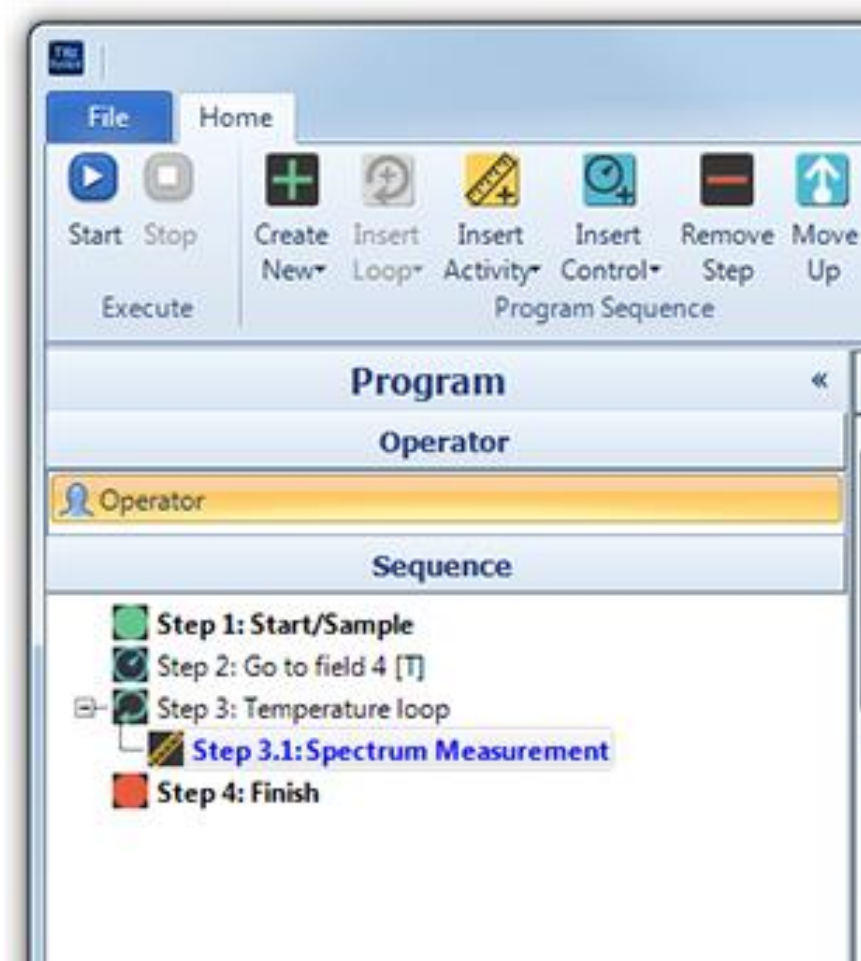
**BLOCK**



# Integrated Software for Turn-key Operation



# Integrated Software for Turn-key Operation

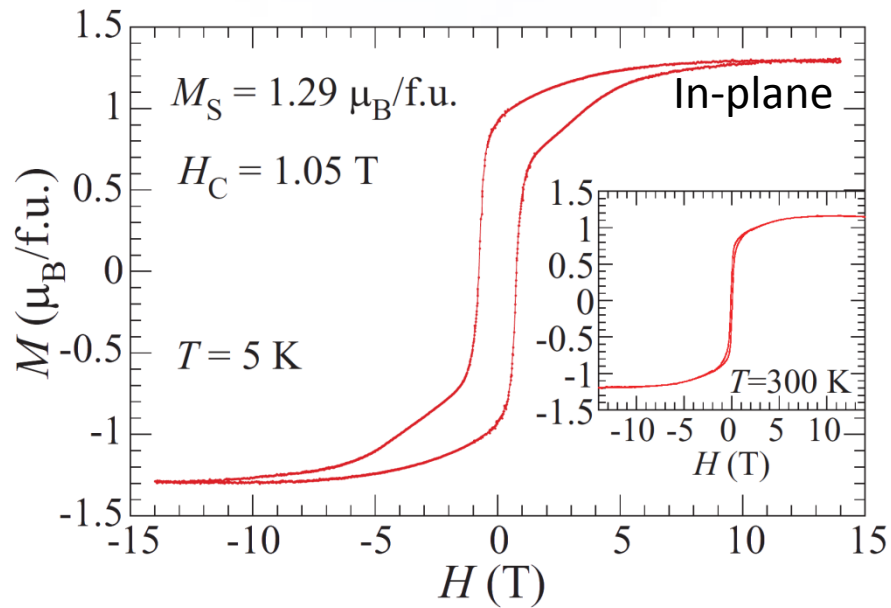




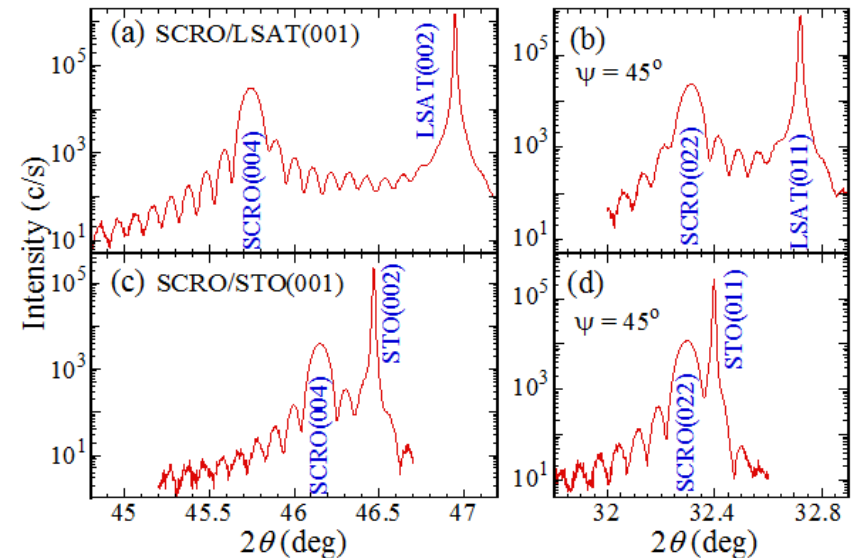
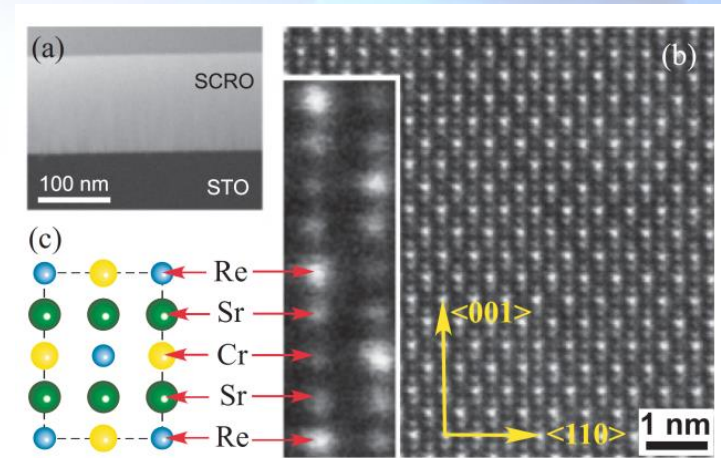
# Case Study: Sr<sub>2</sub>CrReO<sub>6</sub> Thin Films



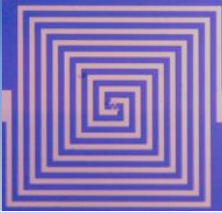
- double-perovskite ferrimagnet ( $T_c = 635$  K)
- $M_s = 1.29 \mu_B$  per f.u.
- Predicted 90% spin polarization
- Large anisotropy



- Stoichiometric, epitaxial, and well ordered
- Well ordered films on STO and LSAT substrates

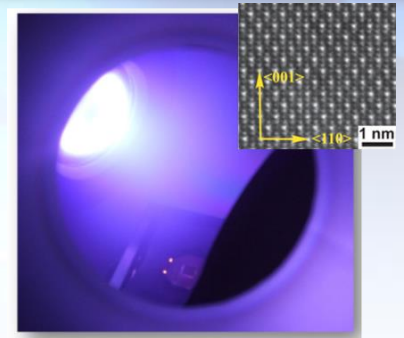


# Case Study: $\text{Sr}_2\text{CrReO}_6$ Thin Films



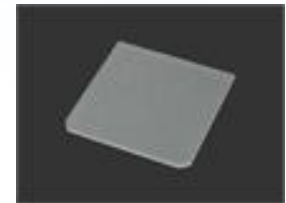
## Stoichiometric, epitaxial, and well ordered

- Grown with off-axis magnetron sputtering  
1  $\mu\text{m}$  thickness  $\sim$  20 hours



