ELLIPSOMETRY OF SEMICONDUCTORS UNDER THERMAL AND LASER EXCITATION

Ph.D. dissertation defense

Carola Emminger
Advisor: Dr. Stefan Zollner

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Department of Physics
New Mexico State University

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MOTIVATION

- **Group IV** (e.g. Ge, Si) and **group III-V** (e.g. GaAs, GaSb, InAs, InSb) semiconductors: important materials for optoelectronic devices
- Group IV photonics applications – examples:
  - Ge-on-Si photodetectors
  - Si-Ge-Sn and Ge-Sn alloys: mid-infrared detectors, photovoltaics, room temperature lasers
- Knowledge on **optical constants** (experiment and theory) is important for simulations and the development of optoelectronic devices.
- **Spectroscopic ellipsometry**: optical contact-free measurement technique; used to determine:
  - Optical constants (complex dielectric function, refractive index)
  - Thickness of a thin layer
  - Information on surface roughness, composition, strain, doping concentration, etc.
- **Femtosecond pump-probe ellipsometry**: study the effects of an ultrashort laser pulse, carrier concentration/doping, scattering etc.

OUTLINE

Introduction:

- Spectroscopic ellipsometry
- Critical point analysis using digital filtering

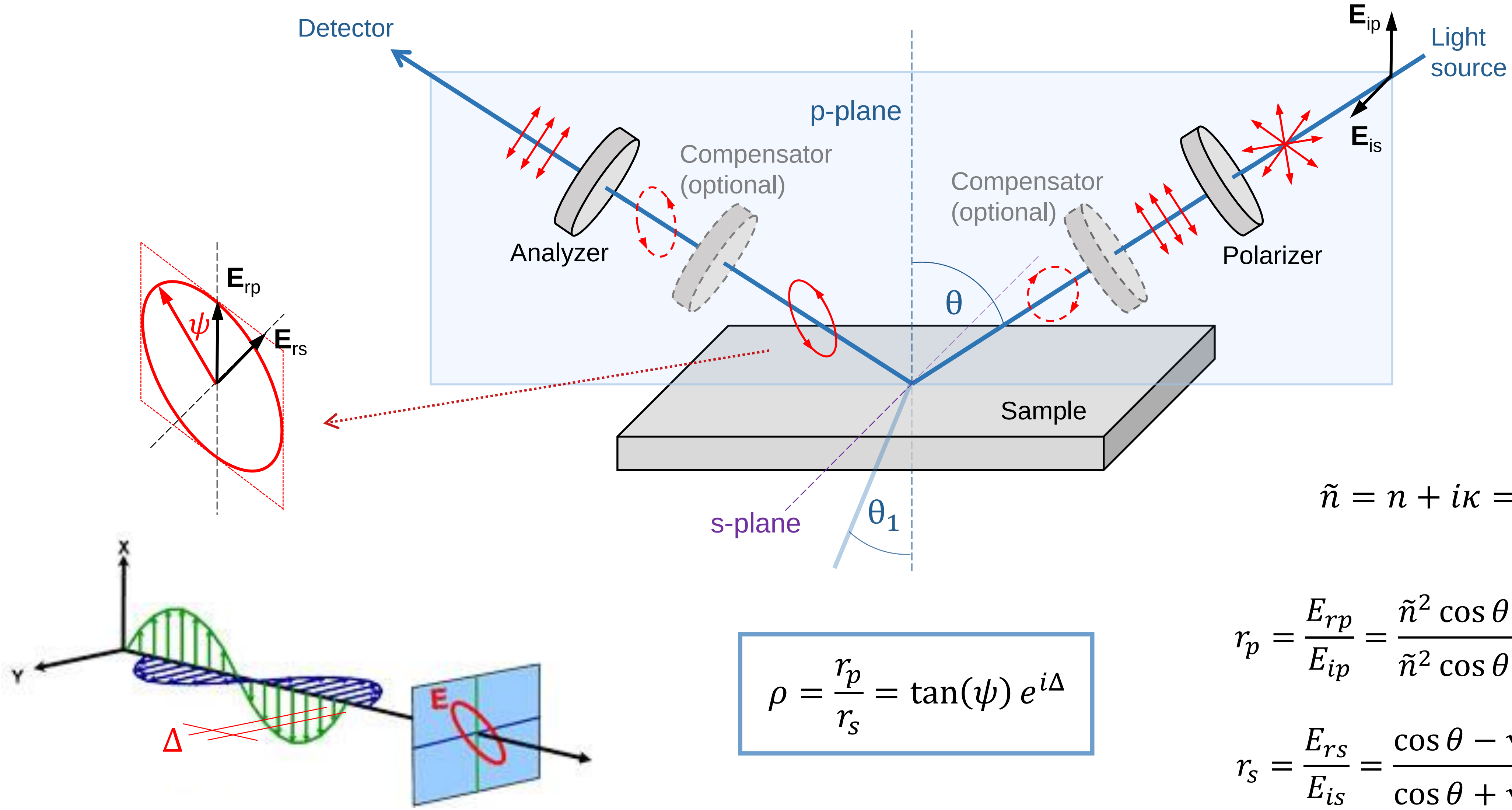
Part 1: Excitonic effects at the direct band gap of Ge

- Hulthén-Tanguy model with parameters from *k·p* theory
- Energy and broadening as functions of temperature

Part 2: Transient critical point parameters of Ge and Si from femtosecond pump-probe ellipsometry

- Critical point parameters as functions of time delay
- Coherent phonon oscillations

Spectroscopic Ellipsometry



$$\tilde{n} = n + i\kappa = \sqrt{\epsilon_1 + i\epsilon_2}$$

$$r_p = \frac{E_{rp}}{E_{ip}} = \frac{\tilde{n}^2 \cos\theta - \sqrt{\tilde{n}^2 - \sin^2\theta}}{\tilde{n}^2 \cos\theta + \sqrt{\tilde{n}^2 - \sin^2\theta}}$$

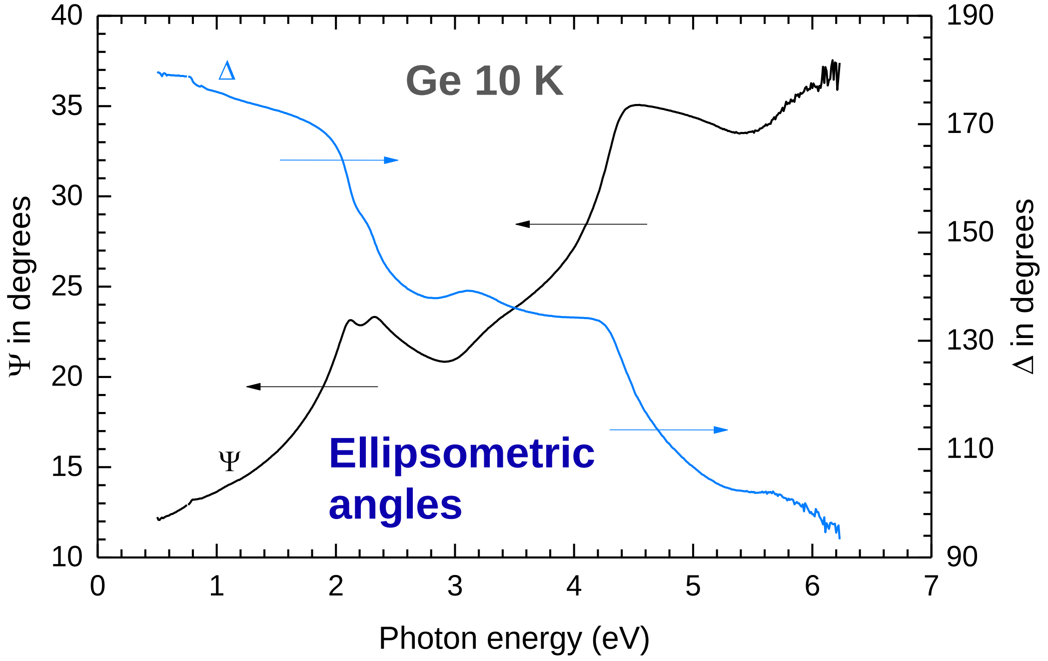
$$\rho = \frac{r_p}{r_s} = \tan(\psi)\, e^{i\Delta}$$

