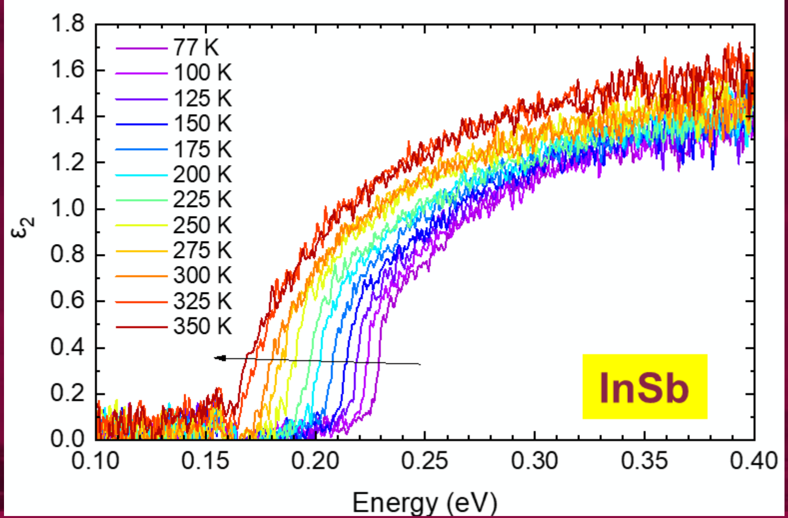
2023 Fall MRS

FA9550-20-1-0135
AFOSR
Gernot Pomrenke

Excitonic absorption in semiconductors with low and high carrier densities



Stefan Zollner

With:

Carola Emminger (Uni Leipzig and HU Berlin, Germany)

Jose Menendez (Arizona State University, Tempe, AZ)

Email: zollner@nmsu.edu. WWW: http://femto.nmsu.edu.

NM STATE

BE BOLD. Shape the Future.

College of Arts and Sciences, Department of Physics
New Mexico State University, Las Cruces, NM, USA

Biography


Regensburg/Stuttgart
Germany

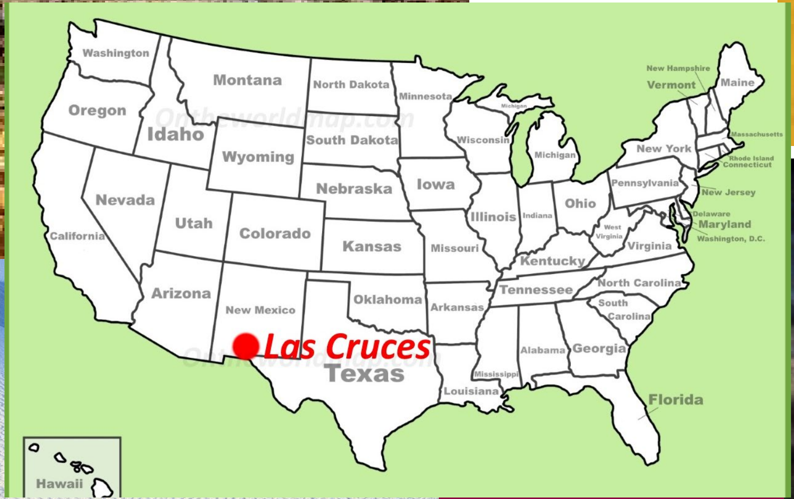

Motorola (Mesa, Tempe)
Arizona, 1997-2005


Freescale, IBM
New York, 91-92; 07-10


NMSU
Las Cruces, NM
Since 2010


Motorola, Freescale
Texas, 2005-2007

Where is Las Cruces, NM ???



White Sands NP

New Mexico State University, Las Cruces

Land grant institution, Carnegie R2 (soon to be R1)
Comprehensive: Arts and Sciences, Education, Business, Agriculture
Ph.D. programs in sciences, engineering, agriculture; Ag extension

14,000 students (11,500 UG, 2,500 GR), 1000 faculty

Minority-serving, Hispanic-serving (60% Hispanic/NA, 26% White)
Small-town setting

Military-friendly institution (Army and Air Force ROTC programs)

Community engagement classification
(first-generation students, Pell grant recipients)

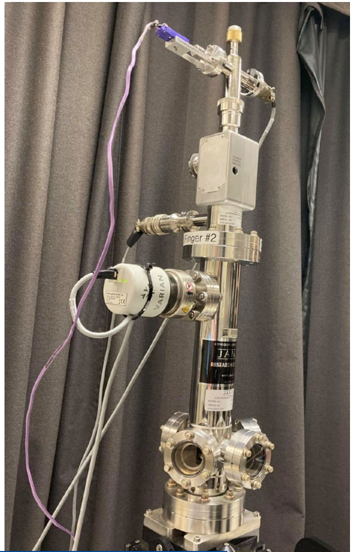
Physics: BS/BA, MS, PhD degrees. 71 UG and 37 GR students.
12 faculty (HE Nuclear and Materials Physics), **1.7 M$ expenditures.**
ABET-accredited BS in Physics and BS in Engineering Physics

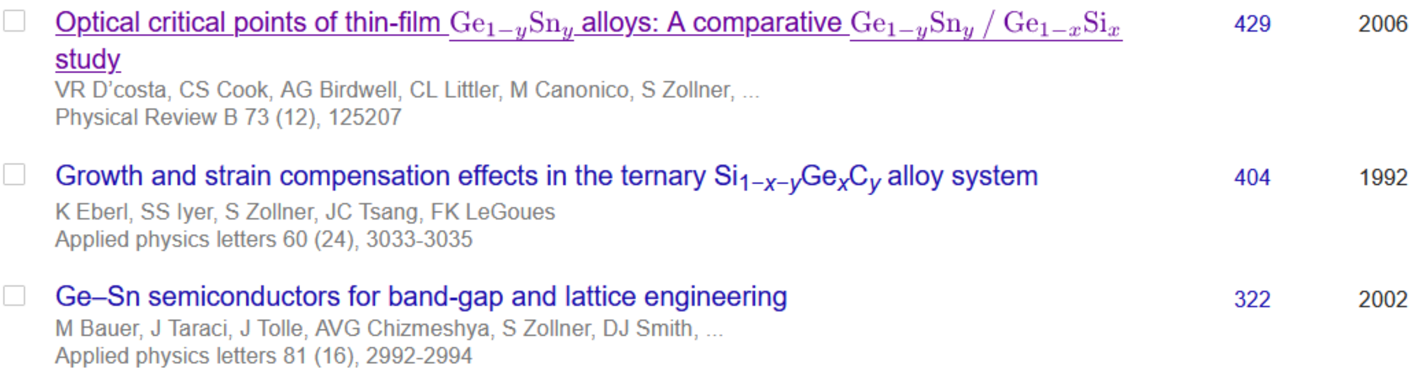
Ellipsometry at NMSU

Ellipsometry on anything (inorganic, 3D)

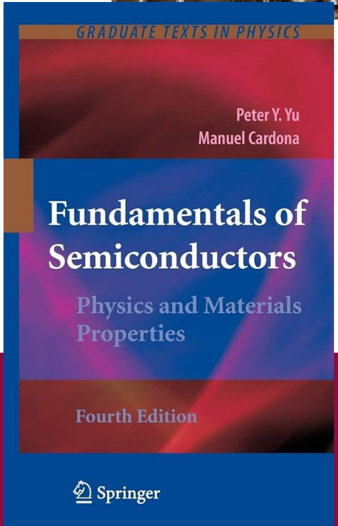
- Metals, insulators, semiconductors
- Mid-IR to vacuum UV
- 10 to 800 K

Ellipsometry tells us a lot about materials quality (not necessarily what we want to know).





☐	Optical critical points of thin-film $Ge_{1-y}Sn_y$ alloys: A comparative $Ge_{1-y}Sn_y$ / $Ge_{1-x}Si_x$ study VR D'costa, CS Cook, AG Birdwell, CL Littler, M Canonico, S Zollner, ... Physical Review B 73 (12), 125207	429	2006
☐	Growth and strain compensation effects in the ternary $Si_{1-x-y}Ge_xC_y$ alloy system K Eberl, SS Iyer, S Zollner, JC Tsang, FK LeGoues Applied physics letters 60 (24), 3033-3035	404	1992
☐	Ge–Sn semiconductors for band-gap and lattice engineering M Bauer, J Taraci, J Tolle, AVG Chizmeshya, S Zollner, DJ Smith, ... Applied physics letters 81 (16), 2992-2994	322	2002



Problem statement

(1) Achieve a **quantitative** understanding of **absorption** and **emission** processes.

- Our **qualitative** understanding of excitonic absorption is 50-100 years old,
- But **insufficient** for modeling of detectors and emitters.

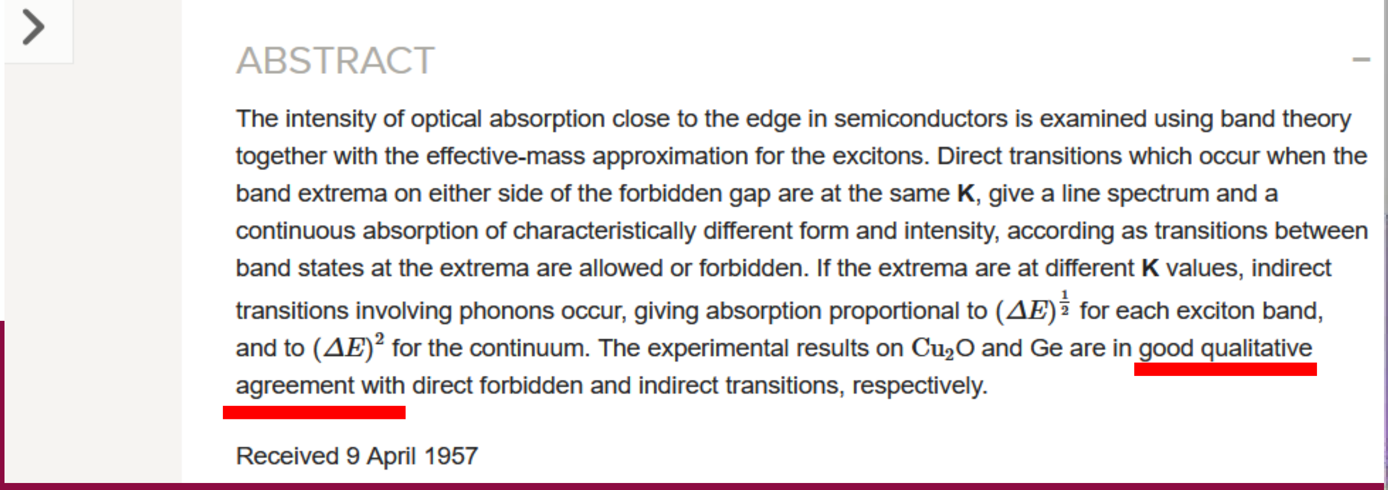
(2) How are optical processes affected by **high carrier concentrations** (screening)?

- High carrier densities can be achieved with
  - **In situ doping** or
  - **ultrafast (femtosecond) lasers** or
  - **high temperatures (narrow-gap or gapless semiconductors)**
- **Goal:** CMOS-integrated mid-infrared camera (thermal imaging with a phone).
- Future: How are optical processes affected by an electric field (pin diode or thin layer)?

Intensity of Optical Absorption by Excitons

R. J. Elliott

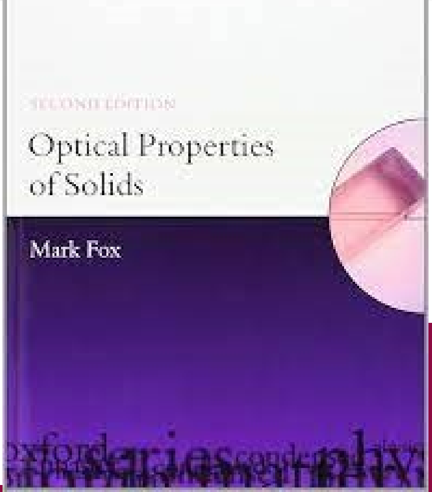
Phys. Rev. **108**, 1384 – Published 15 December 1957



ABSTRACT

The intensity of optical absorption close to the edge in semiconductors is examined using band theory together with the effective-mass approximation for the excitons. Direct transitions which occur when the band extrema on either side of the forbidden gap are at the same **K**, give a line spectrum and a continuous absorption of characteristically different form and intensity, according as transitions between band states at the extrema are allowed or forbidden. If the extrema are at different **K** values, indirect transitions involving phonons occur, giving absorption proportional to $(\Delta E)^{\frac{1}{2}}$ for each exciton band, and to $(\Delta E)^2$ for the continuum. The experimental results on $\mathrm{Cu_2O}$ and Ge are in good qualitative agreement with direct forbidden and indirect transitions, respectively.

Received 9 April 1957


OXFORD MASTER SERIES IN CONDENSED MATTER PHYSICS

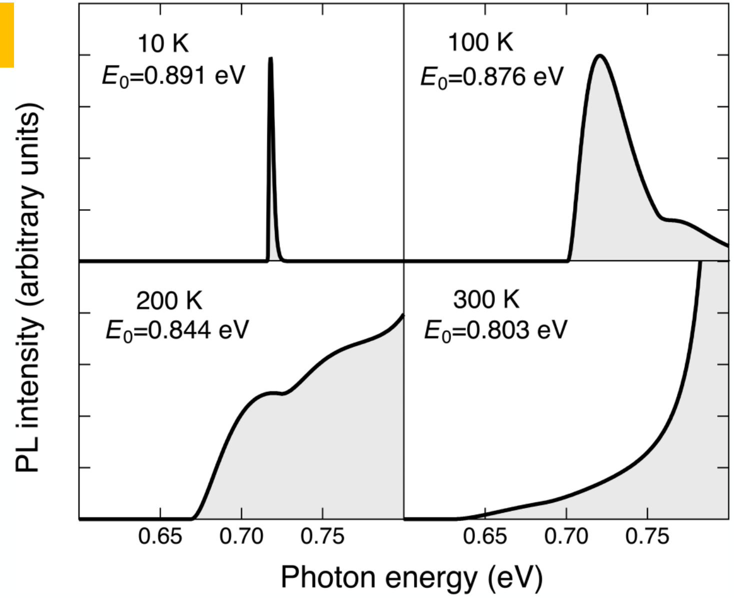
SECOND EDITION

Optical Properties of Solids

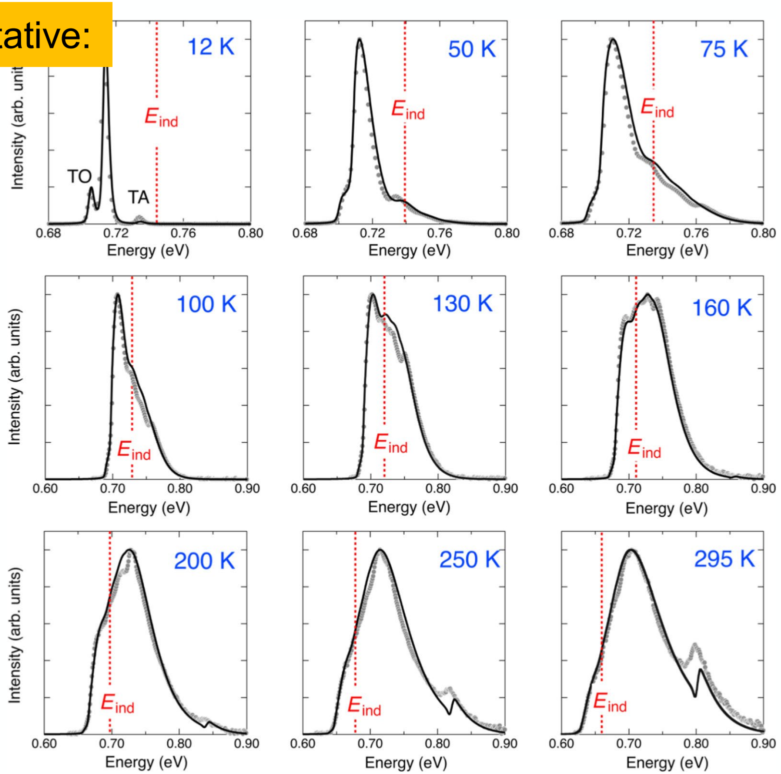
Mark Fox

Example 1: photoluminescence in germanium

Qualitative:



Quantitative:

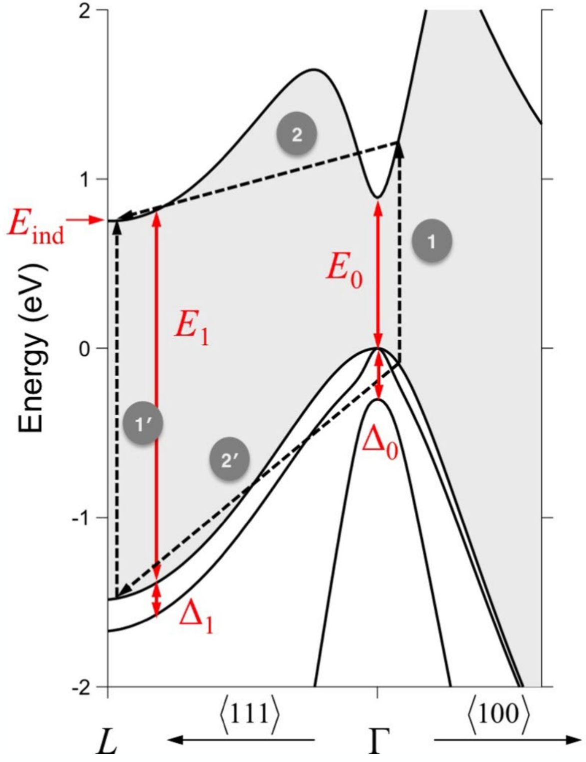


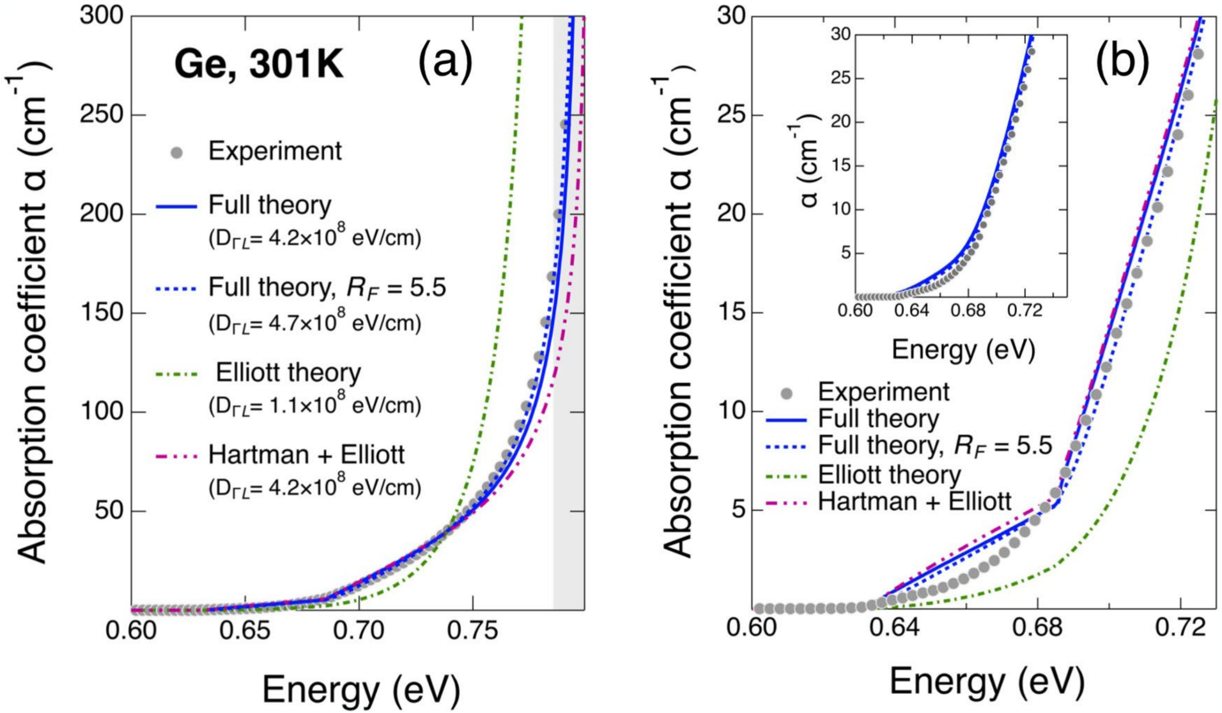
Roosbroeck-Shockley equation (PR **94**, 1558, 1954).

Complete failure above 200 K and at RT.

Menendez, Poweleit, Tilton, PRB **101**, 195204 (2020).

Example 2: indirect absorption in germanium



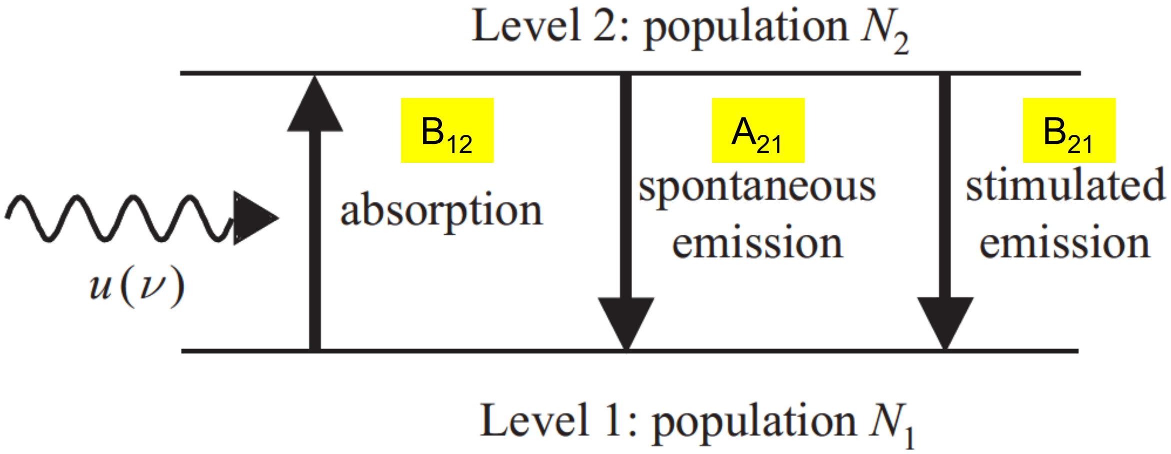


R. J. Elliott, Phys. Rev. **108**, 1384 (1957).
Menendez, Lockwood, Zwinkels, Noel, PRB **98**, 165207 (2018).

Optical constants of highly excited semiconductors

- Einstein coefficients, Fermi’s Golden Rule, Elliott-Tanguy excitons
- Direct gap absorption in **germanium** from 10 to 800 K
- Optical constants of **highly excited semiconductors**
  - Direct gap absorption in **InSb** from 10 to 800 K
  - Intravalence band absorption in topological insulators (**α-tin**)
  - Optical constants of **highly excited germanium** (femtosecond ellipsometry at ELI Beamlines in Prague)
- Conclusion and Outlook

Einstein coefficients



In equilibrium: N_1, N_2 constant.
Absorption and emission balance.
Black-body radiation $u(\hbar\omega)$

$$B_{12}N_1u(\hbar\omega) = A_{21}N_2 + B_{21}N_2u(\hbar\omega)$$

One coefficient is sufficient:

$$g_1B_{12} = g_2B_{21}$$

$$A_{21} = \frac{2\hbar\omega^3}{\pi c^3}B_{21}$$

Use Fermi’s Golden Rule to calculate B_{12}

Albert Einstein, *Strahlungs-Emission und Absorption nach der Quantentheorie*, DPG Verh. **18**, 318 (1916); Phys. Z. **18**, 121 (1917).

Fermi's Golden Rule: Tauc plot

Direct band gap absorption

constant **k·p** matrix element

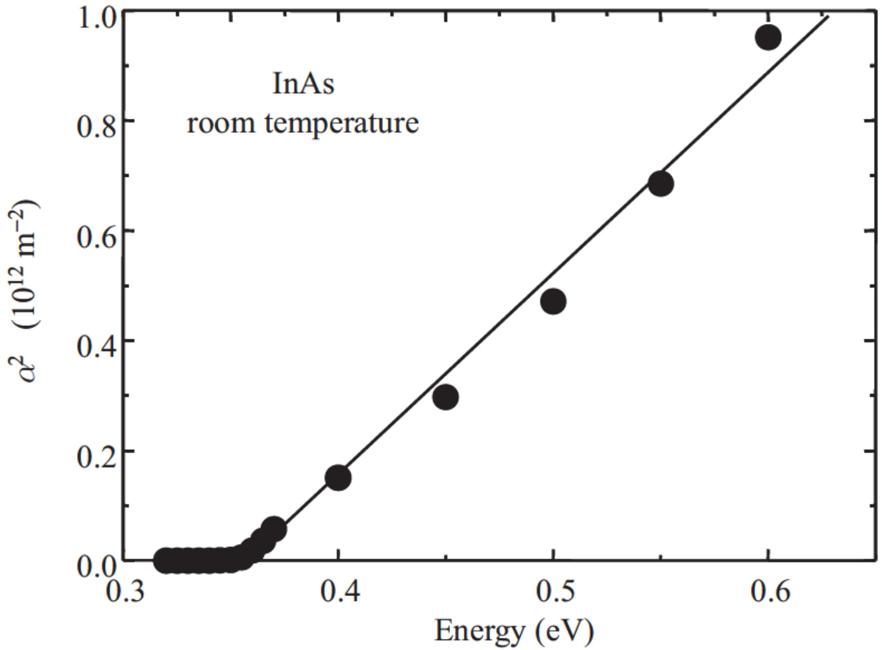
$$\frac{1}{\tau}=\frac{2\pi}{\hbar}\int_{i,f}|\langle f|H_{eR}|i\rangle|^2\delta\left(E_f-E_i-\hbar\omega\right)=\frac{2\pi}{\hbar}|\langle f|H_{eR}|i\rangle|^2 g_{fi}(\hbar\omega)$$

Joint DOS
parabolic bands

$$\langle f|H_{eR}|i\rangle=\frac{e}{m_0}\langle \boldsymbol{f}|\vec{\boldsymbol{p}}|\boldsymbol{i}\rangle\cdot\vec{A}_0$$

Use **k·p** matrix element *P*: $E_P=2P^2/m_0$

$$\varepsilon_2(\hbar\omega)=\frac{e^2\sqrt{m_0}\mu^{\frac{3}{2}}}{3\pi\sqrt{2}\varepsilon_0\hbar}\frac{E_P\sqrt{E_0}}{(\hbar\omega)^2}\sqrt{\frac{\hbar\omega}{E_0}-1}$$



Fox, Chapter 3

Fermi's Golden Rule: Tauc plot

Direct band gap absorption

$$\frac{1}{\tau}=\frac{2\pi}{\hbar}\int_{i,f}|\langle f|H_{eR}|i\rangle|^2\delta\left(E_f-E_i-\hbar\omega\right)=\frac{2\pi}{\hbar}|\langle f|H_{eR}|i\rangle|^2 g_{fi}(\hbar\omega)$$

$$\langle f|H_{eR}|i\rangle=\frac{e}{m_0}\langle \boldsymbol{f}|\vec{\boldsymbol{p}}|\boldsymbol{i}\rangle\cdot\vec{A}_0$$

Use **k·p** matrix element *P*: $E_P=2P^2/m_0$

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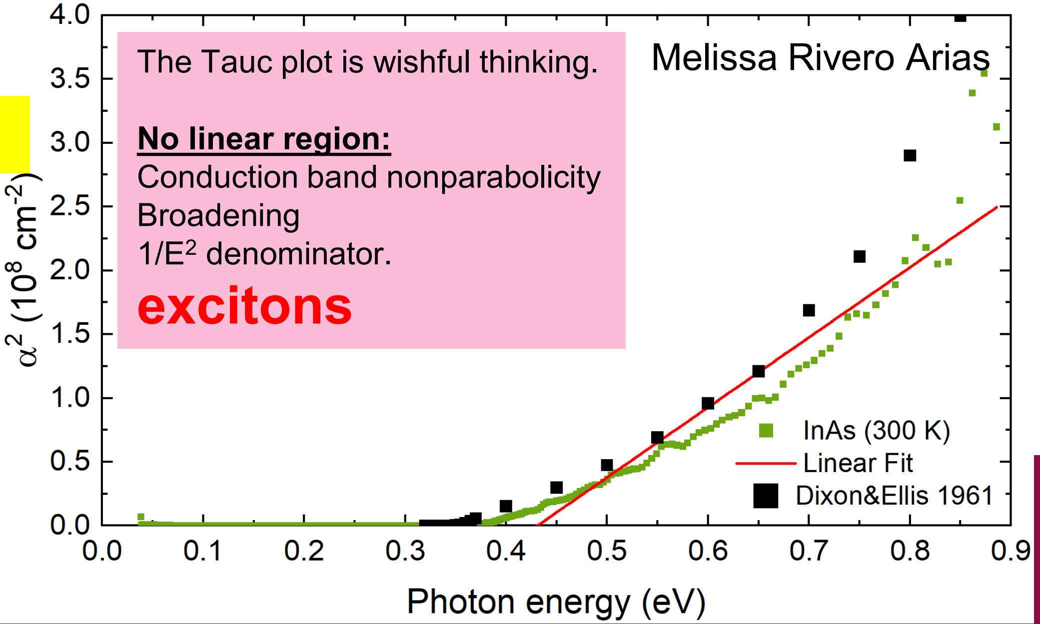


The Tauc plot is wishful thinking.

