Optical Spectroscopy of Advanced Materials
Basic optics, nonlinear and ultrafast optics

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Outline (Feb 9th -20th)

1. Feb 9th, 11th and 13th: overview, basic optics and spectroscopy

2. Feb 16th, 18th and 20th: Advanced optics, ultrafast and nonlinear spectroscopy
   - femtosecond lasers: case study; spectroscopy techniques: incoherent & coherent transient, magneto-optical, infrared & time-domain THz

General References:

Demtroder, Laser Spectroscopy: Basic Concepts and Instrumentation
Diels and Rudolph (DR), Ultrashort Laser Pulse Phenomena
Shah, Ultrashort Spectroscopy of Semiconductors and Semiconductor Nanostructures
Chemla, D.S., Ultrafast Transient Nonlinear Optical Processes in Semiconductors
For example, see Copper, S.L, Optical Spectroscopy Studies of Metal-Insulator Transitions in Perovskite-related Oxides

Jigang Wang, Feb, 2009
Overview and Introduction
“Light is, in short, the most refined form of matter.”

Louis de Broglie

Berkeley, California
The first spectroscopy experiment

A brief history of optics

17th-century  18th-century  19th-century  20th-century

Kepler, Huygens

Newton...

Fresnel, Young...

Maxwell Michelson...

Einstein...

Total internal reflection, Telescope, geometrical optics, the wave theory, prism dispersion, the particle theory of light

Interference, diffraction, expressions for reflected and transmitted waves, unified electricity and magnetism

Light is
(1) “a phenomenon of empty space”
(2) both a wave and a particle

The equations of optics

Maxwell’s equations

\[ \vec{\nabla} \cdot \vec{E} = \rho / \epsilon \]
\[ \vec{\nabla} \times \vec{E} = -\frac{\partial \vec{B}}{\partial t} \]
\[ \vec{\nabla} \cdot \vec{B} = 0 \]
\[ \vec{\nabla} \times \vec{B} = \mu \epsilon \frac{\partial \vec{E}}{\partial t} \]

\( \epsilon \) is the permittivity,
\( \mu \) is the permeability of the medium
Solving Maxwell’s equations

\[ \nabla^2 \vec{E} - \mu \varepsilon \frac{\partial^2 \vec{E}}{\partial t^2} = 0 \]

\[ \vec{E}(\vec{r}, t) \propto \cos(\omega t \pm \vec{k} \cdot \vec{r}) \]

Light is an Electromagnetic Wave
Wave Properties – Velocity

\[ \frac{\partial^2 E}{\partial x^2} - \mu \varepsilon \frac{\partial^2 E}{\partial t^2} = 0 \]

**Phase velocity**

\[ \frac{\omega}{k} = v = \frac{1}{\sqrt{\mu \varepsilon}} \]

**Group velocity**

\[ v_g \equiv [dk / d\omega]^{-1} \]

\[ v_g = v_{phase} \left/ \left( 1 + \frac{\omega}{n} \frac{dn}{d\omega} \right) \right. \]
Wave Properties – Spectrum

1 THz = 300 µm = 33 cm⁻¹ = 4.1 meV
At an oblique angle, light can be completely transmitted or completely reflected.

"Total internal reflection" is the basis of optical fibers, a billion dollar industry.
Conventional Spectroscopy Methods

- Absorption, Reflection, Emission, Interference, Scattering
- Spectrally resolved before or after sample

Absorption Spectroscopy

Penetration depth into water vs. wavelength

Dispersion elements

Scattering Spectroscopy

\[ I_{\text{Raman}} \propto \lambda^{-4} \]

Fourier Transform Infrared (FTIR) Spectrometer

White light *Interference*

Figure 1
Typical SWLI Signal

Nature can do similar tricks by itself

Nature knows interference

The amazing light – Laser

A laser will lase if the beam increases in irradiance during a round trip: that is, if $I_3 > I_0$.

Continuous vs. ultrashort pulses of laser

Irradiance vs. time

Continuous beam:

Ultrashort pulse:

Spectrum

- time

- frequency

- time

- frequency
# How fast is Ultra-fast?