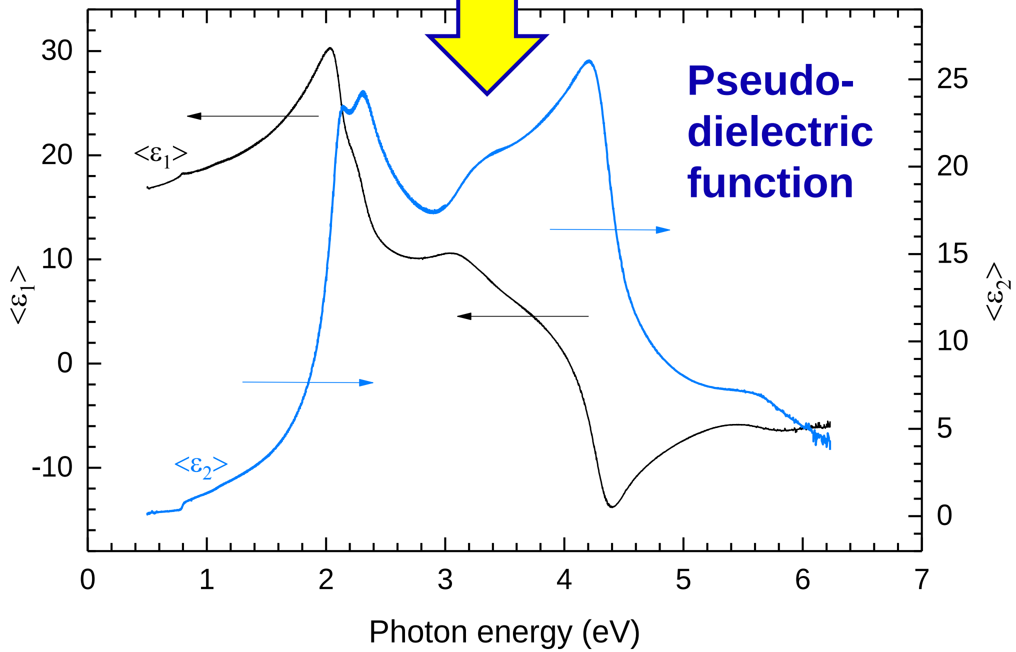
$$r_s = \frac{E_{rs}}{E_{is}} = \frac{\cos\theta - \sqrt{\tilde{n} - \sin^2\theta}}{\cos\theta + \sqrt{\tilde{n} - \sin^2\theta}}$$

https://www.jawoollam.com/resources/ellipsometry-tutorial/polarized-light

www.jawoollam.com

$$\rho = \tan(\psi)\, e^{i\Delta}$$



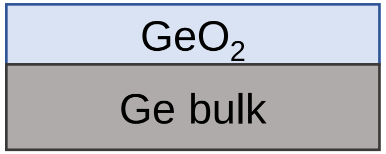


Dielectric function

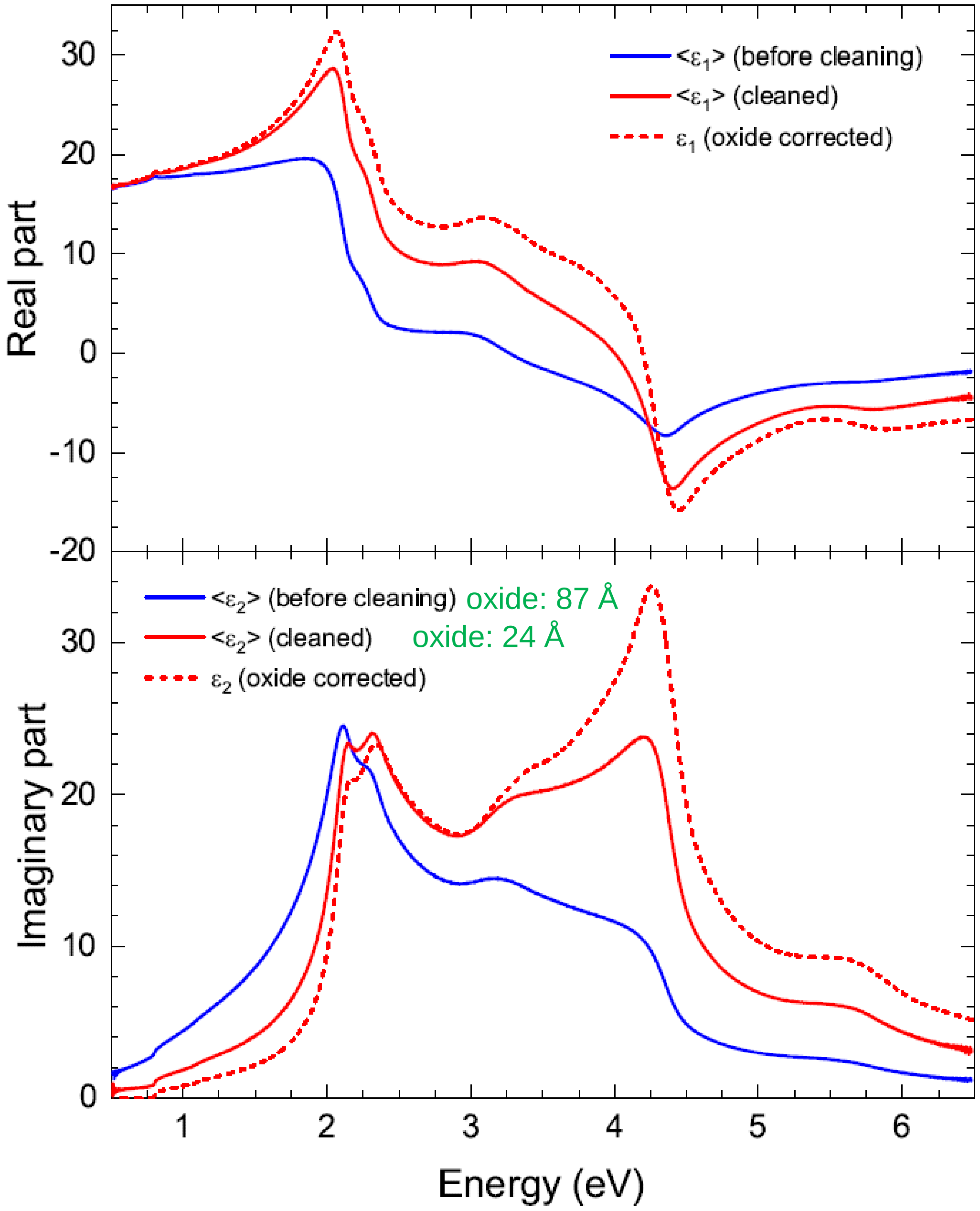
$\epsilon_1 + i\epsilon_2$

Native oxide layer correction

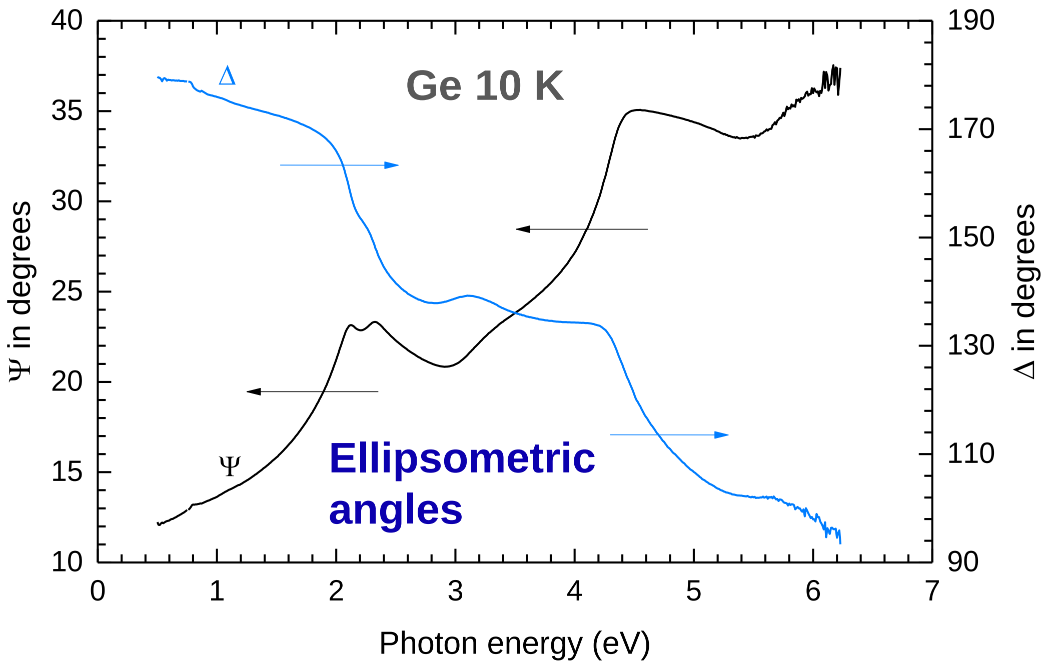
Two-layer model
(Ge substrate + native oxide)
Parametric semiconductor model
=> dielectric function ϵ