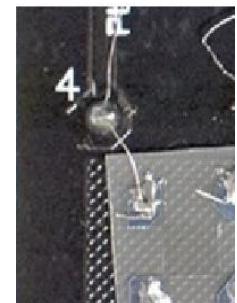
## Low Material yields

- 10 to 200 nm thick film on 10 x 10 mm substrate
- After characterization, wafer is diced & distributed for device manufacturing

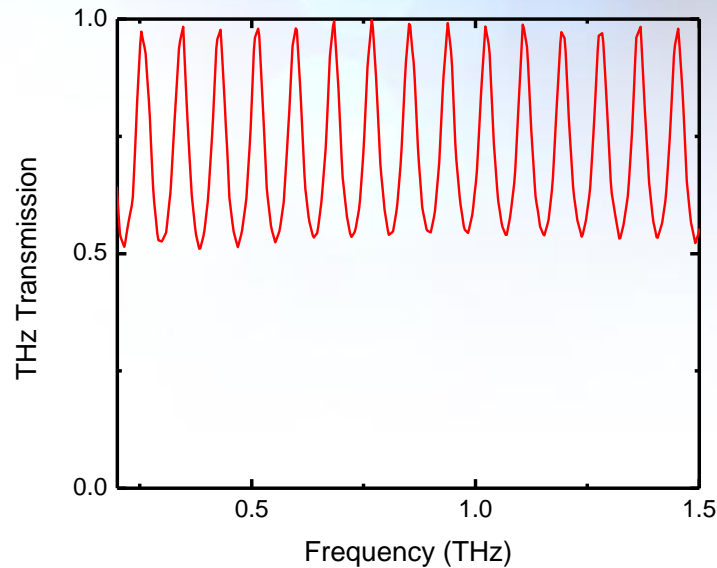
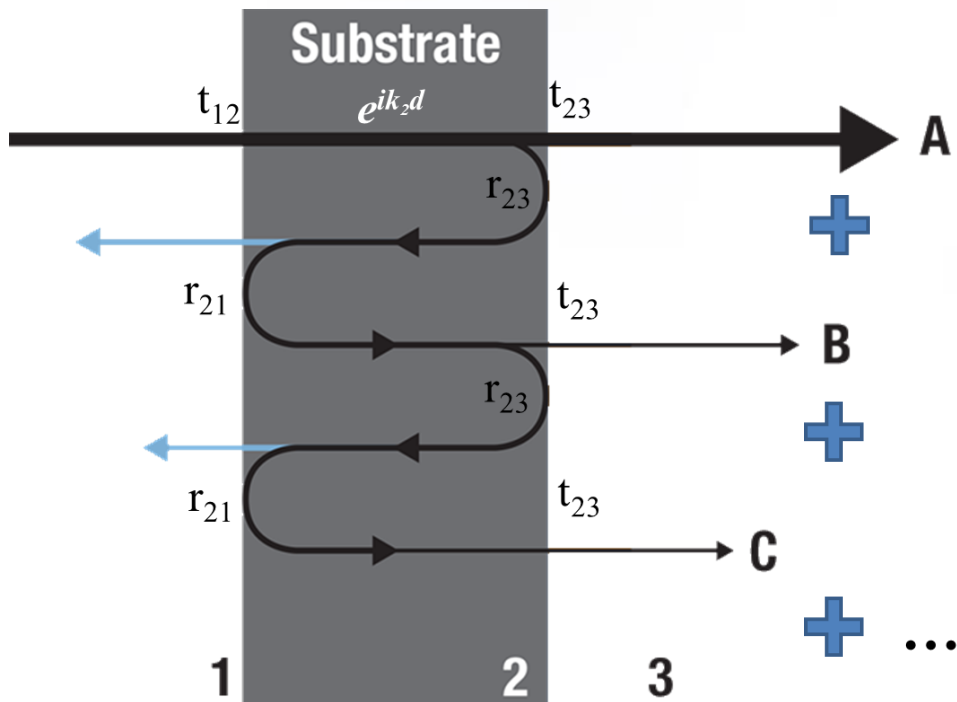