No linear region:
Conduction band nonparabolicity
Broadening
$1/E^2$ denominator.

excitons

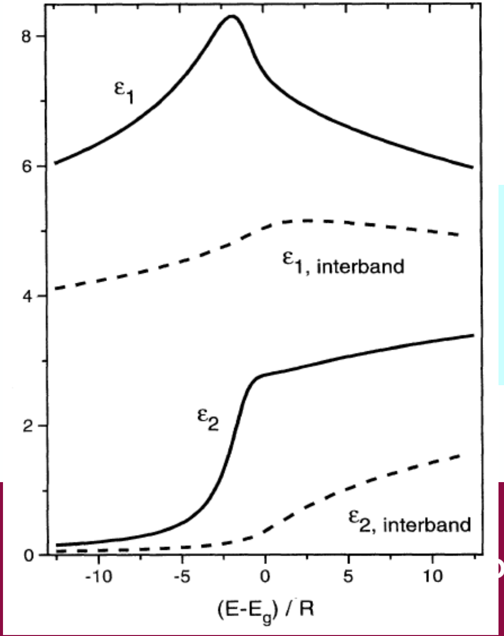
Melissa Rivero Arias

Elliott-Tanguy exciton absorption

Direct band gap absorption

Excitonic binding energy: $R=R_H\times\mu_h/\varepsilon_s^2$

$$\varepsilon_2(\hbar\omega) = \frac{e^2\sqrt{m_0}\,\mu^{\frac{3}{2}}}{3\pi\sqrt{2}\,\varepsilon_0\hbar}\,\frac{E_P\sqrt{R}}{(\hbar\omega)^2}\left[\sum_{n=1}^{\infty}\frac{4\pi R}{n^3}\,\delta\left(\hbar\omega - E_0 + \frac{R}{n^2}\right) + \frac{2\pi H(\hbar\omega - E_0)}{1-\exp\left(-2\pi\sqrt{R/\hbar\omega - E_0}\right)}\right]$$

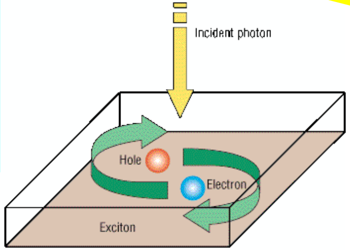


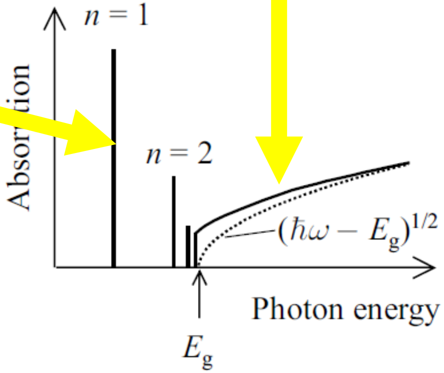
Tanguy's contributions:

- Add Lorentzian broadening
- Kramers-Kronig transform to get the real part.

bound excitons

exciton continuum enhancement

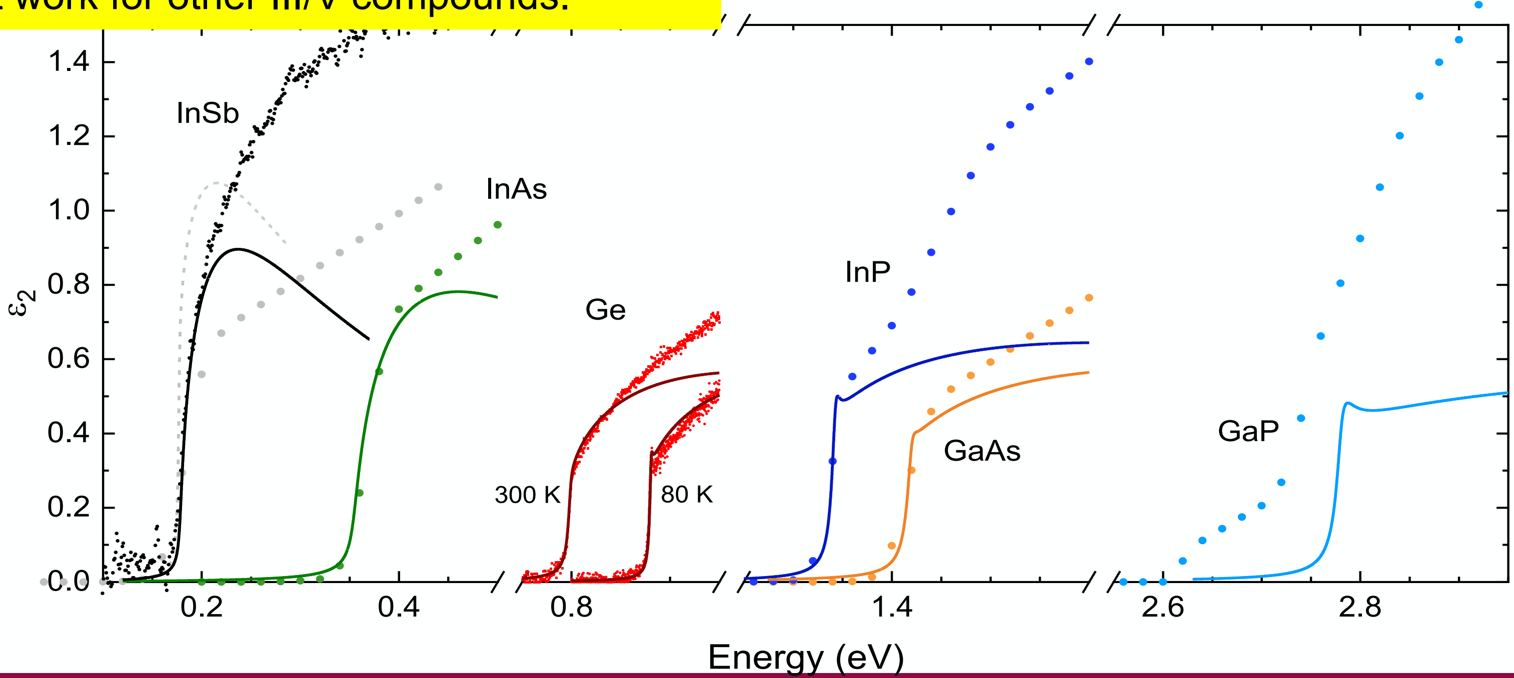




R. J. Elliott, Phys. Rev. **108**, 1384 (1957).
Christian Tanguy, Phys. Rev. Lett. **75**, 4090 (1995) + (E)

Calculation of absorption spectrum from k·p theory

Can we calculate the absorption spectrum?
Yes, we can for Ge in the low carrier density limit.
It does not work for other III/V compounds.



Carola Emminger *et al.*, JAP **131**, 165701 (2022).

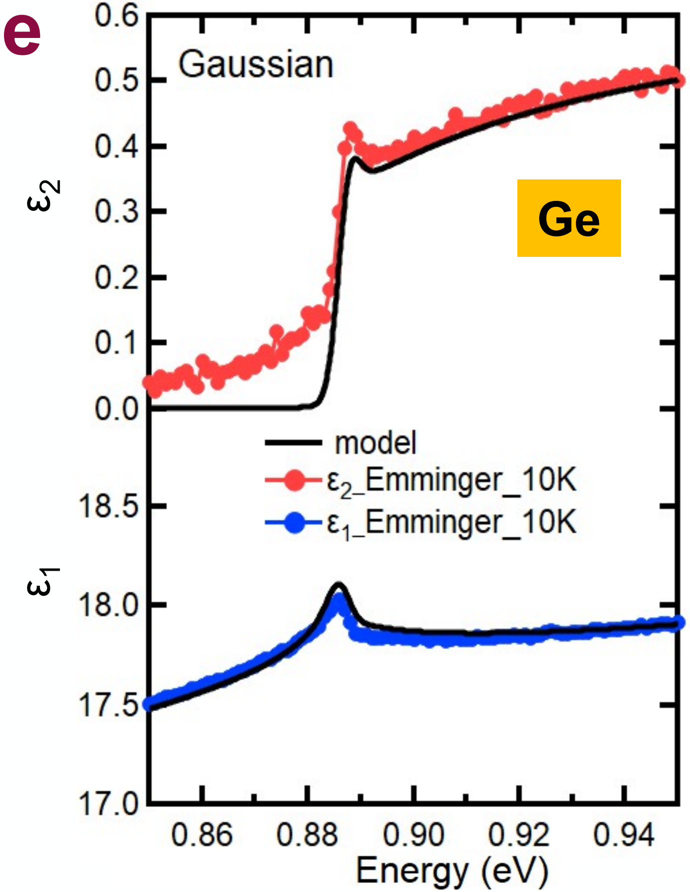
Elliott-Tanguy theory applied to Ge

- **Fixed parameters:**
- Electron and hole masses (temperature dependent)
- Excitonic binding energy R
- Amplitude A (derived from matrix element P)

- **Adjustable parameters:**
- Broadening Γ: 2.3 meV
- Band gap E_0
- Linear background A_1 and B_1 (contribution from E_1 to real part of ε)

Quantitative agreement

- **Problems:**
- Broadening below the gap (band tail, oxide correction)



Carola Emminger *et al.*, JAP **131**, 165701 (2022).

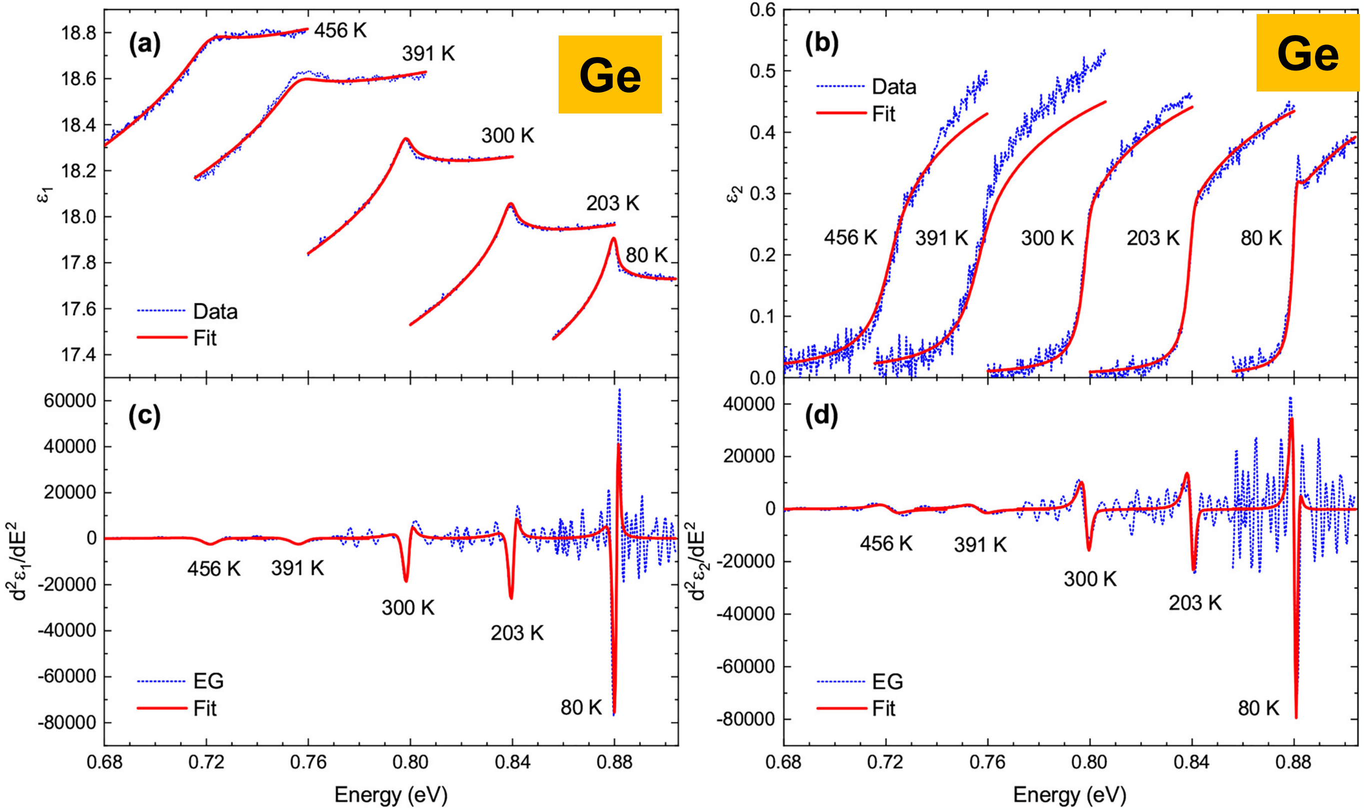
Elliott-Tanguy theory applied to Ge

Good agreement at low temperatures.

Model also describes second derivatives.

Potential problems:

- Matrix element k-dependent
- Nonparabolicity
- Resonant indirect absorption
- ??? at high T.



Carola Emminger *et al.*, JAP **131**, 165701 (2022).

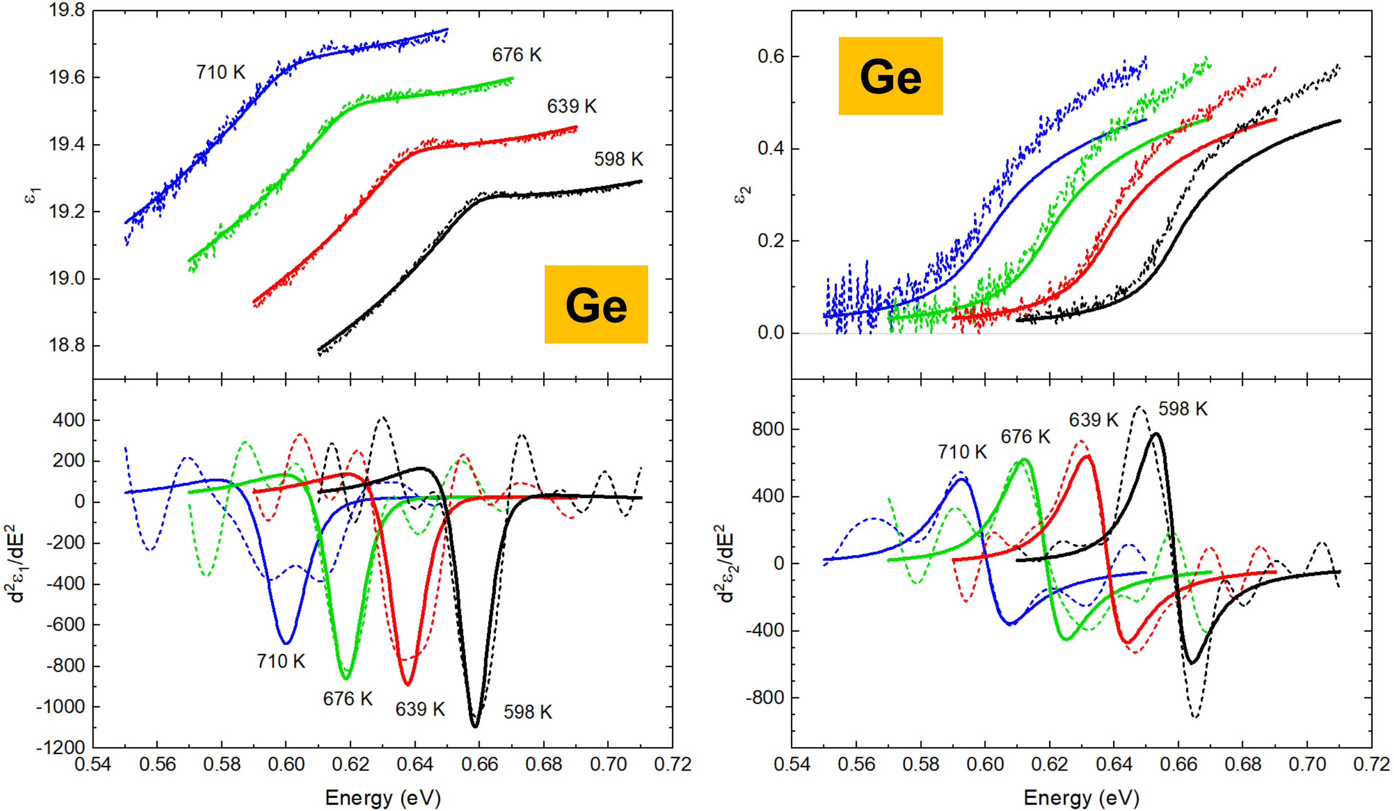
Elliott-Tanguy theory: problems for Ge at high T

Good agreement at low temperatures.

Model also describes second derivatives.

Potential problems:

- Matrix element k-dependent
- Nonparabolicity
- Resonant indirect absorption
- Temperature dependence of the effective mass.



Carola Emminger *et al.*, JAP **131**, 165701 (2022).