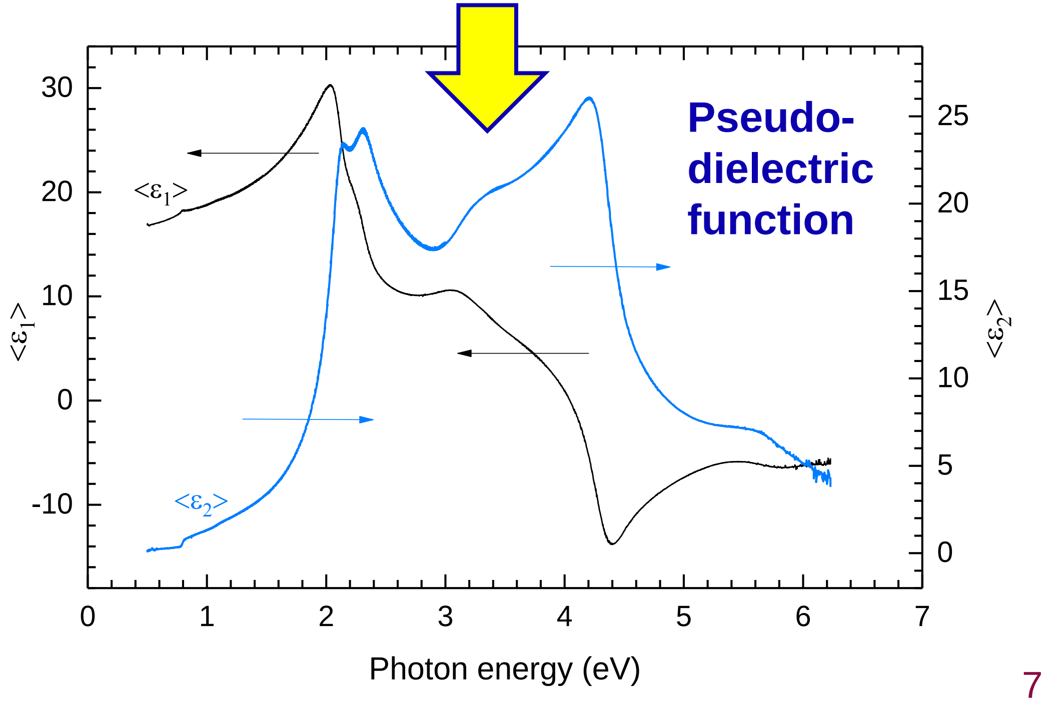


C. Emminger, F. Abadizaman, N.S. Samarasingha, T.E. Tiwald, S. Zollner, J. Vac. Sci. Technol. B **38**, 012202 (2020).



$$\rho = \tan(\psi)\, e^{i\Delta}$$



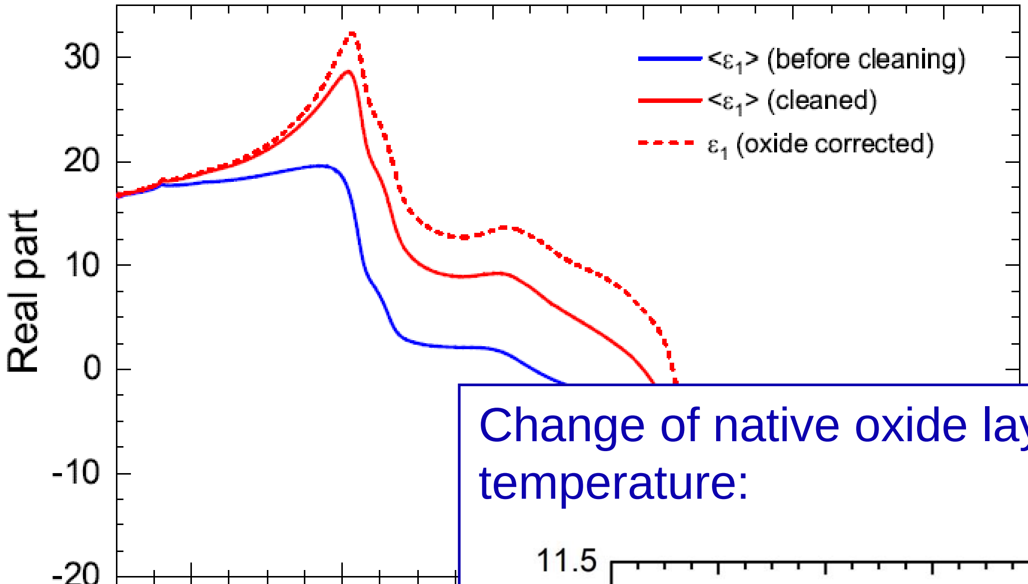


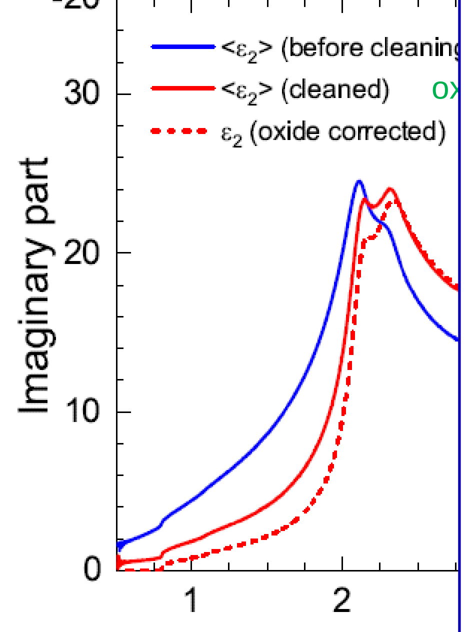
Native oxide layer correction

C. Emminger, F. Abadizaman, N.S. Samarasingha, T.E. Tiwald, S. Zollner, J. Vac. Sci. Technol. B **38**, 012202 (2020).

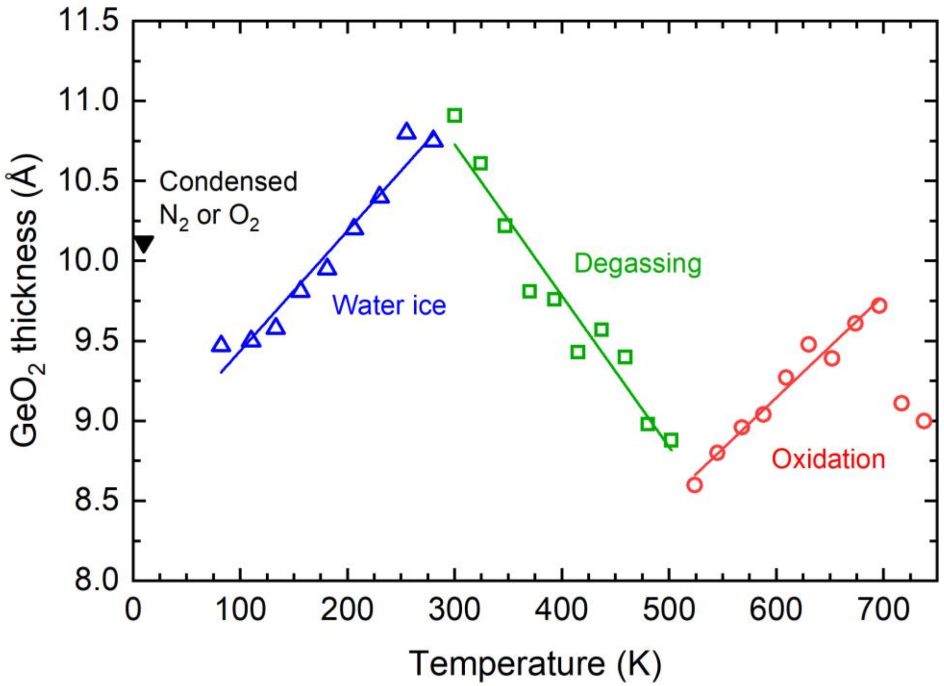
To stabilize native oxide layer:

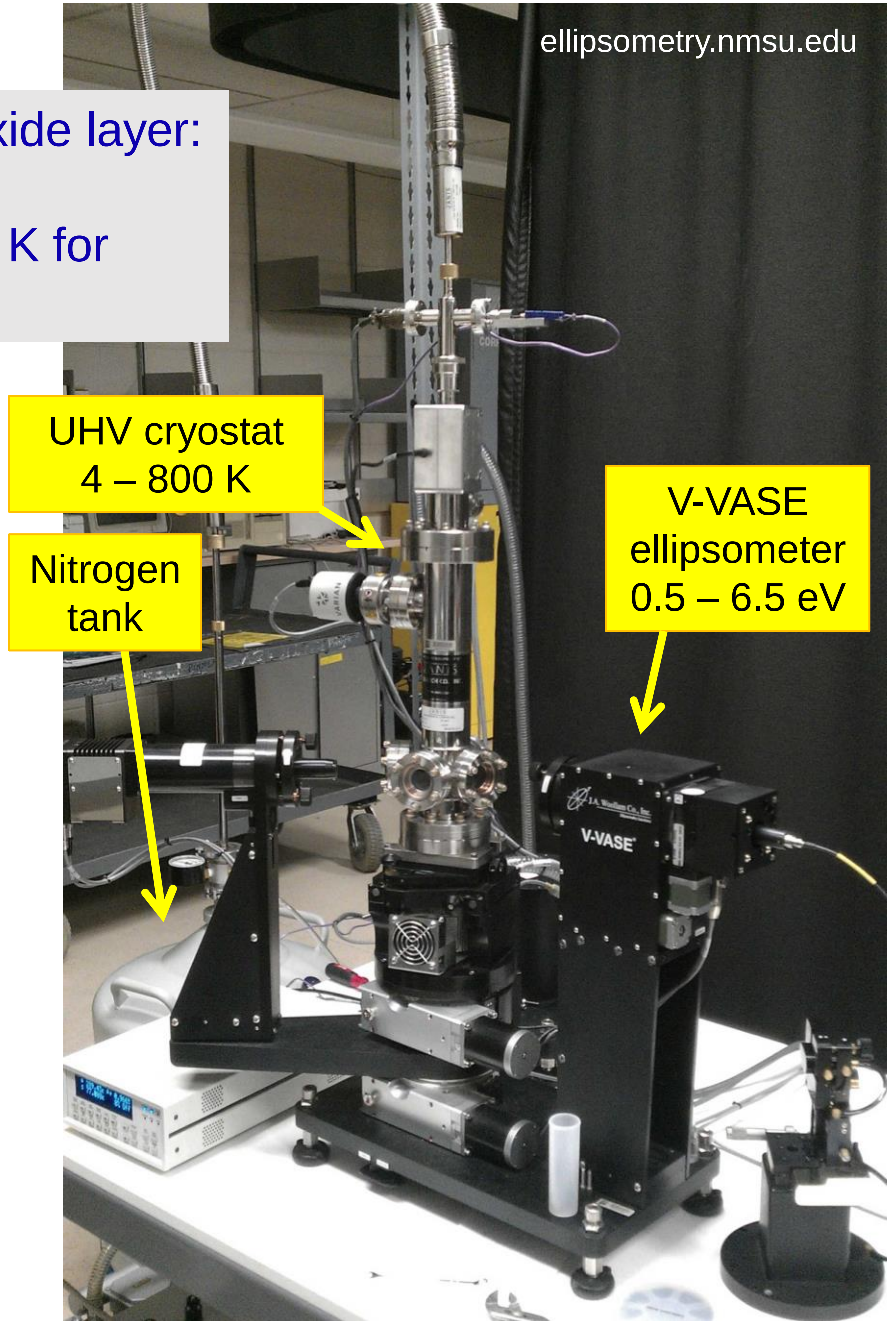
- UHV cryostat
- Heated up to 700 K for several hours





Change of native oxide layer thickness with temperature:



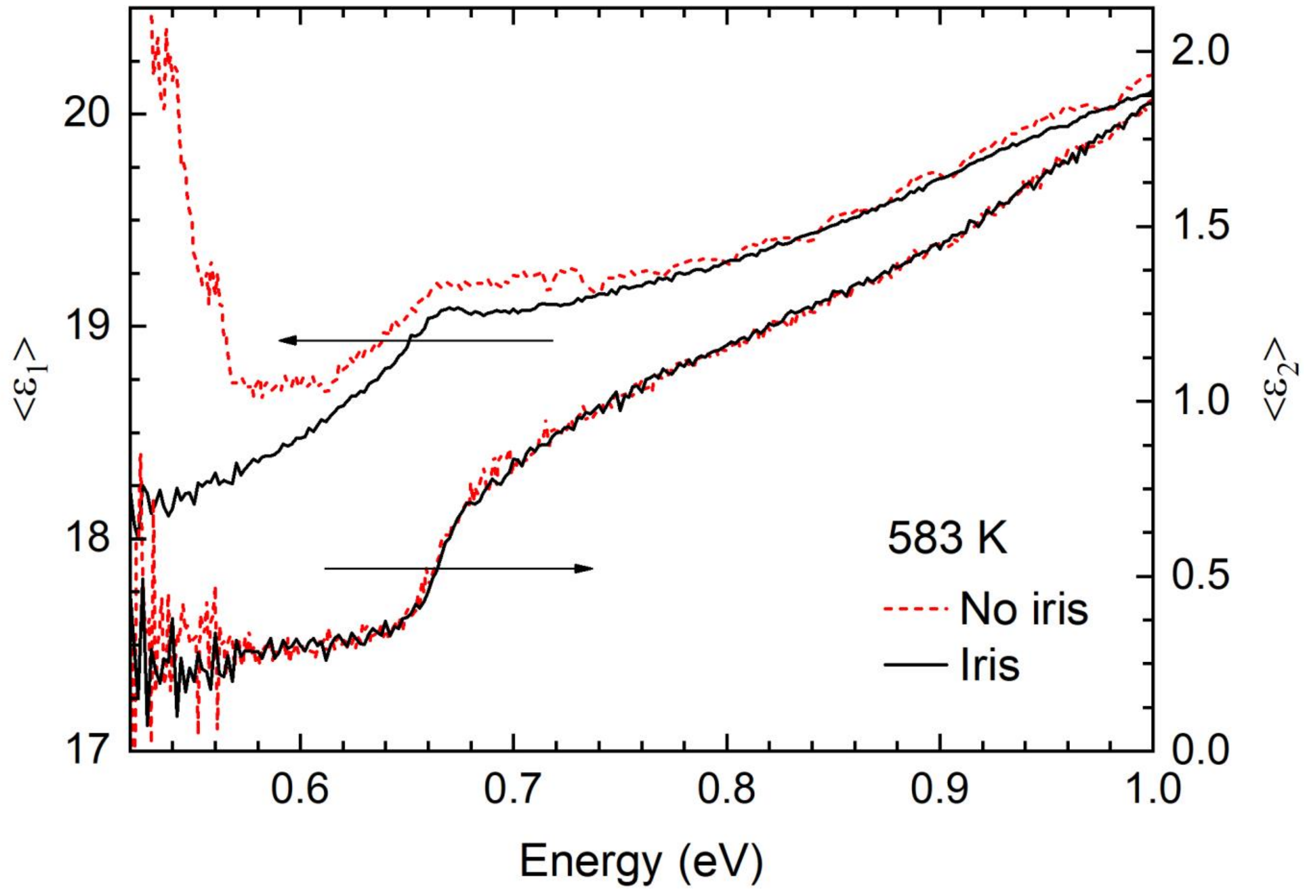


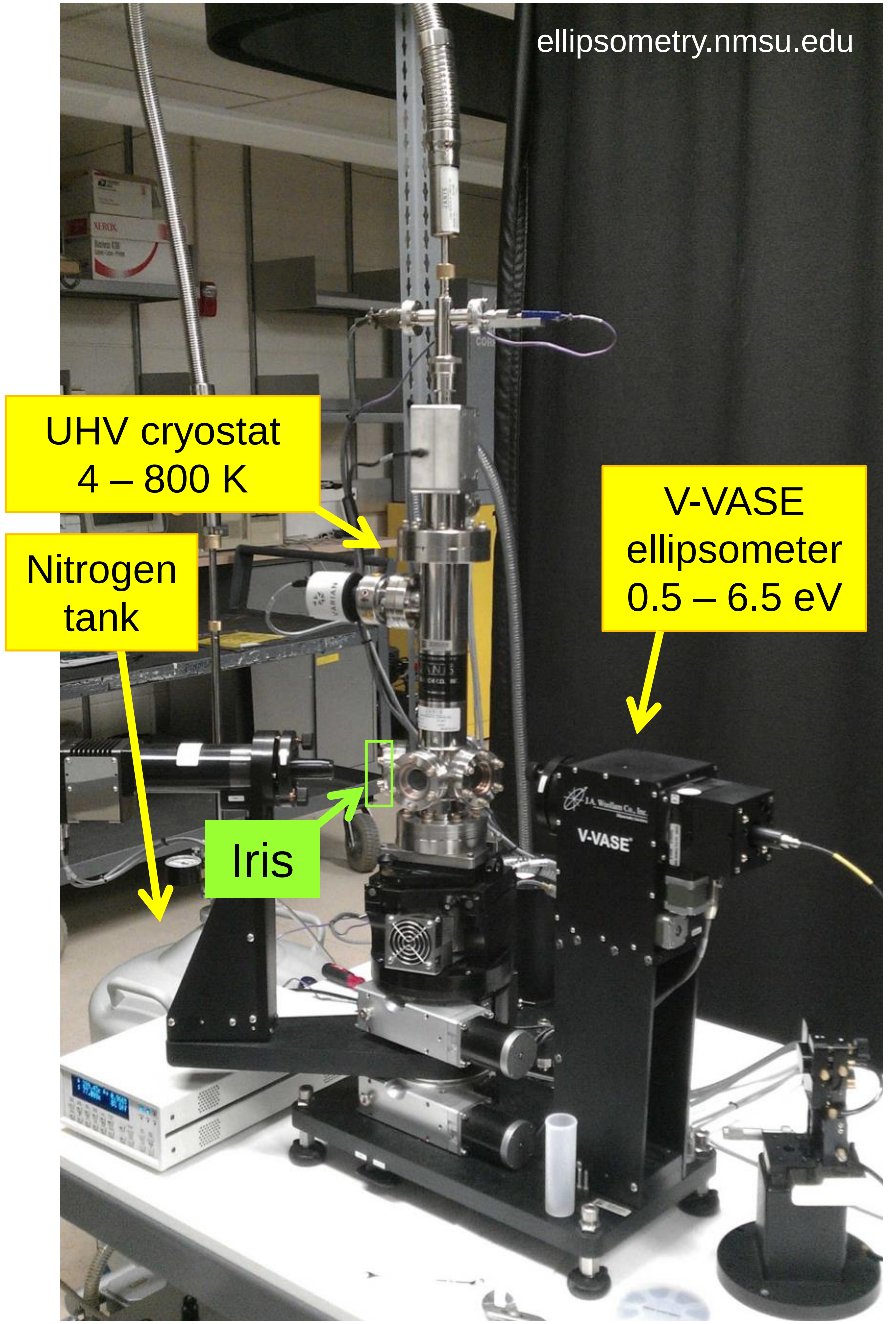
C. Emminger, F. Abadizaman, N.S. Samarasingha, T.E. Tiwald, S. Zollner, J. Vac. Sci. Technol. B **38**, 012202 (2020).

Black body radiation

At higher temperatures: Distortions due to black body radiation

⇒ Can be improved by placing an iris at the exit window of the cryostat

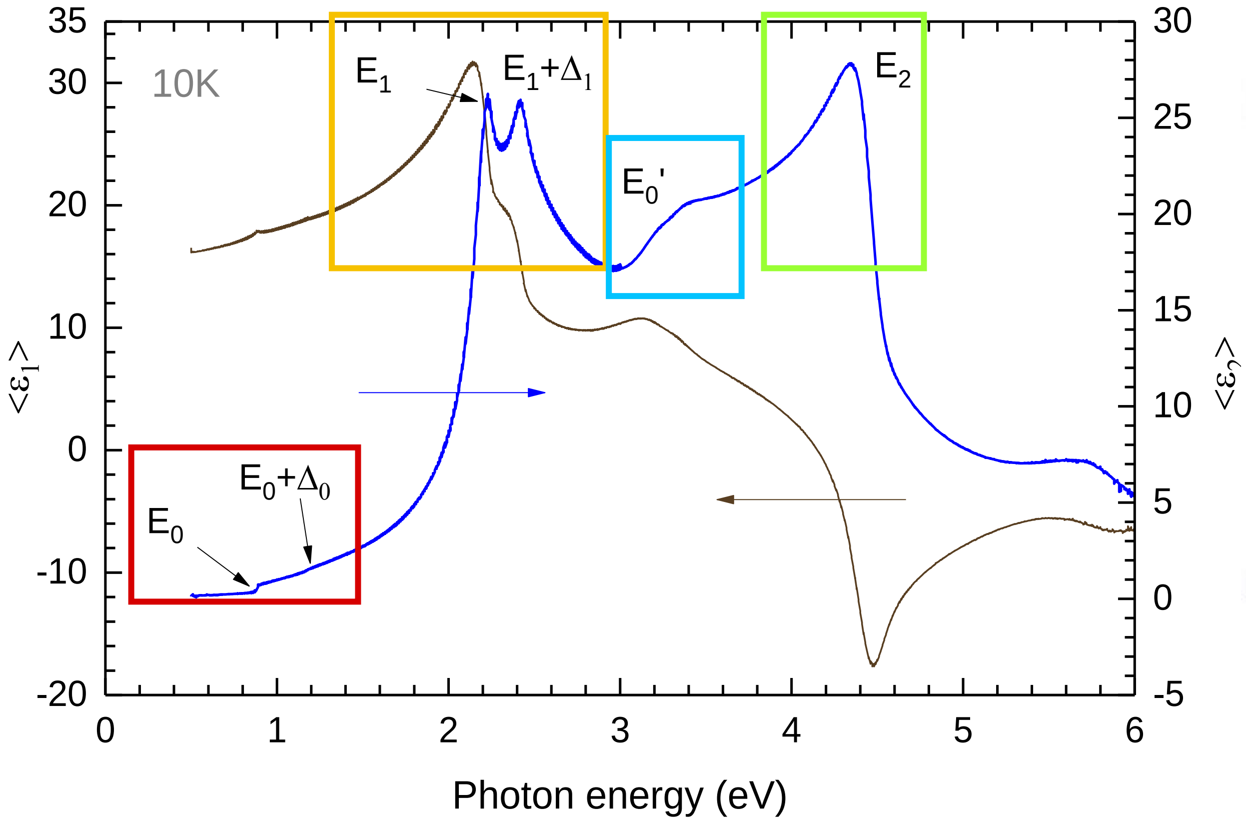


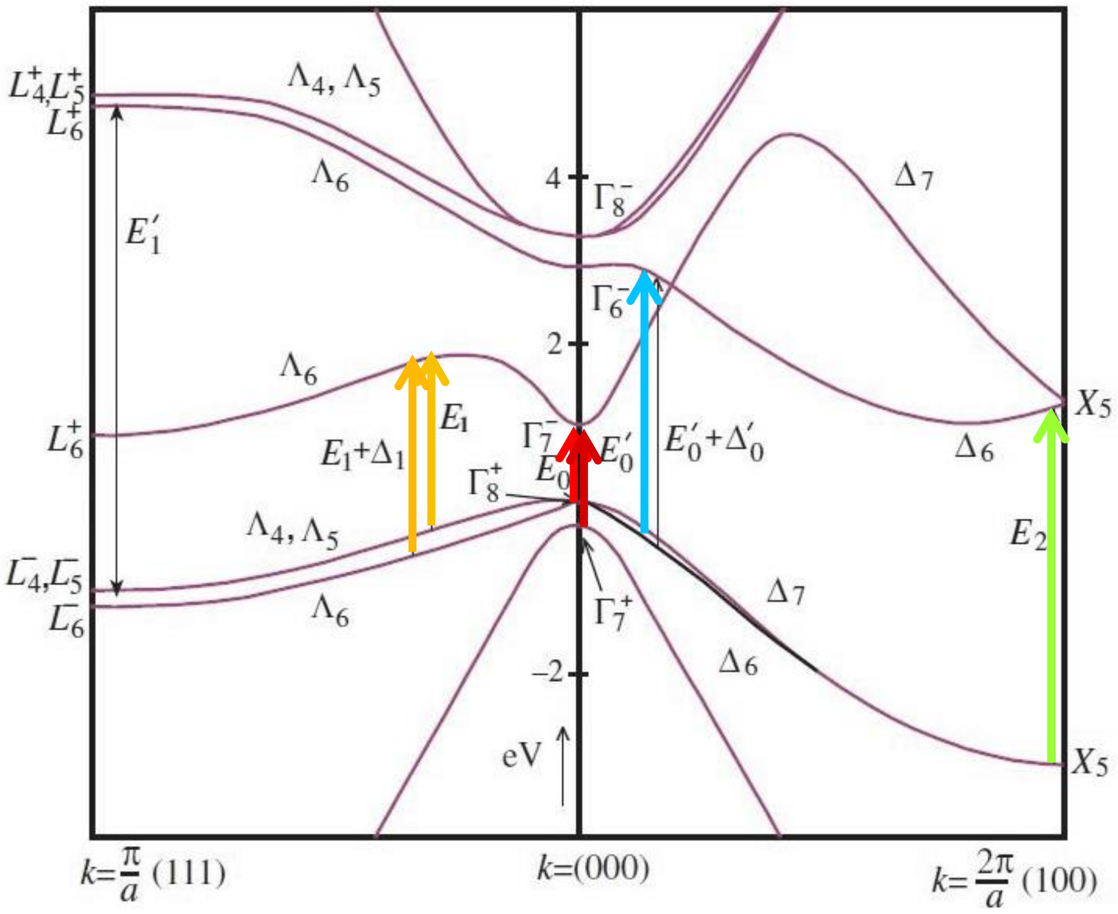


Critical points of Ge

- Structures in the dielectric function due to interband transitions
- Joint density of states – Van Hove singularities

$$D_j(E_{CV}) = \frac{1}{4\pi^3}\int \frac{dS_k}{|\nabla_k(E_{CV})|}$$





P. Y. Yu and M. Cardona: *Fundamentals of Semiconductors: Physics and Materials Properties*. (Springer-Verlag, Berlin, 2010)

