fs-time-resolved spectroscopic ellipsometry

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• Why time-resolved SE?

• Setup: Experimental requirements and solutions

• Experimental demonstration: First results on...
  • ZnO
  • Ge

• Perspective
Time-resolved ellipsometry

• Time-resolved: Charge carrier dynamic

• Probing JDOS, not occupation/luminescence => already before recombination

• Single-wvl? Bare reflectance? => SE!
**Time-resolved ellipsometry**

- Time-resolved:
  - Charge carrier dynamic
  - Probing JDOS, not occupation/luminescence => already before recombination
  - Single-wvl? Bare reflectance? => SE!

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**Pump-probe with white light pulses!**

![Graph showing optical transition](image-url)
Pump-probe ellipsometry

- Laser: 35fs, 6mJ, 800nm, 1kHz (amplified Ti:Sapph)
  - Probe: continuum white light generation in CaF\textsubscript{2}
  - Pump: e.g. 266nm (THG)
Pump-probe ellipsometry

- Laser: 35fs, 6mJ, 800nm, 1kHz (amplified Ti:Sapph)
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  • Probe: continuum *white light* generation in CaF$_2$
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• 1m delay line (6ns); resolution 500 nm (3fs)
Pump-probe ellipsometry

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- Probe: continuum white light generation in CaF₂
- Pump: e.g. 266nm (THG)
• Polarization optics (PSCA, rotating polarizer)
Pump-probe ellipsometry

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Ahead the sample:
- reflective optics
- thin polarizer

GVD (chirp)
Pump-probe ellipsometry

- Laser: 35fs, 6mJ, **800nm**, 1kHz (amplified Ti:Sapphire)
- Probe: continuum **white light** generation in CaF$_2$
- Pump: e.g. **266nm** (THG)
• Two-chopper scheme

⇒ Reflectance difference spectra at different delays for different compensator azimuth angles (polarizers fix)

⇒ Compute transient ellipsometric parameters using reference
Experimental results
ZnO
Pump-probe ellipsometry

- Example:
  ZnO thin film (30nm on SiO2), pumped by 266nm “entire film homogeneously pumped” ($10^{20}$ cm$^{-3}$)
Pump-probe ellipsometry

• Experimental aspects:

  • Time resolution: 70fs? (< 200fs!) [clean-up by spectrometer entrance slit]

  • Spot sizes: pump 400um, probe 200um, 15-20° off-angle

  • Group velocity delay (chirp) corrected

  • Depolarization
Pump-probe ellipsometry

- Modeling:
  - Dielectric function of ZnO by Kramers-Kronig-consistent B-spline
  - For each delay time

Pump-probe ellipsometry

- Sub-picosecond:
  - Immediate bleaching/blocking of above-gap absorption (excitions)
  - “fine structure”
  - Mid-gap absorption
ZnO dynamics

- Intraband absorption

Maciej Neumann, dissertation 2015
Pump-probe ellipsometry

- Sub-picosecond:
  - Immediate bleaching/blocking of above-gap absorption (excitons)
  - “fine structure”
  - Mid-gap absorption

![Graph showing energy vs. time with different curves for different energies and time points.](image)
ZnO dynamics

- Subsequent picoseconds:
  - Relaxation (Thermalization) of hot charge carriers
  - Return of above-gap absorption, vanishing mid-gap absorption
  - vis refractive index increasing again
ZnO dynamics

Maciej Neumann, dissertation 2015
ZnO dynamics

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ZnO dynamics

- Long time-scale (ns):
  - red shift (heating)
  - still “fine structure”

- Note:
  excitons persisted at any time although exceeding Mott density
Experimental results
Ge
• Fermi singularity (band filling singularity)
  • Indirect
  • Parallel bands (2D-like)
  • Analogon Burstein-Moss shift (blocking)

• Here: no doping, but pumping with 800nm (negligible gradient due to low enough)
• \( \langle \epsilon \rangle \)
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• $<e>$

Issues:
- Surface sensitivity
- Interpolation/smoothing for 2$^\text{nd}$ deriv.
General Challenges

Work to be done

• Gradient in depth
• Error estimation
• Imaging?
• ... lots of small experimental improvements...
• User facility near Prague (2019)
• 4 high-power lasers
  (up to 10PW/2kJ or 1kHz/100mJ)

• “ELIps”:
  VUV magneto-ellipsometer

Pump-Probe spectroscopic ellipsometry: *beyond transient reflectance*

- Experimental implementation VIS-NUV
- Demonstration VIS-UV
  - ZnO: hot charge carrier effects on fs to ns time scale
  - Ge: pump-induced band filling singularity
- Perspective: VUV ellipsometer

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Depolarization
As-is measurement

Probe-only spectra at every time delay
MM elements: smaller than +/-0.005
Psi: smaller than +/-0.1°
Delta: smaller than +/-0.1°
same with 6mm quartz in beam
non-collinear pump and probe beam

$\phi = 20^\circ, d = 200 \mu m \Rightarrow \Delta t = 220 \text{ fs}$
Spectral and temporal resolution

\[ \Delta E \Delta t \geq \frac{\hbar}{4\pi} \]

50fs: 7meV

(factor ½ depending on understanding)