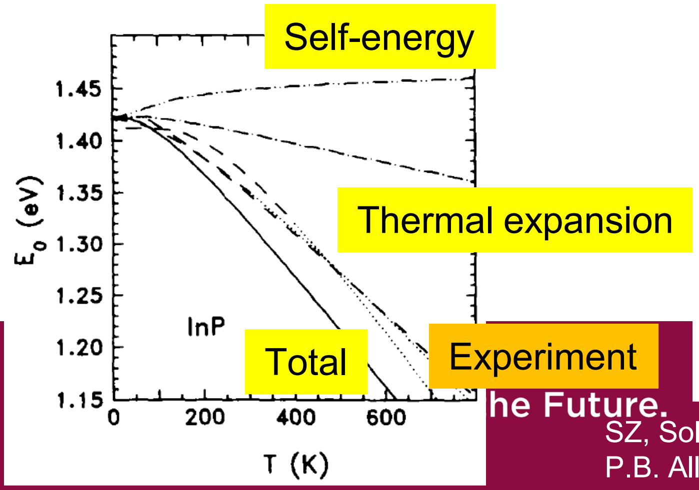
Temperature dependence of the effective mass

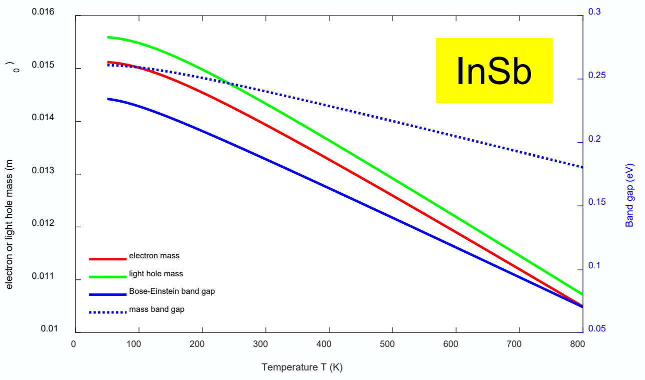
- Effective electron mass given by k·p theory

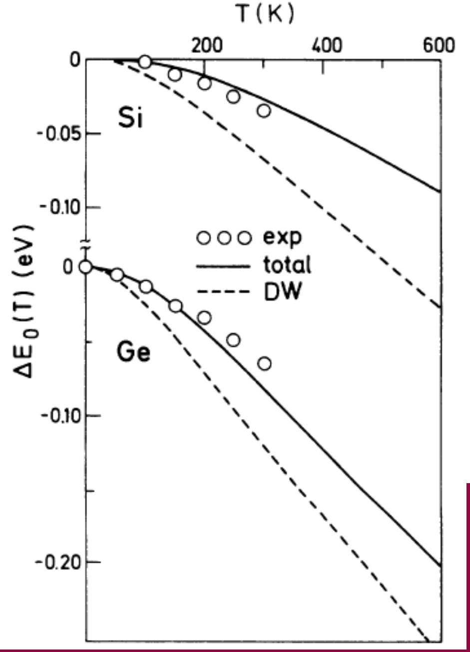
$$\frac{1}{m_e(T)} = 1 + \frac{E_P}{3}\left(\frac{2}{E_0(T)} + \frac{1}{E_0(T)+\Delta_0}\right)$$

E_0: direct band gap
k·p matrix element *P*: $E_P = 2P^2/m_0$

- Temperature dependence of the direct band gap has two contributions:
  - Thermal expansion of the lattice
  - Electron-phonon scattering (Debye-Waller term and self-energy)
- "Mass band gap" should **only include the thermal expansion**.

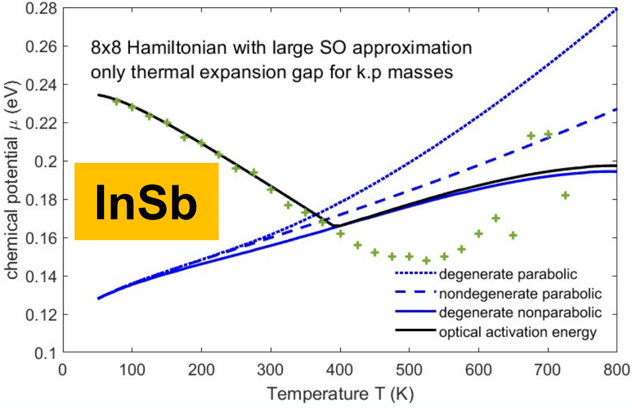


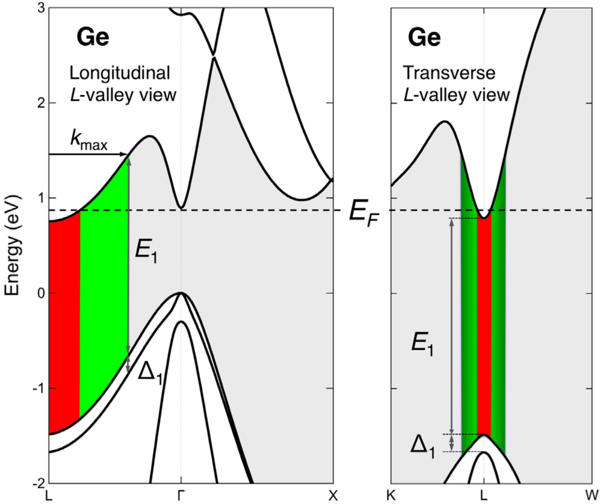


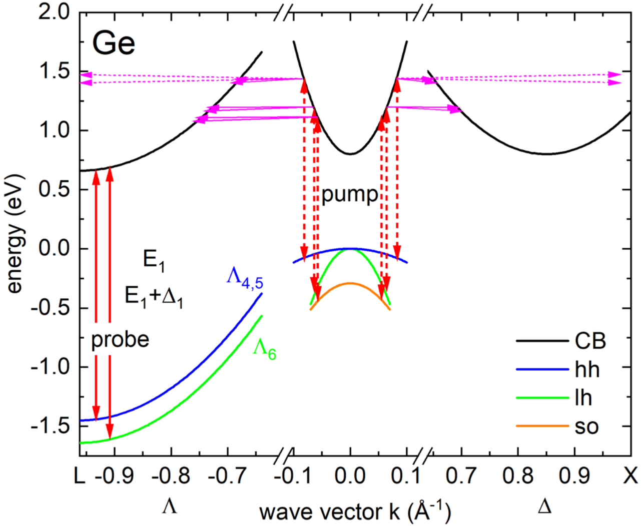


SZ, Solid State Commun. **77**, 485 (1991).
P.B. Allen and M. Cardona, Phys. Rev. B **27** 4760 (1983).

Optical Absorption at High Carrier Densities







High temperature
(thermal excitation of e-h pairs)
constant m and E_0

High n-doping of Ge with P
(free electrons pile up at L-point)

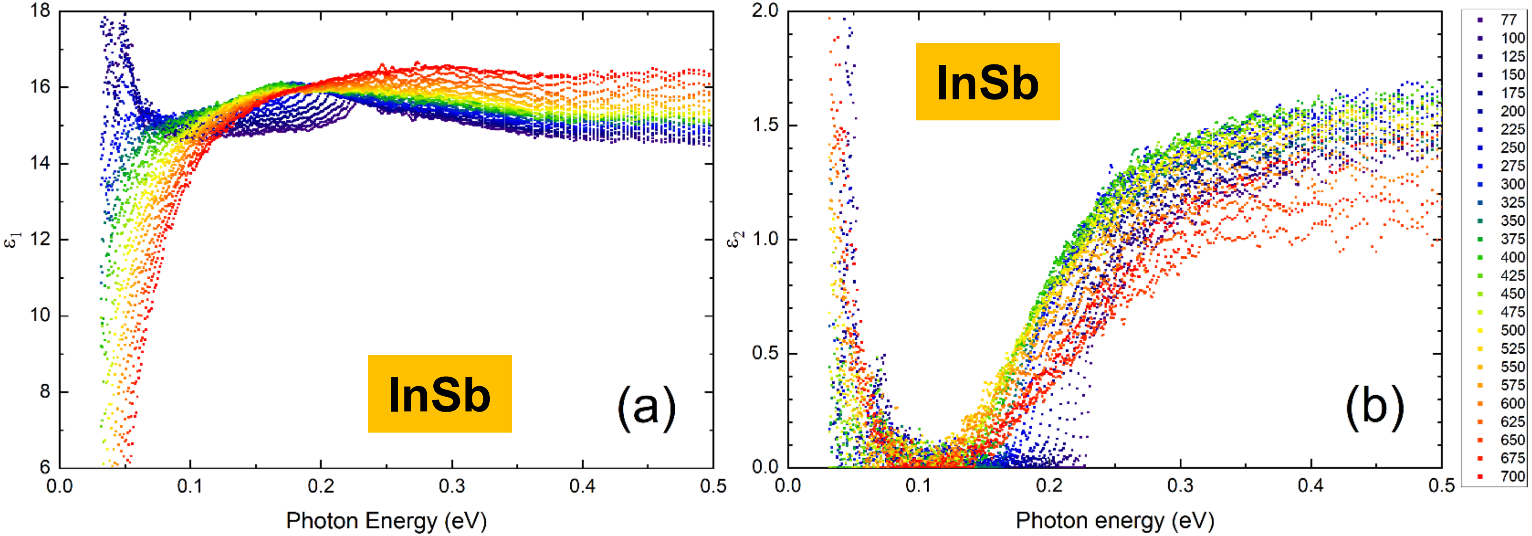
Intense femtosecond **laser excitation** (ELI Beamlines)
(electrons pile at L-point)

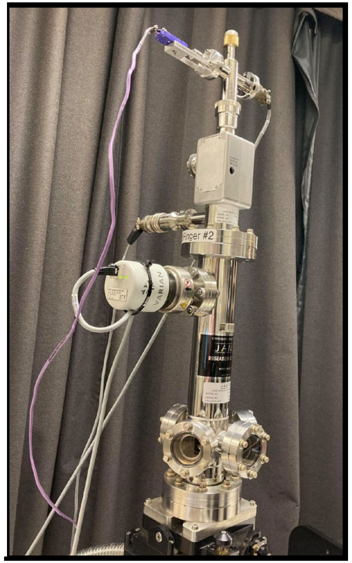
Rivero, JVSTB **41**, 022203 (2023)

Xu et al., PRL **118**, 267402 (2017)

Espinoza, APL **115**, 052105 (2019)

(1) Dielectric function of InSb from 80 to 800 K





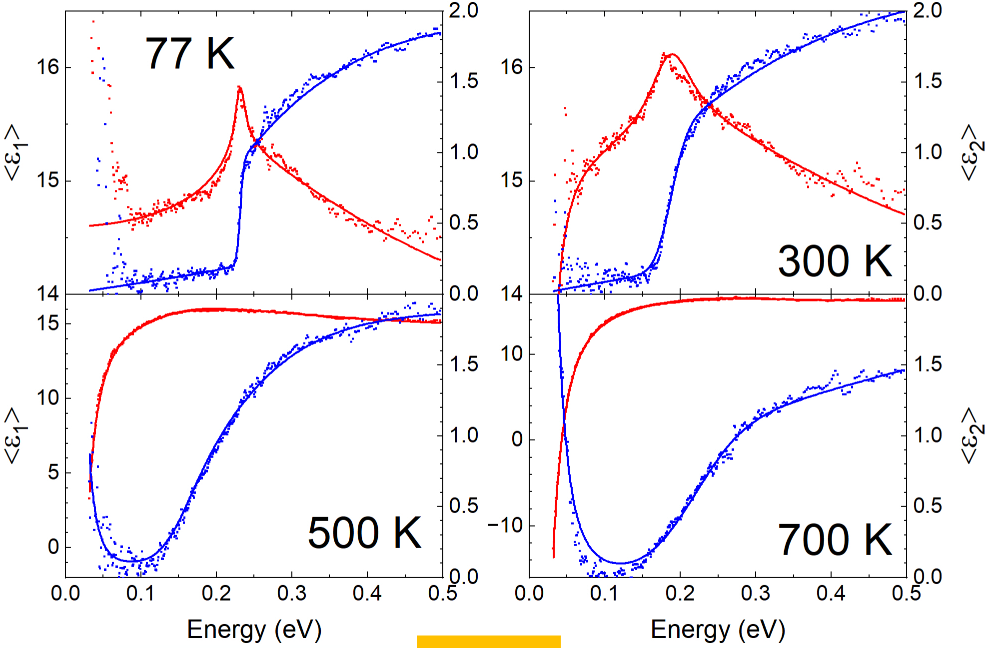
Woollam FTIR-VASE cryostat with CVD diamond windows

- **Band gap** changes with temperature (but only below 500 K).
- **Amplitude reduction at high temperatures (Pauli blocking, bleaching)**
- **Drude response** at high temperatures (thermally excited carriers).
- Depolarization artifacts at long wavelengths (below 300 K).

Rivero Arias, JVSTB **41**, 022203 (2023)

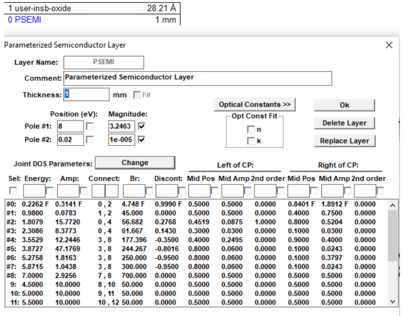
Band gap analysis for InSb

How does the band gap of InSb change with temperature?



InSb

Parametric-Semiconductor Model:



1 user-insb-oxide	28.21 Å
0 PSEMI	1 mm

Parameterized Semiconductor Layer

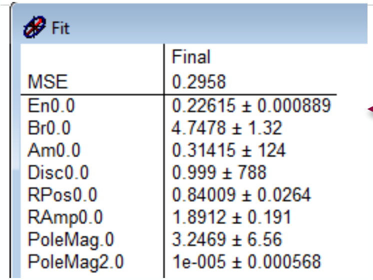
Layer Name: PSEMI

Comment: Parameterized Semiconductor Layer

Thickness: 1 mm ☐ Fit

	Position (eV):		Magnitude:	
Pole #1:	8	☐	3.2463	☑
Pole #2:	0.02	☐	1e-005	☑

						Left of CP:			Right of CP:		
Sel:	Energy:	Amp:	Connect:	Br:	Discont:	Mid Pos	Mid Amp	2nd order	Mid Pos	Mid Amp	2nd order
#0:	0.2262 F	0.3141 F	0 , 2	4.748 F	0.9990 F	0.5000	0.5000	0.0000	0.8401 F	1.8912 F	0.0000
#1:	0.9800	0.0783	1 , 2	45.000	0.0000	0.5000	0.5000	0.0000	0.4000	0.7500	0.0000
#2:	1.8079	15.7720	0 , 4	56.682	0.2768	0.4519	0.0875	1.0000	0.8000	0.5204	0.0000
#3:	2.3086	8.3773	0 , 4	61.667	0.1430	0.3000	0.0300	0.0000	0.1000	0.0300	0.0000
#4:	3.5529	12.2446	3 , 8	177.396	-0.3500	0.4000	0.2495	0.0000	0.9000	0.4000	0.0000
#5:	3.8727	47.1769	3 , 8	244.267	-0.8016	0.8000	0.0600	0.0000	0.1000	0.0243	0.0000
#6:	5.2758	1.8163	3 , 8	250.000	-0.9500	0.8000	0.0600	0.0000	0.1000	0.3797	0.0000
#7:	5.8715	1.0438	3 , 8	300.000	-0.9500	0.8000	0.0600	0.0000	0.1000	0.0243	0.0000
#8:	7.0000	2.9256	7 , 8	700.000	0.0000	0.5000	0.5000	0.0000	0.5000	0.5000	0.0000
9:	4.5000	10.0000	8 , 10	50.000	0.0000	0.5000	0.5000	0.0000	0.5000	0.5000	0.0000
10:	5.0000	10.0000	9 , 11	50.000	0.0000	0.5000	0.5000	0.0000	0.5000	0.5000	0.0000
11:	5.5000	10.0000	10 , 12	50.000	0.0000	0.5000	0.5000	0.0000	0.5000	0.5000	0.0000



Fit

	Final
MSE	0.2958
En0.0	0.22615 ± 0.000889
Br0.0	4.7478 ± 1.32
Am0.0	0.31415 ± 124
Disc0.0	0.999 ± 788
RPos0.0	0.84009 ± 0.0264
RAmp0.0	1.8912 ± 0.191
PoleMag.0	3.2469 ± 6.56
PoleMag2.0	1e-005 ± 0.000568

Also vary “shape parameters”.

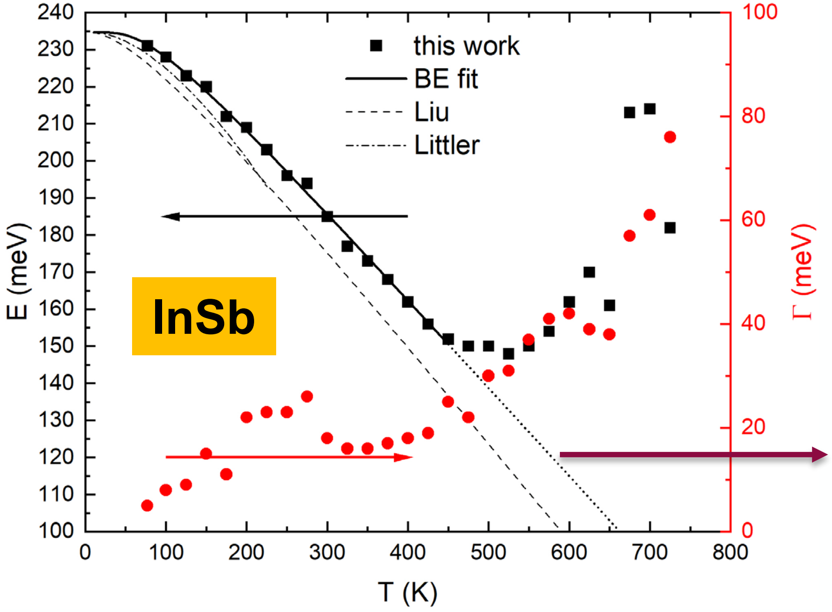
Asymmetric peak shape poorly described.