## Wafer real estate is at a premium

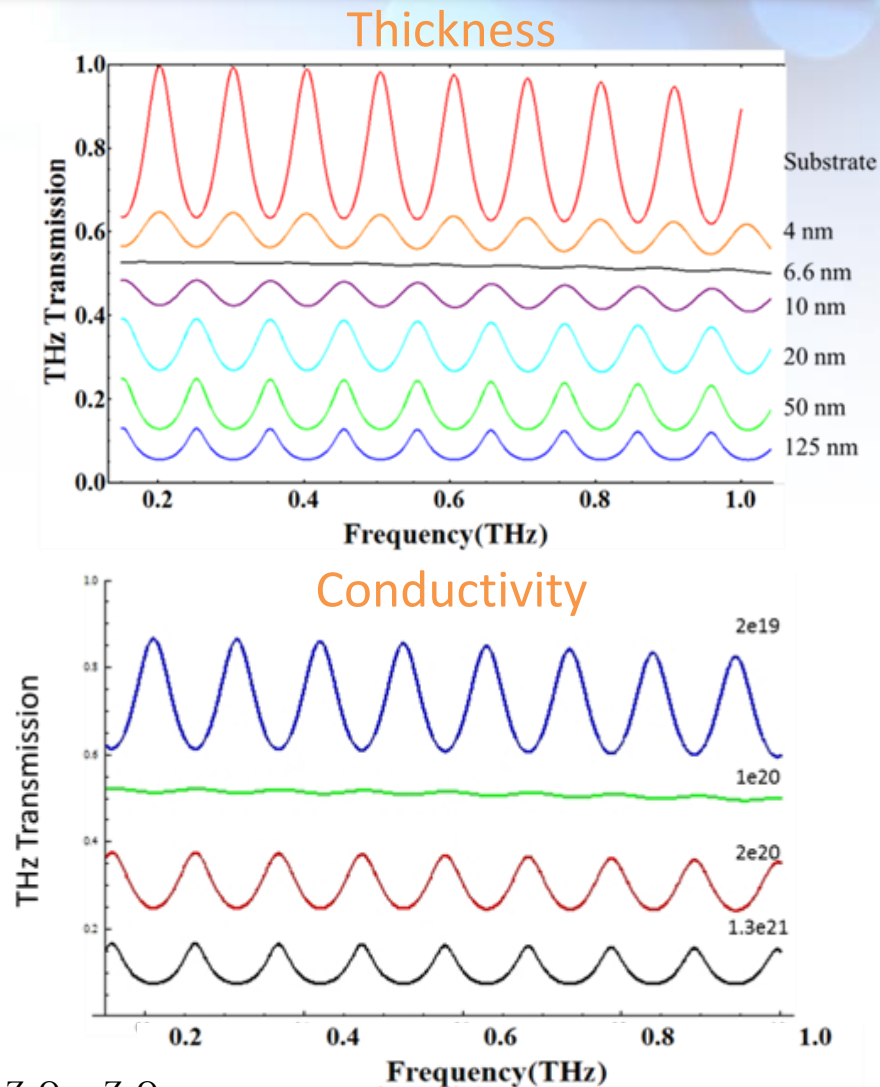
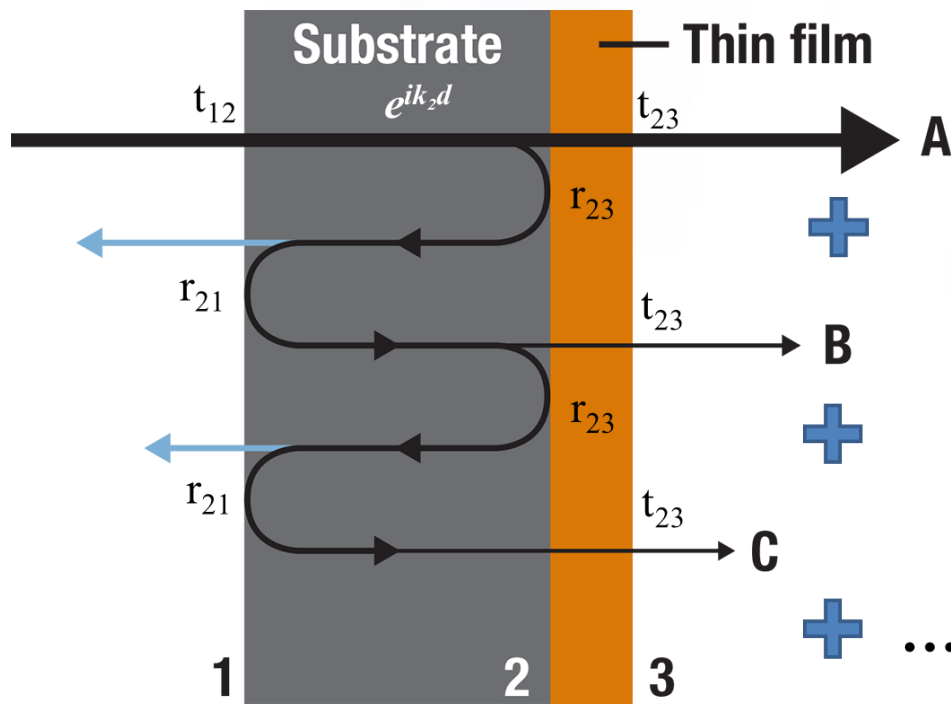
- Electrical contacts for material evaluation reduce number of devices – non contact evaluation
- Cryogenic characterization elucidates conduction mechanisms