- Critical point analysis: Second derivative of dielectric function to suppress non-resonant background

C. Emminger, F. Abadizaman, N.S. Samarasingha, T.E. Tiwald, S. Zollner, JVST-B **38**, 012202 (2020)

Critical point analysis using linear filters

- <u>Linear filter method (Le *et al.* 2019) can be used for:</u>
  - Scale change (energy ↔ wavelength)
  - Interpolation and filtering of the dielectric function
  - Calculation of second derivatives of the dielectric function

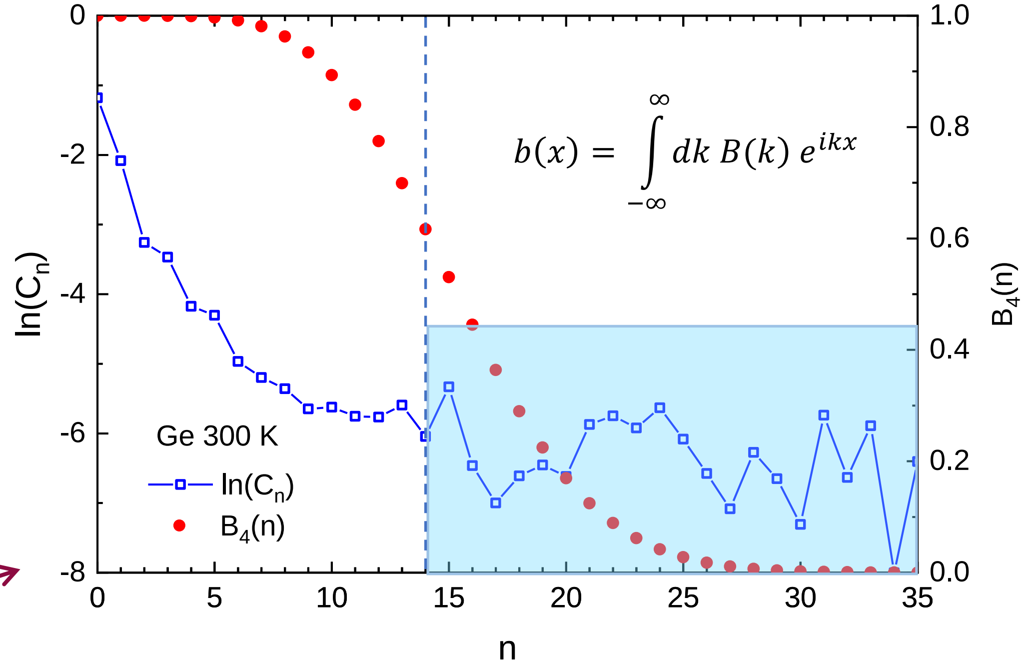
- Direct space convolution $\bar{f}(E) = \int_{-\infty}^{\infty} dE' f(E') b_M(E - E')$

  with Extended Gauss (EG) filters

$$b_M(x) = \left(1 - \frac{a}{1!}\frac{d}{da} + \frac{a^2}{2!}\frac{d^2}{da^2} - \cdots + (-1)^M \frac{a^M}{M!}\frac{d^M}{da^M}\right)\frac{a^{-1/2}}{2\sqrt{\pi}} e^{-x^2/4a}$$

$$a = 1/\Delta k^2 = \Delta E^2$$

Discrete Fourier transform
+ removal of endpoint discontinuities
(according to Yoo and Aspnes, 2001)



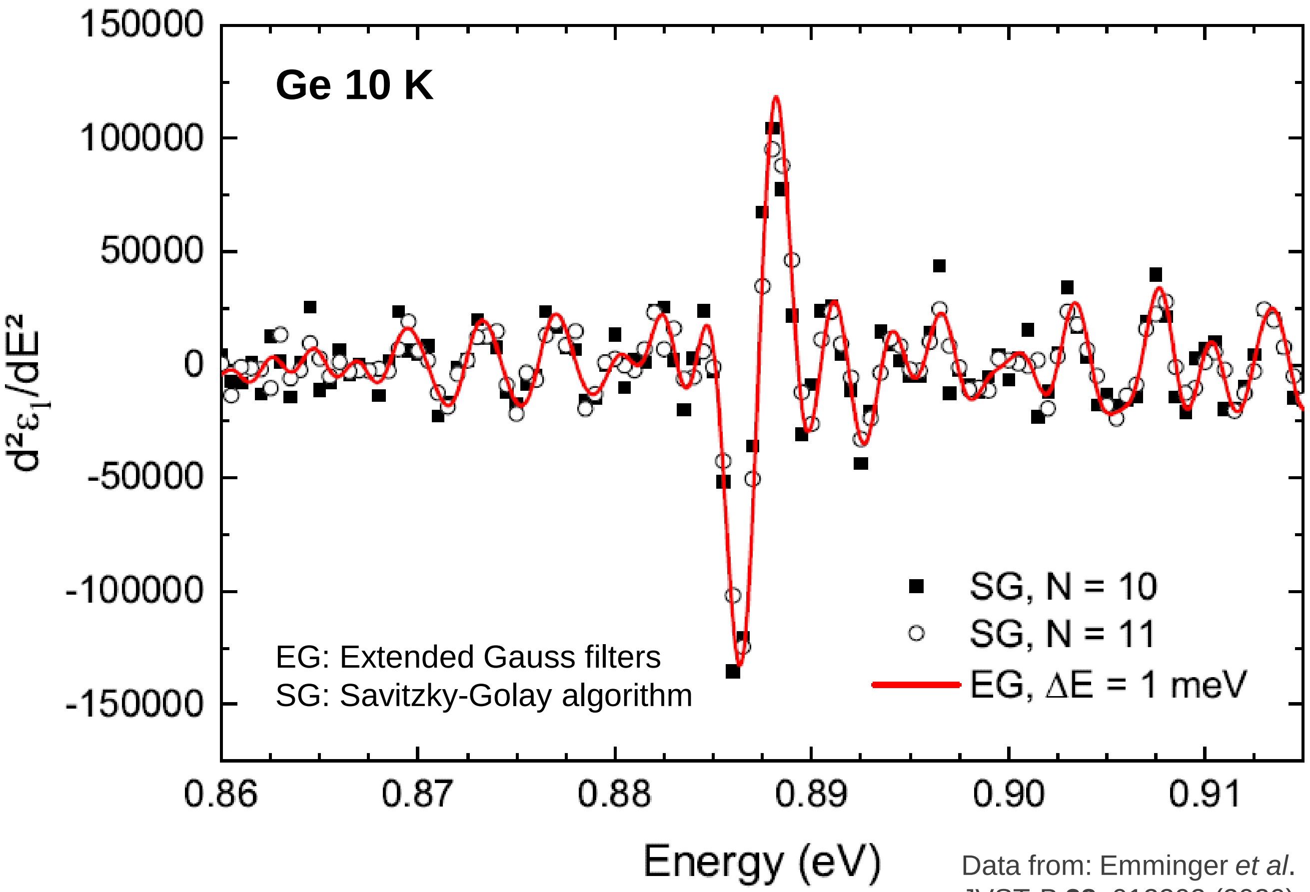
- Filter width ΔE chosen according to white noise onset

V. L. Le, T. J. Kim, Y. D. Kim, and D. E. Aspnes, J. Vac. Sci. Technol. B **37**, 052903 (2019)
S. D. Yoo and D. E. Aspnes, J. Appl. Phys. **89**, 8183 (2001)
Private communication with Prof. David E. Aspnes

Calculation of the second derivatives

Second derivative of ϵ using EG-filters for *M* = 4 for data sets with equidistant energy steps $\Delta E'$:

$$\frac{d^2\bar{\epsilon}(E)}{dE^2} \approx \frac{\Delta E'}{49152\sqrt{\pi}\Delta E^{13}} \sum_{j=-\infty}^{\infty} \epsilon(E_j)\Big(\big(E-E_j\big)^{10} - 106\big(E-E_j\big)^8 \Delta E^2 + 3608\big(E-E_j\big)^6 \Delta E^4 - 45936\big(E-E_j\big)^4 \Delta E^6 + 188496\big(E-E_j\big)^2 \Delta E^8 - 110880\,\Delta E^{10}\Big) e^{-\frac{(E-E_j)^2}{4\Delta E^2}}$$