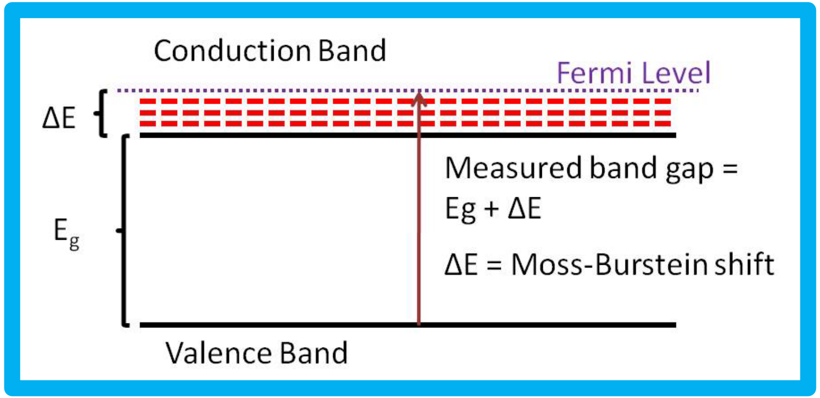
Try Tanguy oscillator for excitonic line shape.

BE BOLD. Shape the Future.

C. M. Herzinger, B. Johs, et al., J. Appl. Phys. **83**, 3323 (1998)
Rivero Arias, JVSTB **41**, 022203 (2023)

Band gap of InSb from 80 to 800 K




Conduction Band

Fermi Level

ΔE

E_g

Measured band gap = Eg + ΔE

ΔE = Moss-Burstein shift

Valence Band

Bose-Einstein Model

$$E_0(T) = E^{\mathrm{un}} - b\left[1 + \frac{2}{\exp\left({}^{\Omega}/_{k_B T}\right)}\right]$$

- Band gap changes with temperature (but only below 500 K)
- Described by Bose-Einstein model below 500 K: Logothetidis, PRB **31**, 947 (1985).
- No redshift above 500 K: **Thermal Burstein-Moss shift**

T.S. Moss, Proc. Phys. Soc. **67**, 775 (1954).
E. Burstein, Phys. Rev. **93**, 632 (1954).

k·p theory (band structure method)

Schrödinger equation

$$H\Phi_{n\vec{k}} = \left(\frac{\vec{p}^2}{2m_0} + V\right)\Phi_{n\vec{k}} = E_{n\vec{k}}\Phi_{n\vec{k}}$$

Use Bloch's theorem:

$$\Phi_{n\vec{k}}(\vec{r}) = e^{i\vec{k}\cdot\vec{r}}u_{n\vec{k}}(\vec{r})$$

Product rule

$$(fg)'' = f''g + 2f'g' + fg''$$

Solve equation for **k**=0.

$$\left(\frac{\vec{p}^2}{2m_0} + \frac{\hbar^2\vec{\boldsymbol{k}}^2}{2\boldsymbol{m_0}} + \frac{\hbar\vec{\boldsymbol{k}}\cdot\vec{\boldsymbol{p}}}{\boldsymbol{m_0}} + V\right)u_{n\vec{k}} = E_{n\vec{k}}u_{n\vec{k}}$$

Eliminate green free-electron term with substitution of variables (Kane 1957).

Then treat red term in perturbation theory.

Works very well for semiconductors with local V(**r**) potentials.

Yu & Cardona, Fundamentals of Semiconductors
Kane, J. Phys. Chem. Solids **1**, 249 (1957). Kane 1966.

Nonparabolicity of InSb conduction band from k·p theory

Kane 8x8 k·p Hamiltonian:

$$\tilde{H}_{\vec{k}} = \begin{pmatrix} E_0 & 0 & -\frac{\hbar\vec{k}}{m_0}iP & 0 \\ 0 & -\frac{2\Delta_0}{3} & \frac{\sqrt{2}\Delta_0}{3} & 0 \\ \frac{\hbar\vec{k}}{m_0}iP & \frac{\sqrt{2}\Delta_0}{3} & -\frac{\Delta_0}{3} & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}$$

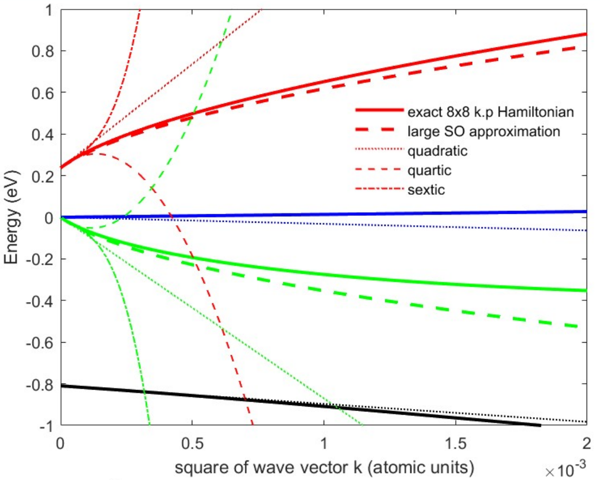
Cubic characteristic equation:

$$\tilde{E}\left(\tilde{E}-E_0\right)\left(\tilde{E}+\Delta_0\right)-\frac{\hbar^2k^2E_P}{2m_0}\left(\tilde{E}+\frac{2\Delta_0}{3}\right)=0$$

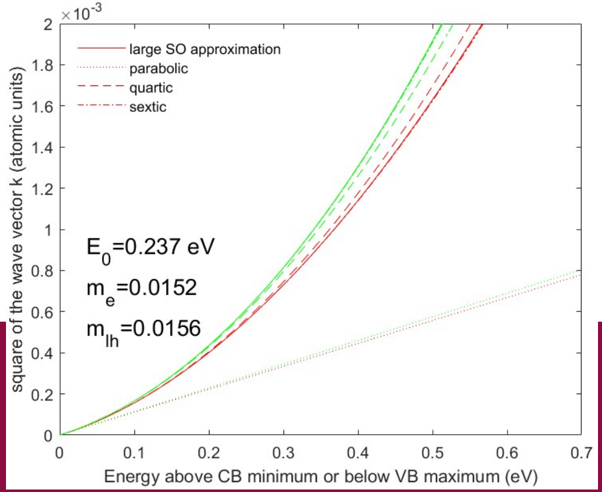
Large spin-orbit approximation:

$$E_{3,4}=\frac{\hbar^2k^2}{2m_0}+\frac{E_0}{2}\left(1\pm\sqrt{1+\frac{\hbar^2k^2}{2m_0}\frac{2}{\mu_{lh}E_0}}\right)$$

Kane, J. Phys. Chem. Solids **1**, 249 (1957).



Energy versus k

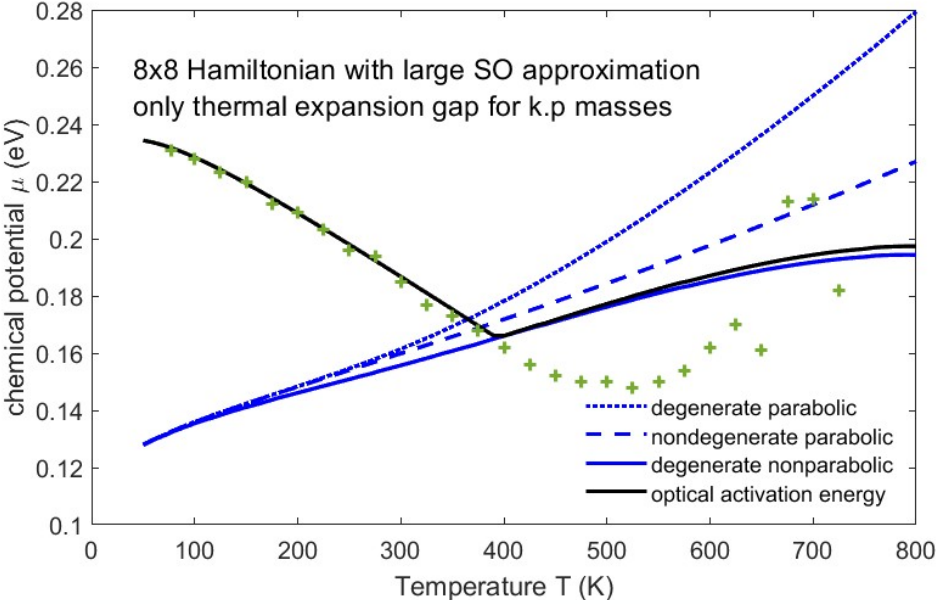


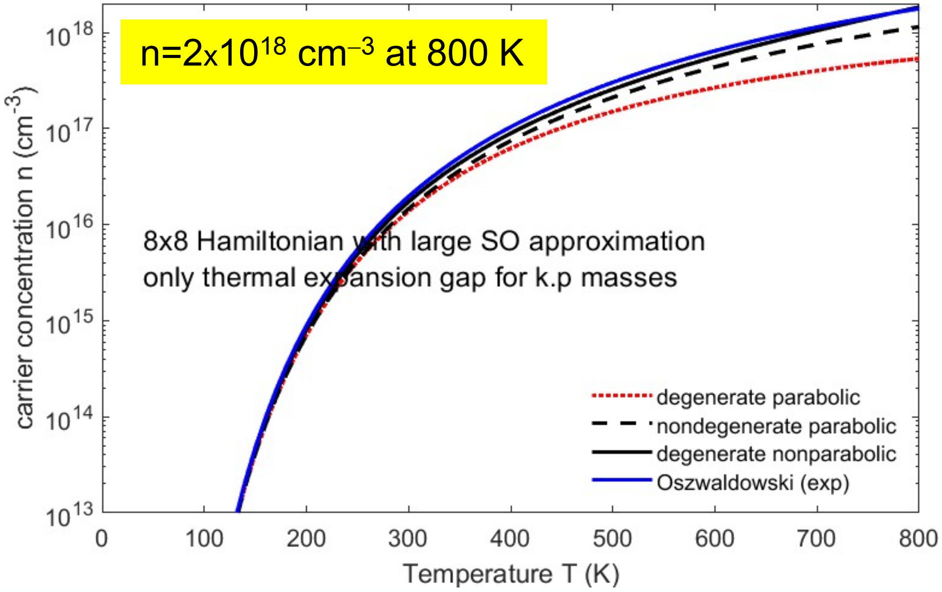
Density of CB states

$$\frac{\hbar^2k^2}{2m_0m^*}=\varepsilon\left(1+\alpha\varepsilon+\beta\epsilon^2\right)$$

$$\alpha=\frac{(1-m^*)^2}{E_0}$$

Thermal excitations of electron-hole pairs in InSb


8x8 Hamiltonian with large SO approximation
only thermal expansion gap for k.p masses


$n=2\times10^{18}$ cm^{-3} at 800 K

8x8 Hamiltonian with large SO approximation
only thermal expansion gap for k.p masses

$k_BT=E_g/4$ at 600 K
Fermi level above
conduction band edge above 450 K.

Thermal Burstein-Moss shift
Drude response of free carriers
Reduction of absorption coefficient

M. Rivero Arias *et al.*, JVSTB **41**, 022203 (2023).
Oswaldowski/Zimpel, J. Phys. Chem. Solids **49**, 1179 (1988).
D. L. Rode, Phys. Rev. B **3**, 3287 (1971).

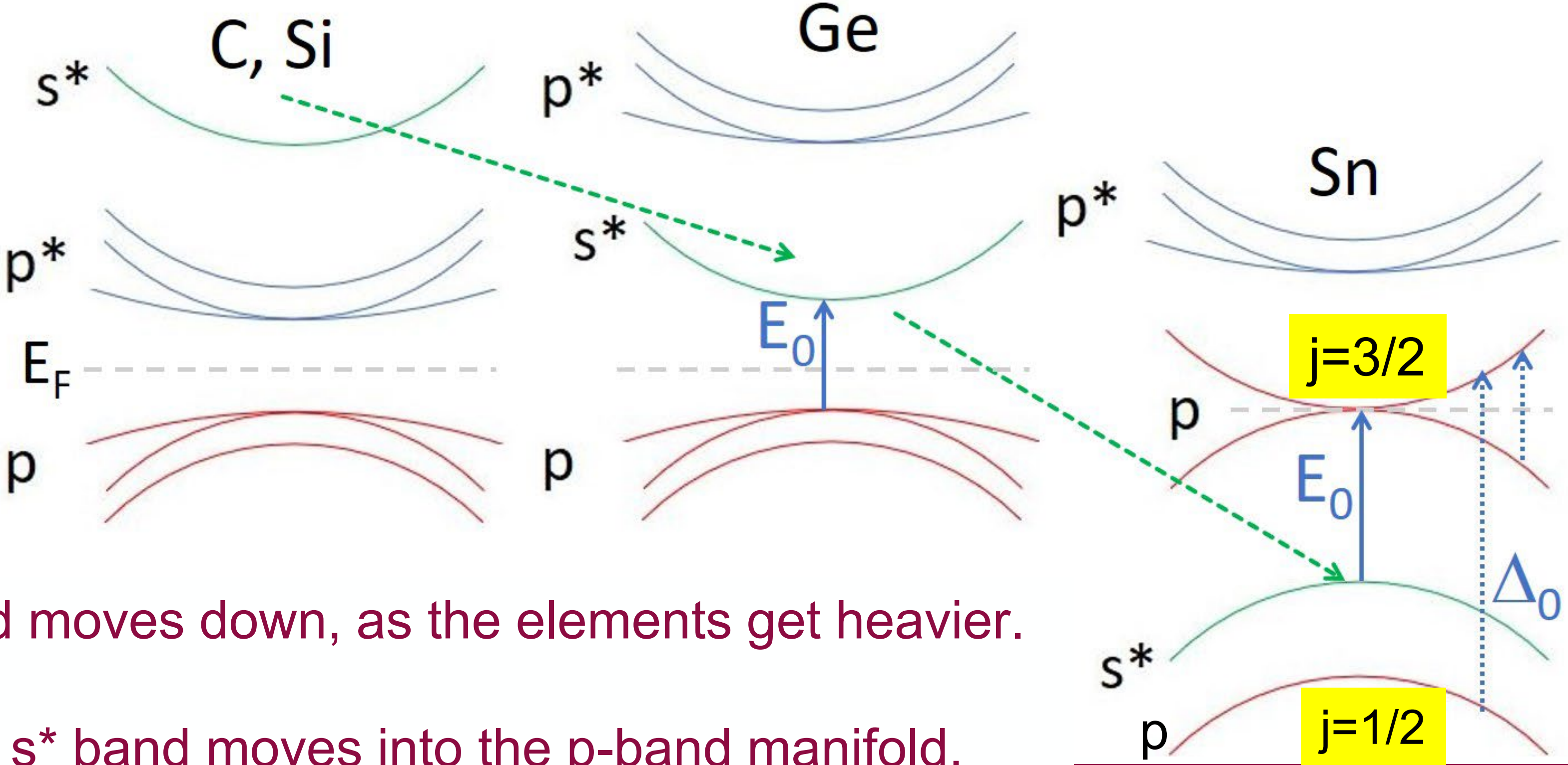
Optical constants model: screened excitons

$$\varepsilon_2(E) = \frac{2\pi A\sqrt{R}}{E^2}\left\{\sum_{n=1}^{\sqrt{g}} \frac{2R}{n}\left(\frac{1}{n^2}-\frac{n^2}{g^2}\right)\delta\left[E-E_0+\frac{R}{n^2}\left(1-\frac{n^2}{g}\right)^2\right]+\frac{\sinh(\pi g k)\mathrm{H}(E-E_0)}{\cosh(\pi g k)-\cosh\left(\pi g\sqrt{k^2-\frac{4}{g}}\right)}\right\}[f_h(E)-f_e(E)]$$

- **Absorption by screened excitons** (Hulthen potential)
- **Degenerate Fermi-Dirac statistics** to calculate f_h and f_e.
- Numerical Kramers-Kronig transform (need occupation factors)
- Two terms for light and heavy excitons
- **Non-parabolicity and temperature-dependent mass** included from k.p theory
- **k-dependent matrix element *P***.
- Screening parameter $g=12/\pi^2 a_R k_{TF}$ (large: no screening)
  Sommerfeld enhancement persists well above the Mott density.
- **Only two free parameters: Band gap E_0 and broadening Γ**
- Amplitude *A* and exciton binding energy *R* from k.p theory and effective masses

Christian Tanguy, Phys. Rev. B **60**, 10660 (1999).
Jose Menendez, Phys. Rev. B **101**, 195204 (2020).
Carola Emminger, J. Appl. Phys. **131**, 165701 (2022).