# CW-THz transmission

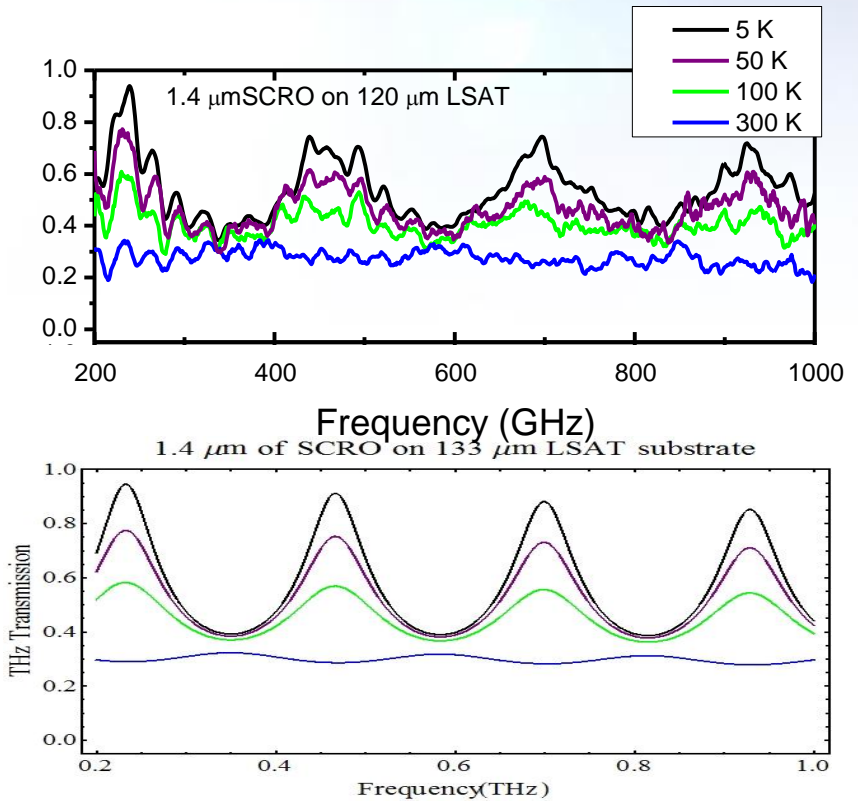
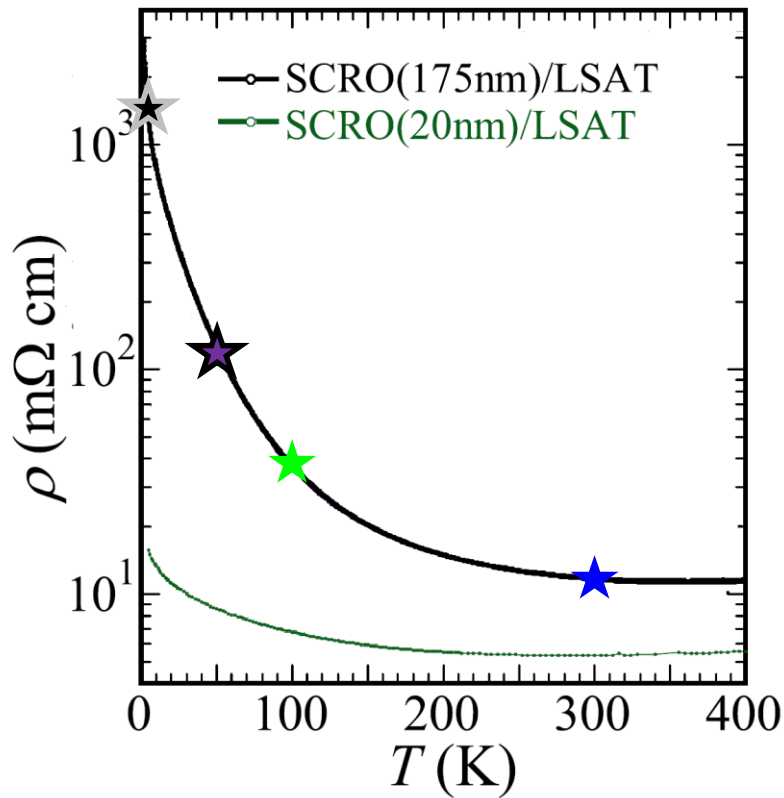