<table>
<thead>
<tr>
<th>Prefix</th>
<th>Value</th>
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<tbody>
<tr>
<td>milli</td>
<td>$10^{-3}$</td>
</tr>
<tr>
<td>micro</td>
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</tr>
<tr>
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<td>$10^{-9}$</td>
</tr>
<tr>
<td>pico</td>
<td>$10^{-12}$</td>
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<tr>
<td>femto</td>
<td>$10^{-15}$</td>
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<tr>
<td>atto</td>
<td>$10^{-18}$</td>
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The evolution of pulse Lasers

<table>
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<tr>
<th>Year</th>
<th>Time resolution (seconds)</th>
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<tr>
<td>1960</td>
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</tr>
<tr>
<td>1970</td>
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<td>1990</td>
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<tr>
<td>2000</td>
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</table>

Electronics
Optics

Courtesy of Trebino

Long vs. short pulses of laser

Irradiance vs. time

Spectrum

Long pulse

Short pulse
Generating short pulses = “mode-locking”

Locking the phases of the laser frequencies yields an ultrashort pulse.

Random phases of all laser modes:
- Out of phase
- Out of phase
- Out of phase

Locked phases of all laser modes:
- In phase!
- Out of phase

Irradiance vs. time:
- Random phases
- Ultrashort pulse!!
- Light bulb

A generic ultrashort-pulse laser

A generic ultrafast laser has a broadband gain medium, a pulse-shortening device, and two or more mirrors:

- Gain medium
- Pulse-shortening device
- Continuous laser pump source
- Partially reflecting output mirror

Pulse-shortening devices include:
- Saturable absorbers
- Phase modulators
- Dispersion compensators
- Optical-Kerr media
Different frequencies travel at different group velocities in materials, causing pulses to expand to highly "chirped" (frequency-swept) pulses.

Ultrashort laser pulse broadening

Input ultrashort pulse

Output pulse, fs

Input Pulse, fs
Ultrafast Optics is Nonlinear Optics
Strategic advantages

Ultrafast
Ultrabroadband
Manipulation
Ultrafast Magneto-optical Spectroscopy

Excitation © J Wang, LBNL

Carriers

Detection

Magnetic Ions

Tunable pump-probe from MIR, NIR to visible

Highly sensitive to time reversal symmetry breaking

Ultrafast THz Spectroscopy

Field-resolved Detection

Complex transmission coefficient

$$t(\omega) \equiv \frac{E_{\text{OUT}}(\omega)}{E_{\text{IN}}(\omega)} \approx \frac{2}{1 + n_s + d Z_0 \sigma(\omega)}$$
Coherent Transient Spectroscopy

Ultrafast FWM

\[ k_1 \quad k_3 \quad k_2 \quad k_1 + k_2 - k_3 \]

'Residual' coulomb interactions → the dynamics between quasiparticles

Ultrafast demagnetization

- $\Delta \theta_K / \theta_K$

Time Delay (ps)

(1) (2) (3) (4)
Ultrafast spectroscopy of HTc superconductor

Cuprate SC: Generic Phase Diagram

Temperature (K)

Antiferromagnetic  No Gap  Superconducting

underdoped  optimally doped  overdoped

CuO₂-plane carrier density


Many-body Effects in SWCN

1D Band picture

van Hove singularity

1D Exciton picture

Photon Energy (meV)

Transmittance

$\Delta t = -500 \text{ fs}$

$\Delta T/T (%)$

$\Delta t = 200 \text{ fs}$

$\Delta t = 1000 \text{ fs}$

$\Delta t = 2000 \text{ fs}$

Ultrafast Laser Lab Tour

www.cmpgroup.ameslab.gov/ultrafast/

Welcome to the Wang Research Group
Our group is with the Condensed Matter Physics program of the Physics Department at Iowa State University and Ames Laboratory-US DOE.

The major challenges that nature poses for current condensed matter and materials physics come from a world of small things and of complex things. Our research currently focuses on the development and application of ultrafast laser spectroscopy and/or microscopy to study the world of nanoscience and complexity (or simply, advanced materials). The combination of femtosecond $[10^{-15}$ s] time resolution, broadband probe capabilities, and specifically designed nanostructures and complex materials opens up many exciting opportunities to understand and manipulate the fundamental properties of advanced materials [Figure 1].

Energy (THz)

1000 100 10 1

SH/SE OPA DFG OR

Ultrafast, Ultrasmall and Ultrabroadband