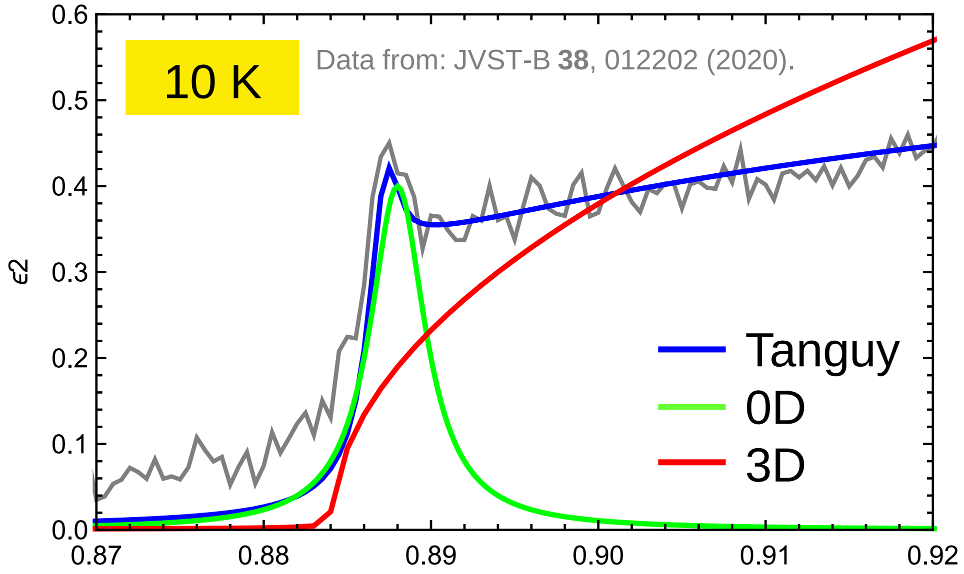
Data from: Emminger *et al*. JVST-B **38**, 012202 (2020).

V. L. Le, T. J. Kim, Y. D. Kim, D. E. Aspnes, J. Vac. Sci. Technol. B **37**, 052903 (2019)
A. Savitzky, and M. J. E. Golay, Anal. Chem. **36**, 1627 (1964).

Part 1: Excitonic effects at the direct band gap of Ge

Choice of lineshape for direct band gap

$$\epsilon(E) = B - Ae^{i\varphi}\left(E - E_g + i\Gamma\right)^{\mu}$$



Which lineshape describes the band gap best?

- Three dimensional lineshape: Square root-like dependence on energy

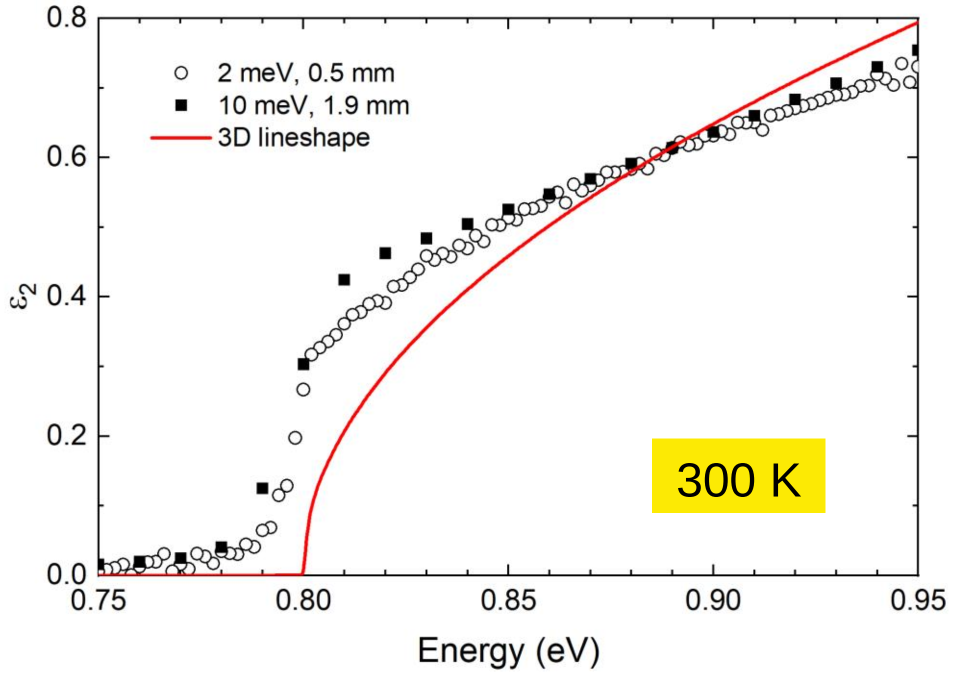
$$\epsilon_{3D}(E) = B + Ae^{i\varphi}\sqrt{E - E_g + i\Gamma}$$

- Lorentzian (0D) lineshape

$$\epsilon_{0D}(E) = B + \frac{Ae^{i\varphi}}{E - E_g + i\Gamma}$$

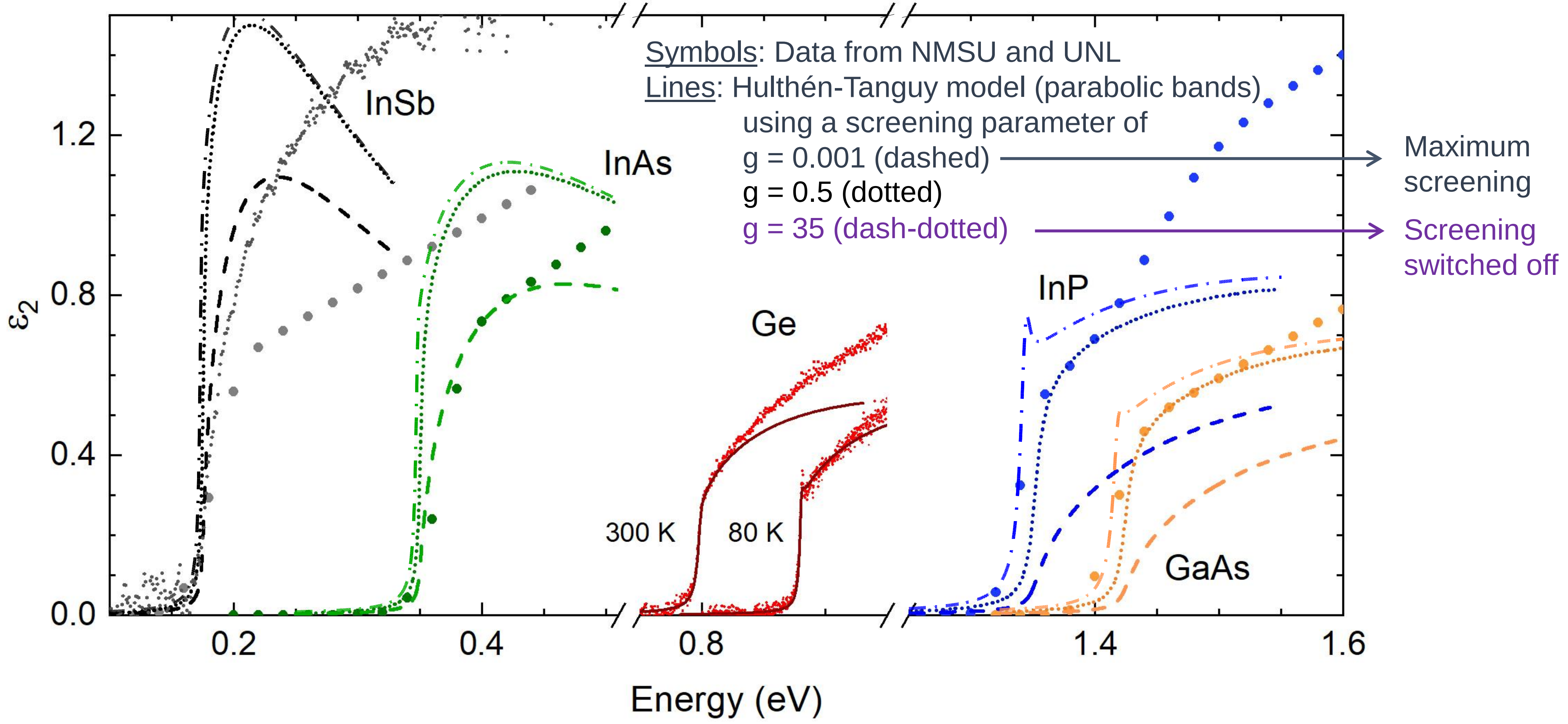
A …. amplitude
E_g … threshold energy
Γ …. broadening
φ …. phase angle

- Hulthén-Tanguy lineshape (blue lines): excitonic effects taken into account => **much better agreement**



D. E. Aspnes, *Handbook on Semiconductors*, edited by M. Balkanski (North-Holland, Amsterdam, 1980).
C. Tanguy, Phys. Rev. B **60**, 10660 (1999)

Motivation: Model of the direct band gap of various semiconductors



Model: C. Tanguy, Phys. Rev. B **60**, 10660 (1999)
Parameters: P. Lawaetz, Phys. Rev. B **4**, 3461 (1971).
L. Pavesi and F. Piazza, Phys. Rrev. B **44**, 9052 (1991).
S. Zollner J. Appl. Phys. **90**, 515 (2001).
S. Zollner, S. Gopalan, and M. Cardona, Solid State Commun. **77**, 485 (1991).