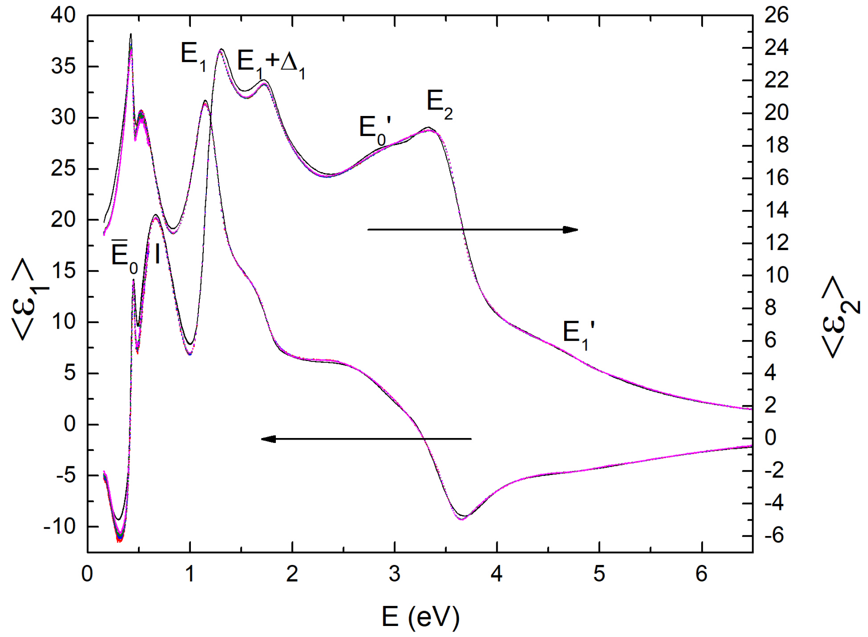
Relativistic Effects: Darwin Shift: C, Si, Ge, Sn

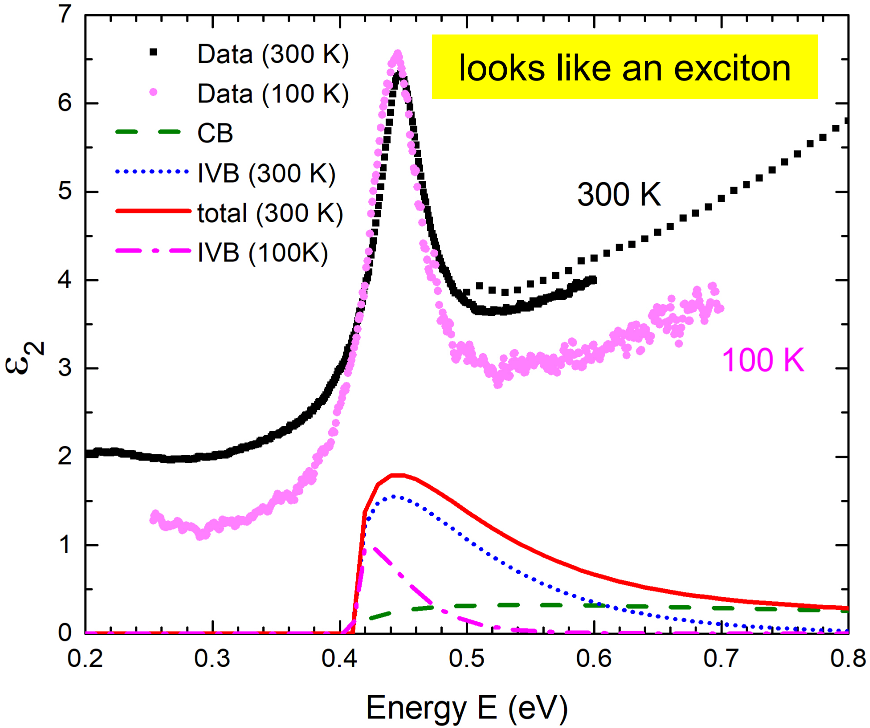


The s* band moves down, as the elements get heavier.

In α-tin, the s* band moves into the p-band manifold, between the j=1/2 and j=3/2 states.
This makes α-tin an (**inverted**) **gapless** semiconductor.

Intravalence band absorption in gapless topological insulators (α-tin)



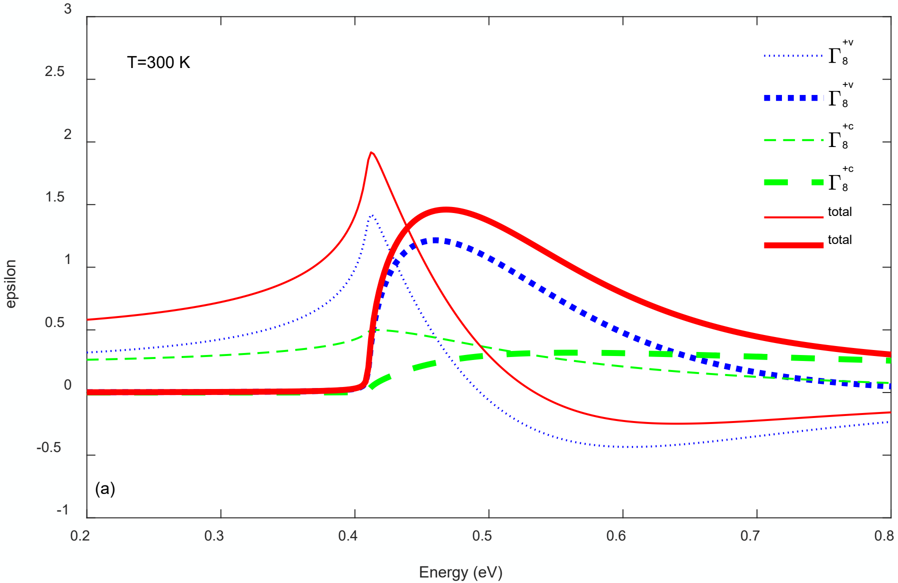


looks like an exciton

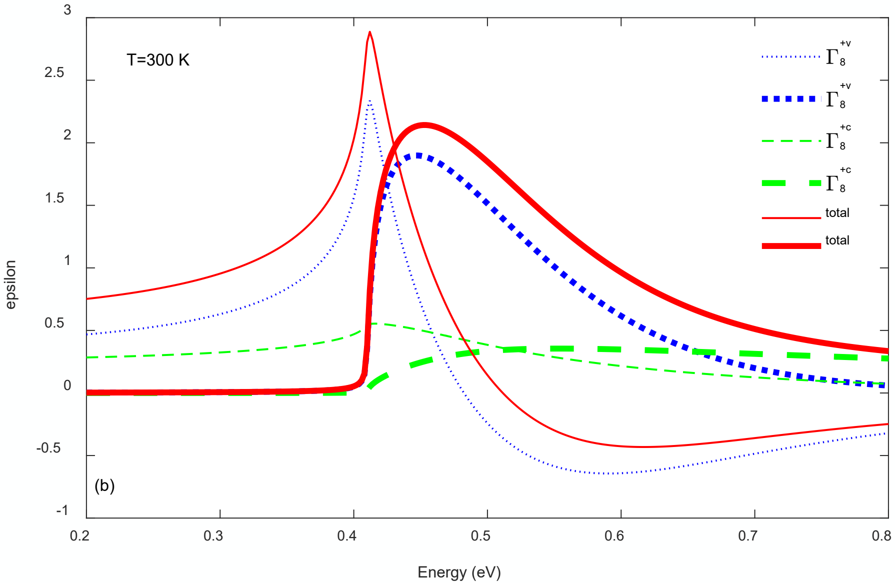
R.A. Carrasco, APL **113**, 232104 (2018).

All gapless (inverted) semiconductors should have this peak.
Theory with same model as Ge IVB (Kaiser 1953, Kahn 1955).

Excitonic intravalence band absorption in gapless topological insulators (α-tin)

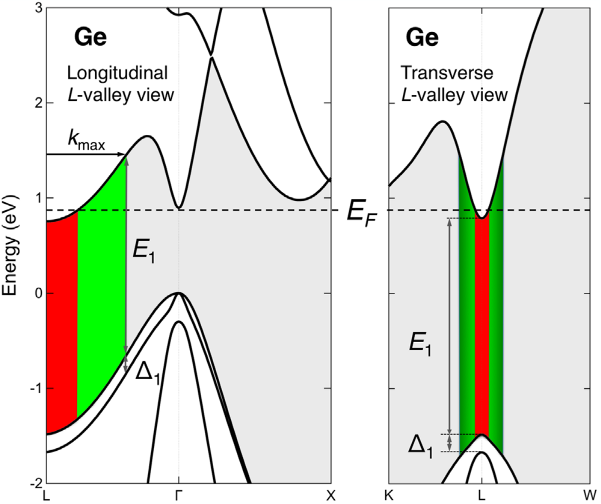


Interband transitions without excitonic effects.



Screened excitons included, parabolic bands. Agreement with experiment is not good.

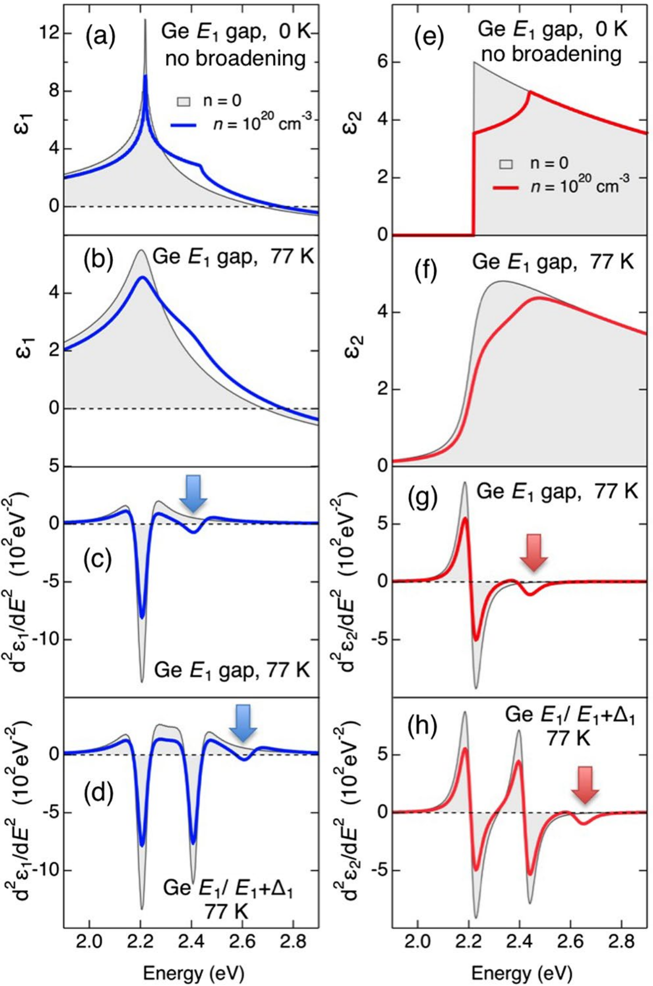
(2) Highly doped Ge (n-type, with phosphorus)

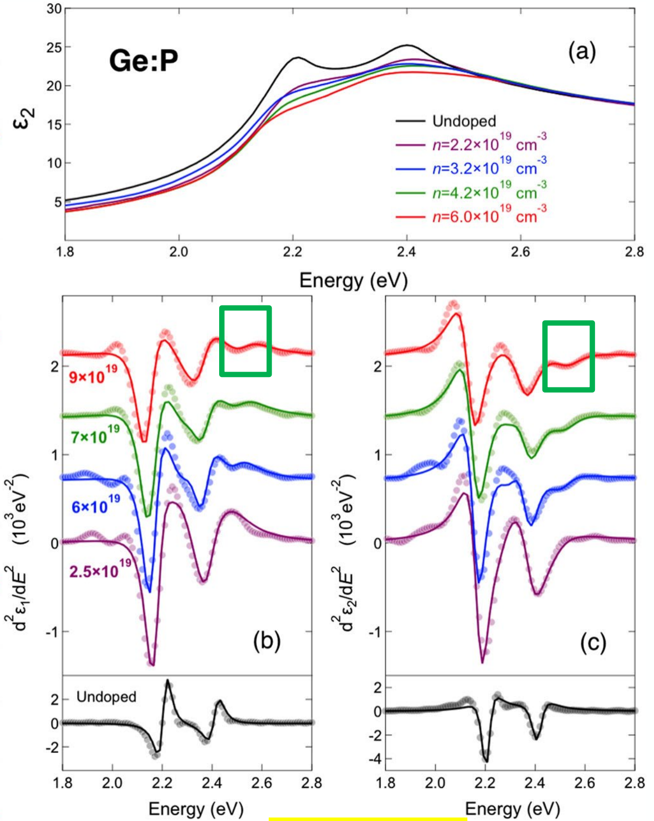


Electrons pile up at the L-point.
Transitions in red region blocked.