# CW-THz Spectroscopy of Conductive Thin Films



Ga-doped ZnO on ZnO

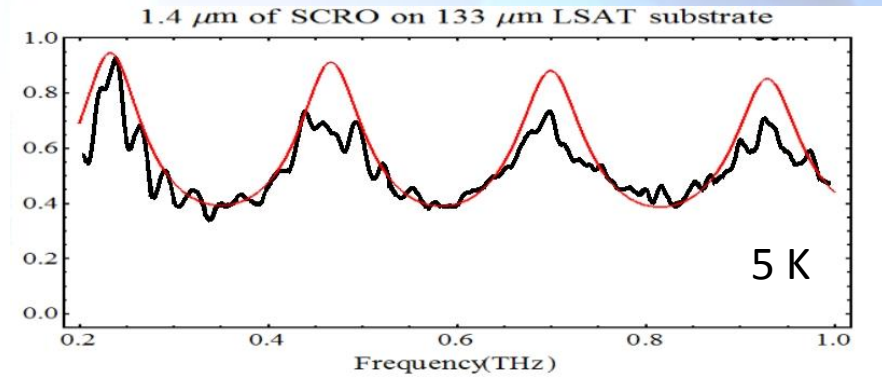
# SCRO Film DC conductivity



# Variable Range Hopping Conductivity



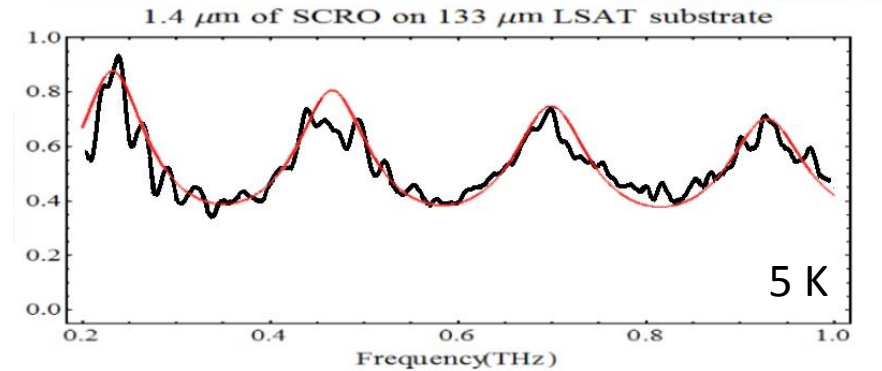
$$\sigma(\omega) = \sigma_{DC}$$



## Variable Range Hopping

$$\sigma(\omega) = A(T)\omega^{s(T)} + \sigma_{DC}$$

$$A(5K) \sim \frac{1000}{2\pi} \quad s(5K) \sim 0.8$$



N. F. Mott and E. A. Davis. *Electronic Processes in Non-Crystalline Materials*. Clarendon Press Oxford, 1971.

# What's Next

## Model 8500

### THz System for Material Characterization

- Turn-key acquisition of temperature and field dependent THz spectra

Taking orders March 2014!



- Customer applications – samples
- Sponsored research program to develop turn-key analysis software for CW-THz spectra

# Acknowledgements

## Collaborators:

- Lake Shore  
Scott Yano & Richard Higgins
- EMCORE  
Joseph Demers
- The Ohio State University  
Center for Emergent Materials  
Chunhui Du & P. Chris Hammel
- Wright State University  
David Look & Tim Cooper
- University of Arizona  
Hao Xin & Min Liang
- IDCAST/UDRI  
G. E. Pacey

## Funding:

- Ohio Third Frontier  
The logo for the Ohio Department of Development, featuring the word "Ohio" in a bold, red, sans-serif font, followed by a vertical line and the words "Department of Development" in a smaller, black, sans-serif font.
- USAF STTR Program