Phase filling singularity

No good model for 2D exciton screening

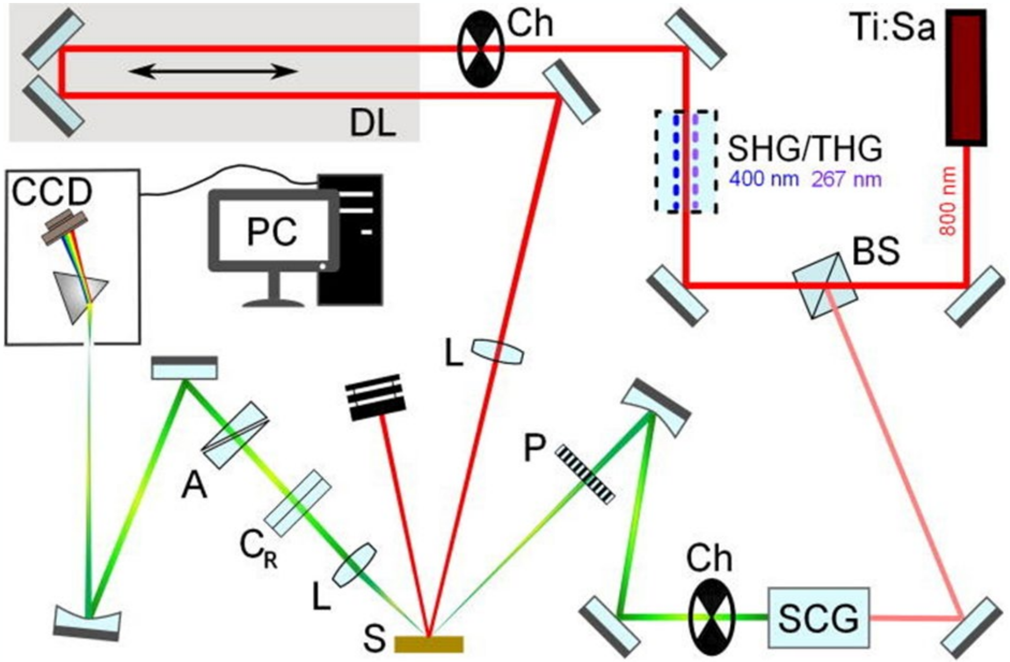




Phase shift

Xu et al., PRL **118**, 267402 (2017)
Xu et al., JAP **125**, 085704 (2019)

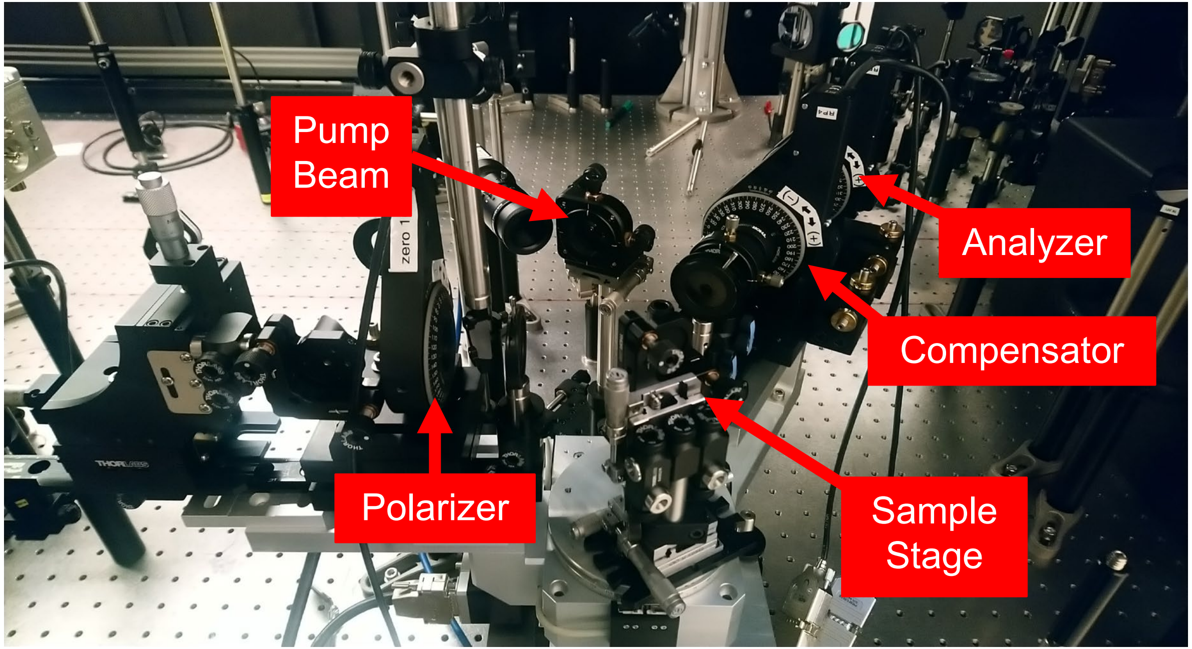
(3) Set-up: Femtosecond pump-probe ellipsometry



- Ch: Chopper (500 Hz, 250 Hz)
- A: Analyzer
- P: Polarizer
- C_R: Rotating compensator
- L: Lens
- S: Sample
- DL: Delay Line (~6.67 ns pump-probe delay and 3 fs resolution)
- BS: Beam splitter
- SHG/THG: $2^{nd}/3^{rd}$ harmonic generation
- SCG: Super-continuum generation
- CCD: Charge-coupled device detector

Espinoza et al., APL **115**, 052105 (2019).

Set-up: Femtosecond pump-probe ellipsometry



Rotating compensator ellipsometer:

Compensator was rotated in steps of 10º for a total of 55-65 angles.

Probe beam of 350-750 nm at 60º incidence angle.

P-polarized pump beam: 35 fs pulses of 800 nm wavelength at 1 kHz repetition rate.

Delay time from -10 to 50 ps.

Time resolution of about 500 fs.

Espinoza et al., APL **115**, 052105 (2019).

ELI Beamlines: ELI ERIC. Dolní Břežany (near Prague)

Third user call was due **October 16th, 2023**: https://up.eli-laser.eu/
Contact Shirly Espinoza: shirly.espinoza@eli-beams.eu

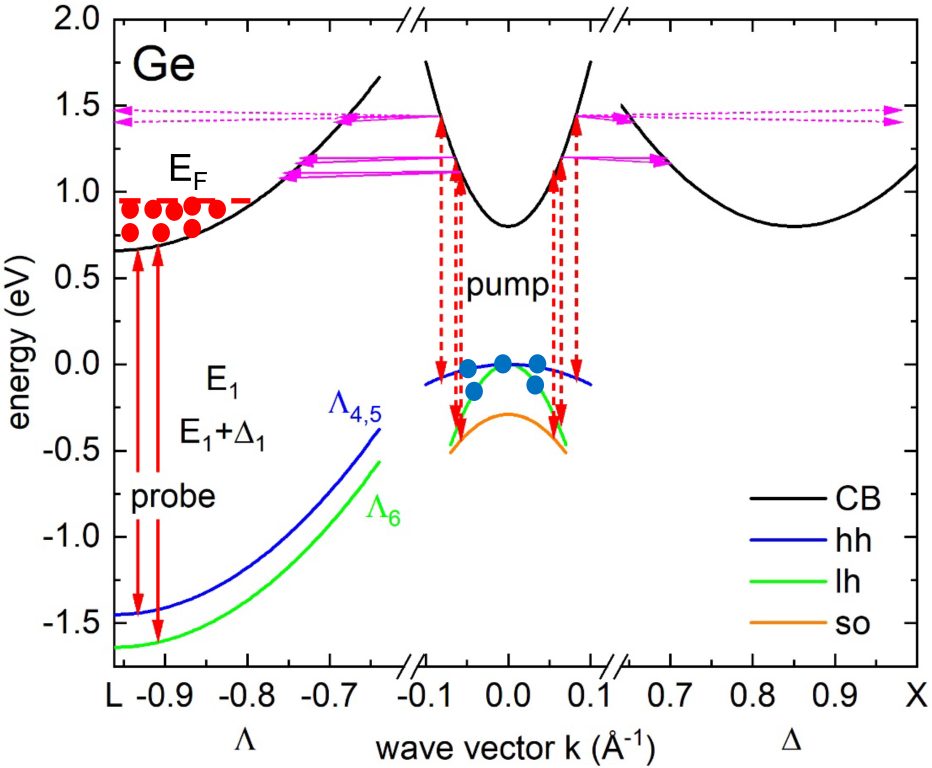
Semiconductors

Metal oxides

Bulk and thin films

Etc.

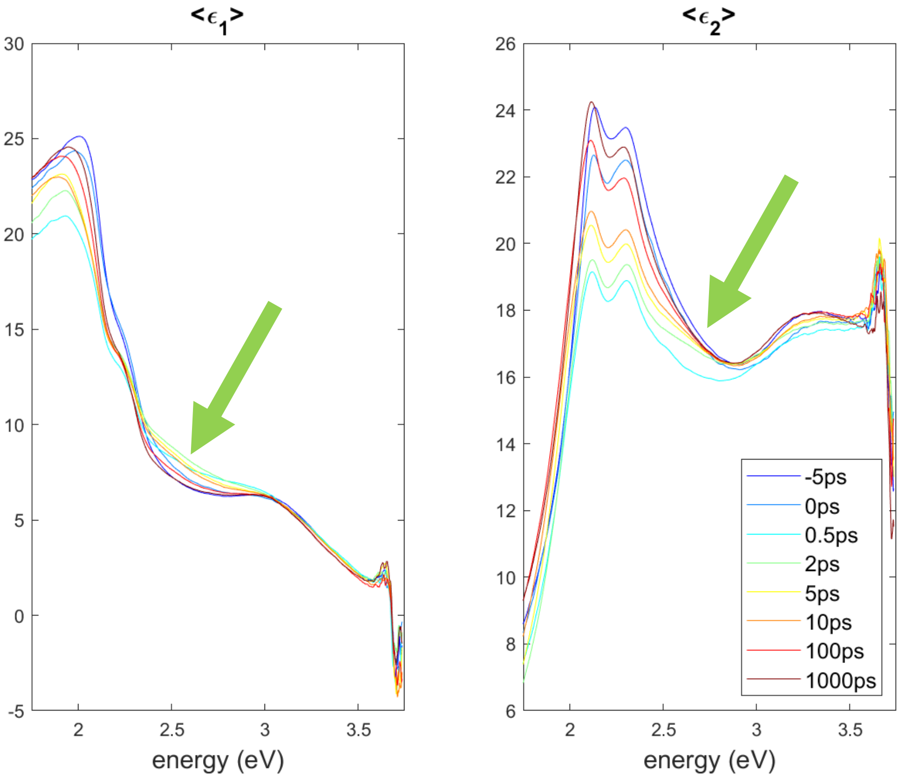
Ultrafast processes in photoexcited Germanium



- 1.55 eV pump beam creates $N=10^{21}$ cm^{-3} electron-hole pairs near Γ-point. 9 mW power.
- Heavy, light, split-off bands.
- Ignore diffusion (for 1.55 eV pump).
- Ignore Auger recombination.
- Thermalization: Fermi-Dirac distribution
- **Intervalley scattering: $\Gamma \rightarrow X \rightarrow L$**
- Electrons accumulate at L.
- Holes remain near Γ.
- Electrons at L block E_1 and $E_1+\Delta_1$ transitions (**Fermi-level singularity**)
- **Bandgap renormalization: redshift expected**
- Lattice heating (25 K): redshift
- **Exciton screening**

Espinoza et al., APL **115**, 052105 (2019).

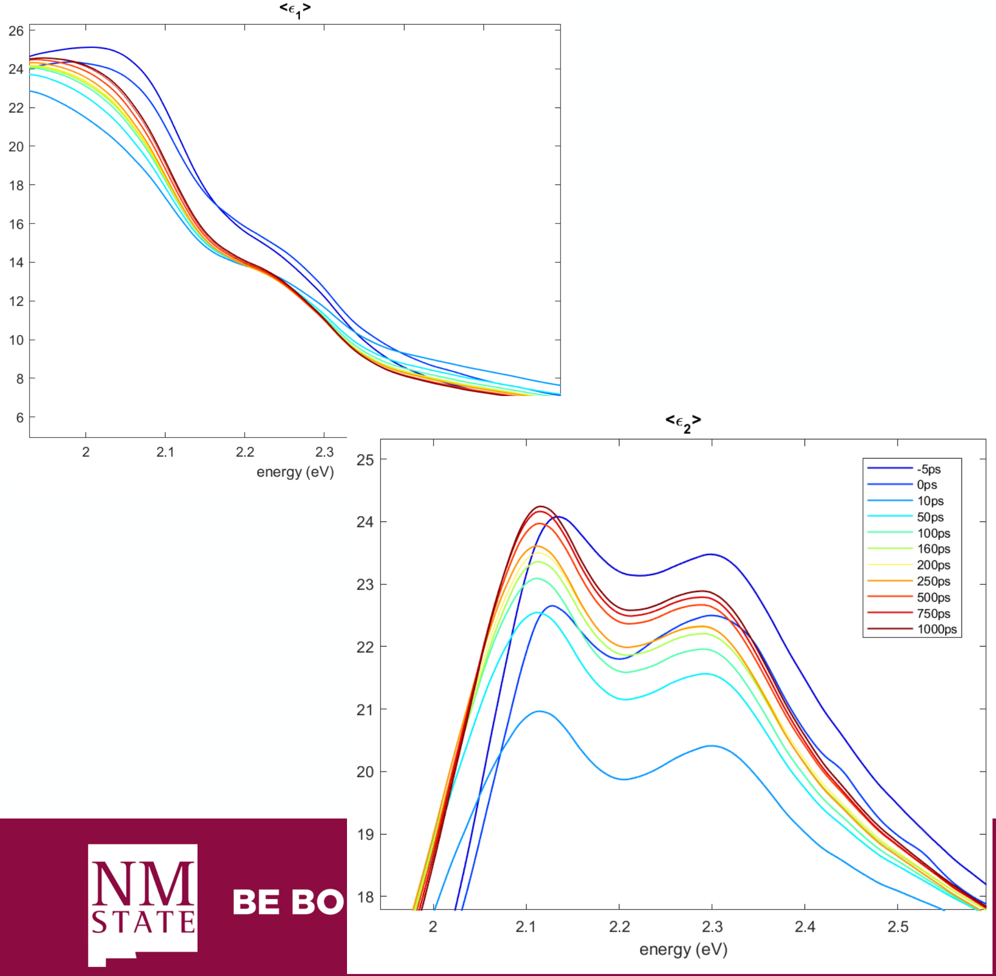
Ultrafast processes in photoexcited Germanium



- 1.55 eV pump beam creates $N=4\times10^{21}$ cm^{-3} electron-hole pairs near Γ-point.
- The E_1 and $E_1+\Delta_1$ peaks decrease within the first two picoseconds and then recover: **Band filling, excitonic screening followed by diffusion, Auger, and radiative recombination.**
- **Maybe a Fermi level singularity near 2.6 eV.**
- **Detailed modeling is required (Tanguy 2-D excitonic line shapes, including Fermi level singularity.)**

Carlos Armenta, February 2023, unpublished.

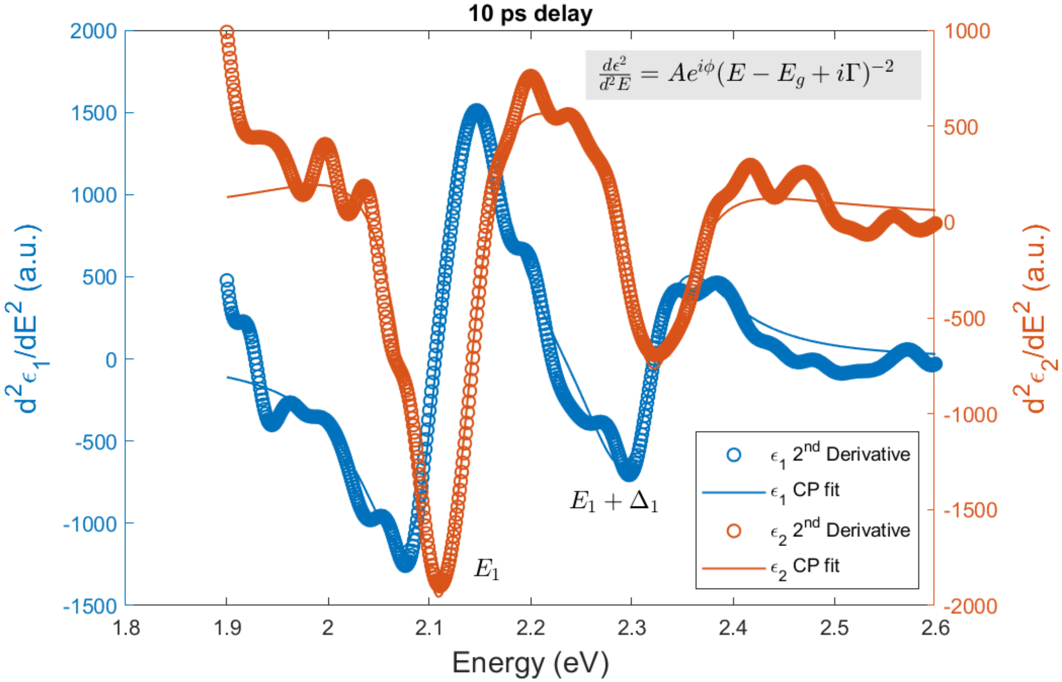
Ultrafast processes in photoexcited Germanium

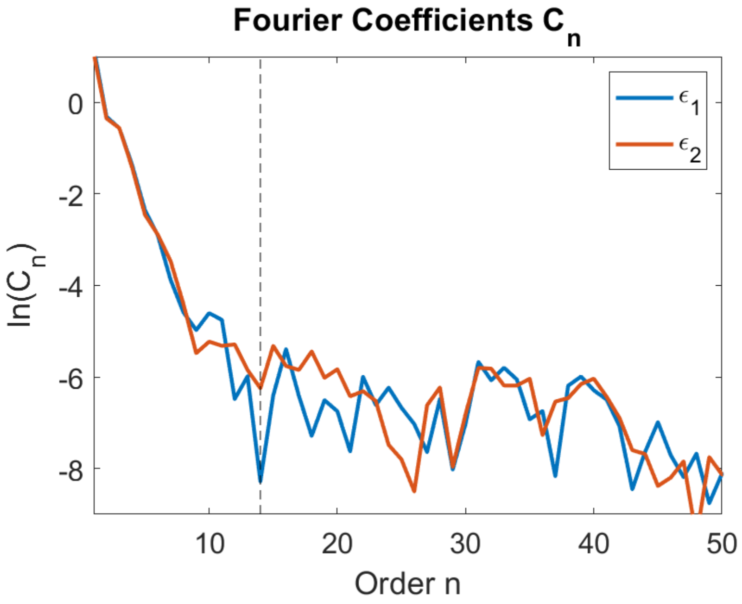


- 1.55 eV pump beam creates N=4x10^{21} cm^{-3} electron-hole pairs near Γ-point.
- The E_1 and $E_1+\Delta_1$ peaks decrease within the first two picoseconds and then recover.
  (Band filling, excitonic screening followed by diffusion, Auger, and radiative recombination)
- Maybe a Fermi level singularity near 2.6 eV.
- **It looks like there is a redshift, but we need a line shape analysis with derivatives.**

Carlos Armenta, February 2023, unpublished.

Derivative analysis: critical point parameters



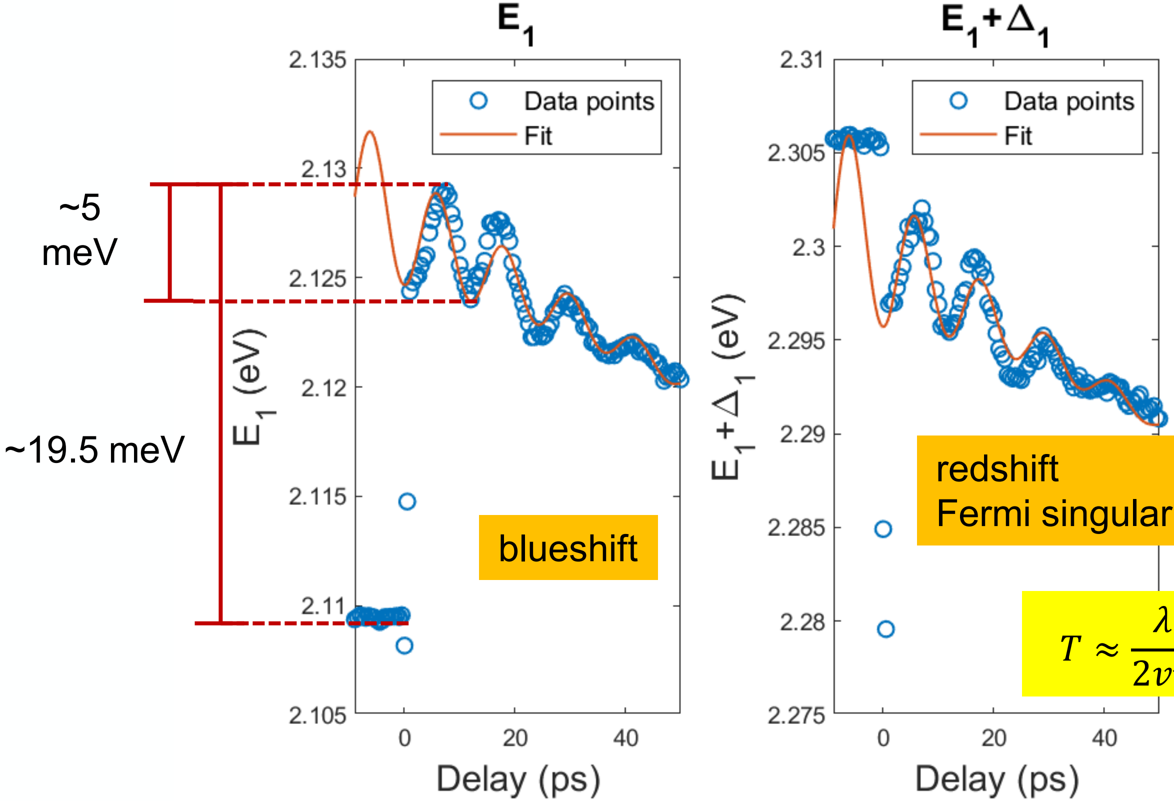


Extended Gauss filter
Filter width determined from Fourier coefficients

BE BOLD. Shape the Future.

Carlos Armenta, February 2023, unpublished.
Emminger *el al.*, PSS RRL **2022**, 2200058.
Le, JVST B **37**, 052903 (2019);

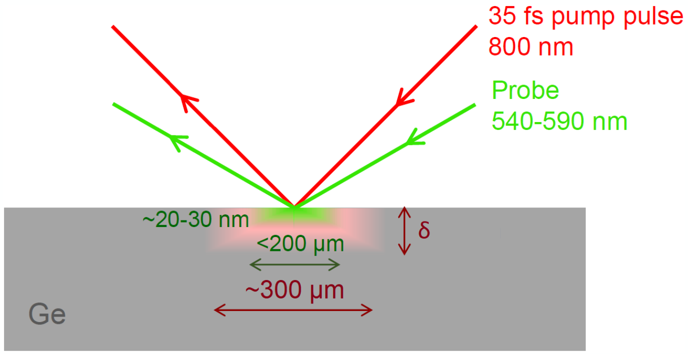
Coherent acoustic phonon oscillations

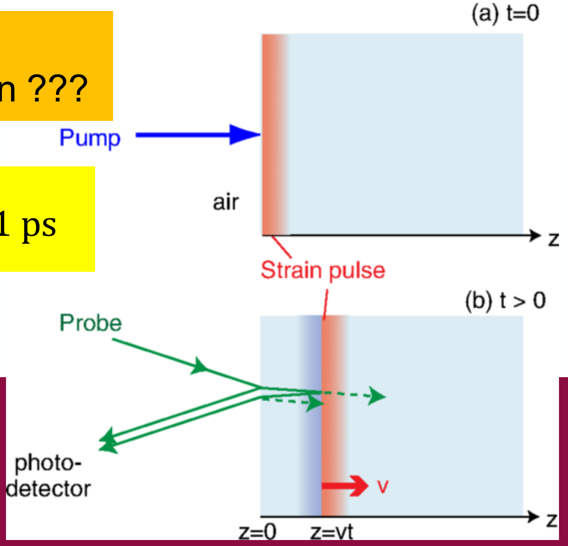


blueshift

redshift
Fermi singularity or strain ???

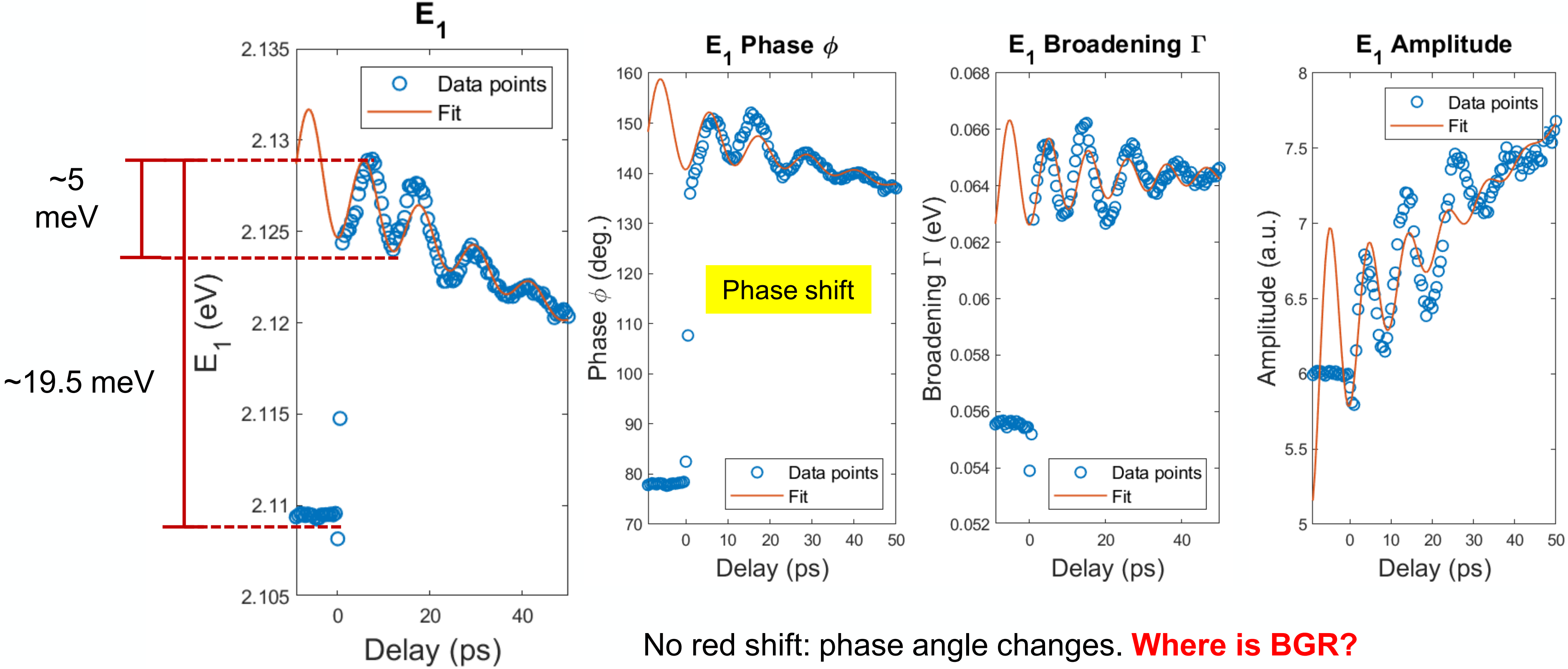
$$T \approx \frac{\lambda}{2vn} = 9 - 11\ \text{ps}$$





Carlos Armenta, February 2023, unpublished.
Emminger *el al.*, PSS RRL **2022**, 2200058.

Critical point parameters for E_1 and $E_1+\Delta_1$

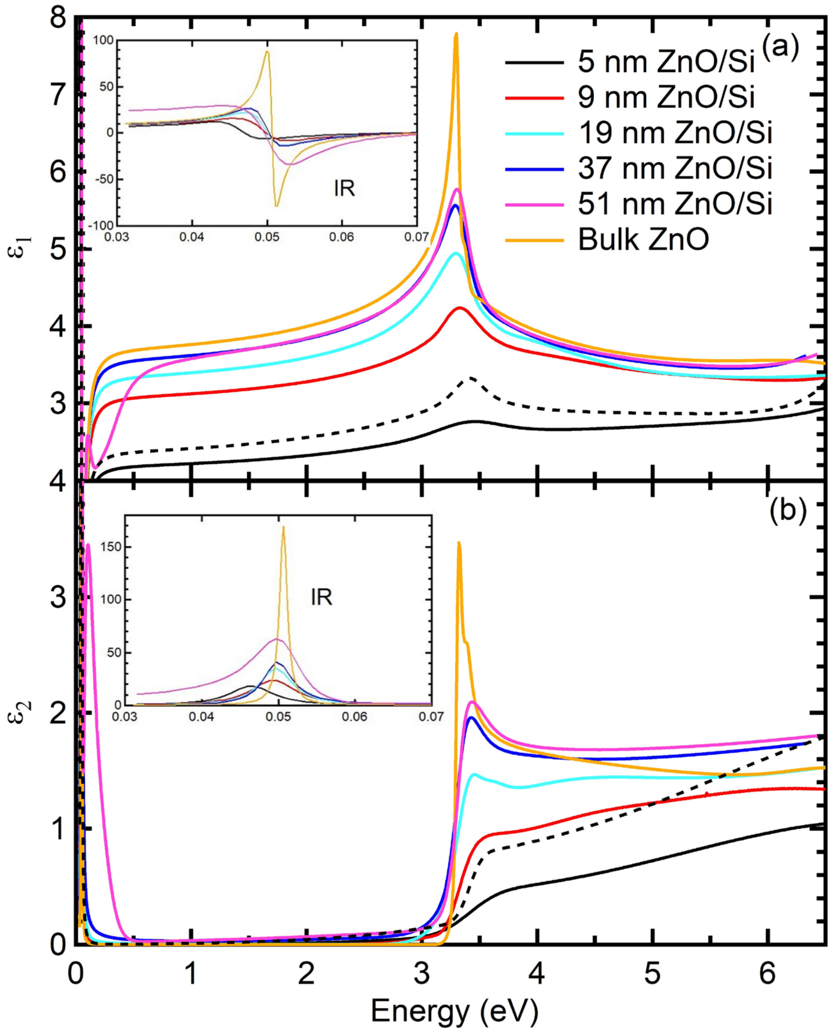


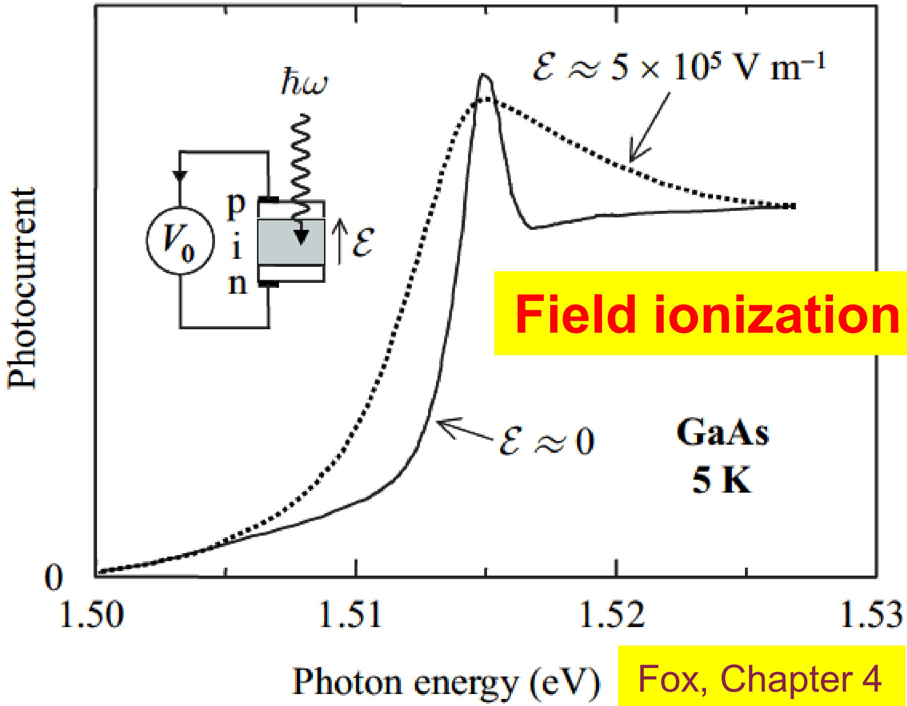
No red shift: phase angle changes. **Where is BGR?**

Carlos Armenta, February 2023, unpublished.
Emminger *el al.*, PSS RRL **2022**, 2200058.

Thickness dependence of excitonic absorption

ZnO on Si with different thicknesses.
This might be an electric field effect.
(Samarasingha, Sudeshna Chattopadhyay, SZ, JVSTB 2020)





Field ionization

Fox, Chapter 4

Conclusions

- Quantitative modeling of low-density optical processes is possible with basic physics and matrix elements from k.p theory:
  - **Photoluminescence in Ge**
  - **Indirect gap absorption in Ge**
  - **Direct gap absorption in Ge at low T**
  - More work is needed at high temperatures and for materials other than Ge.
- High carrier excitations:
  - **High electron doping density in Ge**
  - **Thermal excitation of electron-hole pairs in InSb and α-tin.**
  - **Femtosecond laser generation of electron-hole pairs in Ge (ELI Beamlines)**
  - Experimental data and qualitative explanations exist
- We need more experiments and more detailed theory and simulations.

Thank you!

Questions?

Many students contributed to this project.