Next steps:
- Improve model (warping, non-parabolicity, screening)
- Detailed measurements → better data

Hulthén-Tanguy model to consider excitonic effects

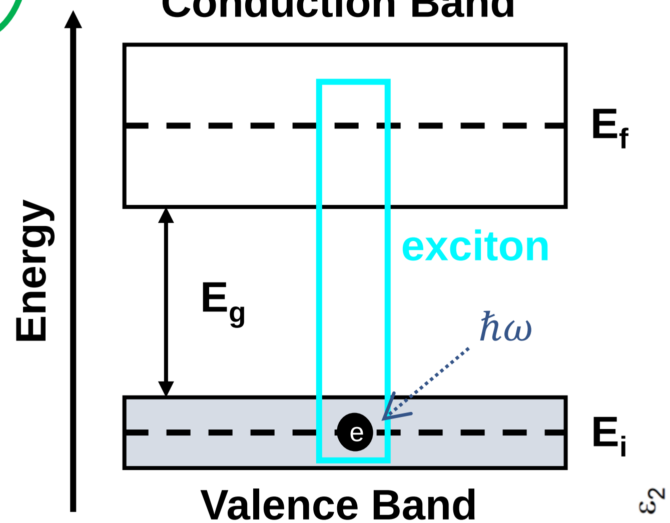
$$\epsilon(E) = \frac{A\sqrt{R}}{(E+i\gamma)^2}\left[\tilde{g}\big(\xi(E+i\Gamma)\big) + \tilde{g}\big(\xi(-E-i\Gamma)\big) - 2\tilde{g}(\xi(0))\right]$$

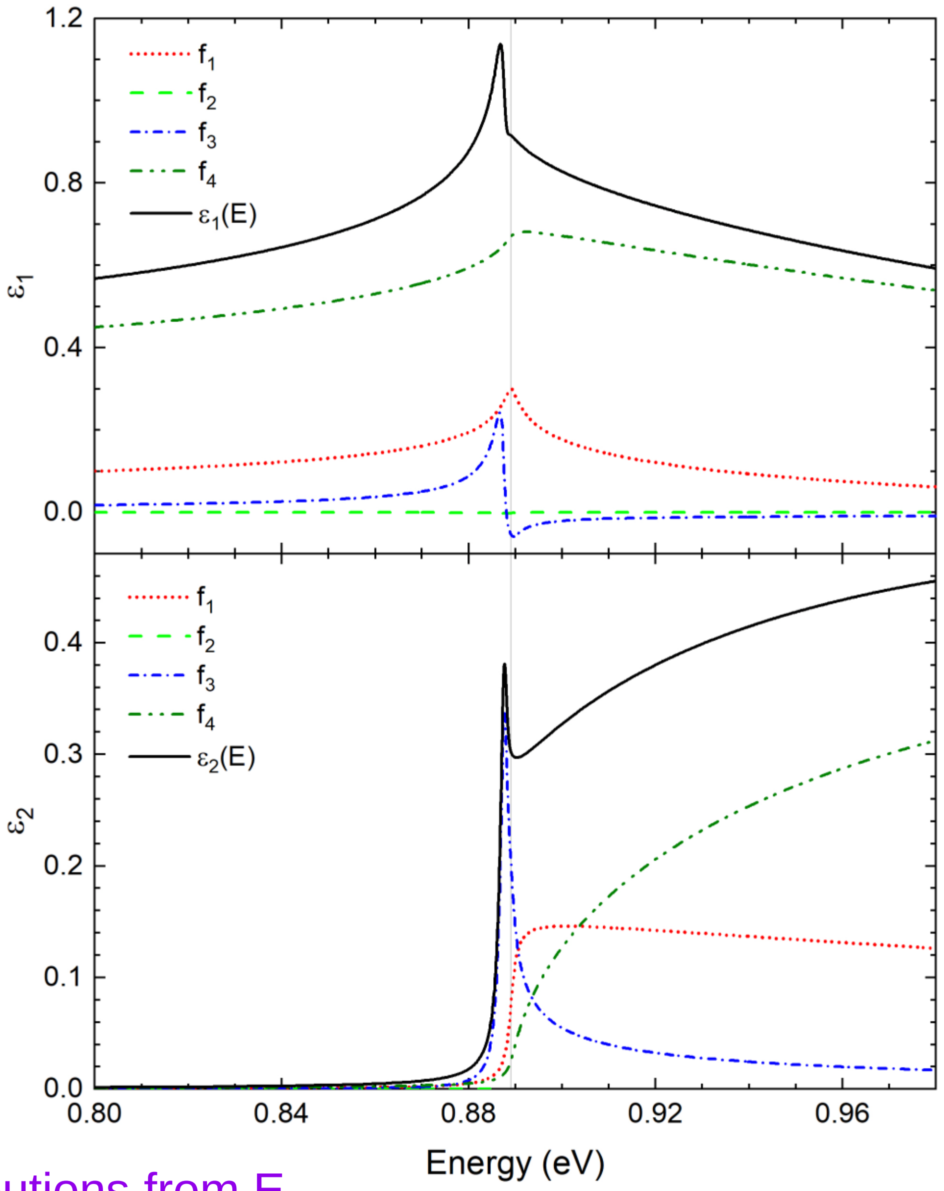
$$\tilde{g}(\xi) = -2\psi\left(\frac{g}{\xi}\right) - \frac{\xi}{g} - 2\psi(1-\xi) - \frac{1}{\xi}$$

unbound: $-2\psi\left(\frac{g}{\xi}\right)$; $\frac{\xi}{g}$ = 0 for $g \to \infty$; bound: $2\psi(1-\xi)$; interband: $\frac{1}{\xi}$

$$\xi(z) = \frac{2}{\left(\frac{E_0 - z}{R}\right)^{1/2} + \left(\frac{E_0 - z}{R} + \frac{4}{g}\right)^{1/2}}$$

$$\psi(x) = \frac{d\ln\Gamma(x)}{dx}$$ (Digamma function)





Heavy hole (hh) and light hole (lh):

$$\epsilon(E) = \epsilon_{hh}(E) + \epsilon_{lh}(E) + 1 + \frac{A_1}{1 - B_1 E^2}$$

Sellmeier term to consider contributions from E_1

C. Tanguy, Phys. Rev. B **60**, 10660 (1999)

Parameters from *k·p* theory

Fit parameters: E_0, γ, (A_1, B_1)

Calculated parameters: $A_{hh/lh}$ and $R_{hh/lh}$

- **Heavy (hh) and light hole (lh) contributions:** $\epsilon(E) = \epsilon_{hh}(E) + \epsilon_{lh}(E) + 1 + \frac{A_1}{1-B_1E^2}$
- **Amplitude:** $A_{hh/lh} = \frac{e^2\sqrt{m_0}}{\sqrt{2}\pi\epsilon_0\hbar}\mu_{hh/lh}^{3/2}\frac{E_P}{3}$ with $E_P = \frac{2P^2}{m_0} \approx 26$ eV
- **Excitonic binding energy**: $R_{hh/lh} = \frac{\mu_{hh/lh}}{\epsilon_{st}^2} 13.6$ eV ($R_{hh} \approx 2$ meV and $R_{lh} \approx 1$ meV at 4 K)
- **Reduced masses**: $\frac{1}{\mu_{hh/lh}} = \frac{1}{m_{hh/lh}} + \frac{1}{m_{e\Gamma}}$
- **Electron effective mass:** $\frac{m_0}{m_{e\Gamma}} = 1 + \frac{E_P}{3}\left[\frac{2}{E_0} + \frac{1}{E_0+\Delta_0}\right]$
- **Hole effective masses:** $\frac{m_0}{m_{hh/lh}} = \frac{1}{\hbar^2}\left[-A \pm \sqrt{B^2 + \frac{C^2}{5}}\right]$
- **DKK parameters:**

$$A = \frac{1}{3}[F + 2G + 2M] + 1$$
$$B = \frac{1}{3}[F + 2G - M]$$
$$C = \frac{1}{3}[(F - G + M)^2 - +(F + 2G - M)^2]$$

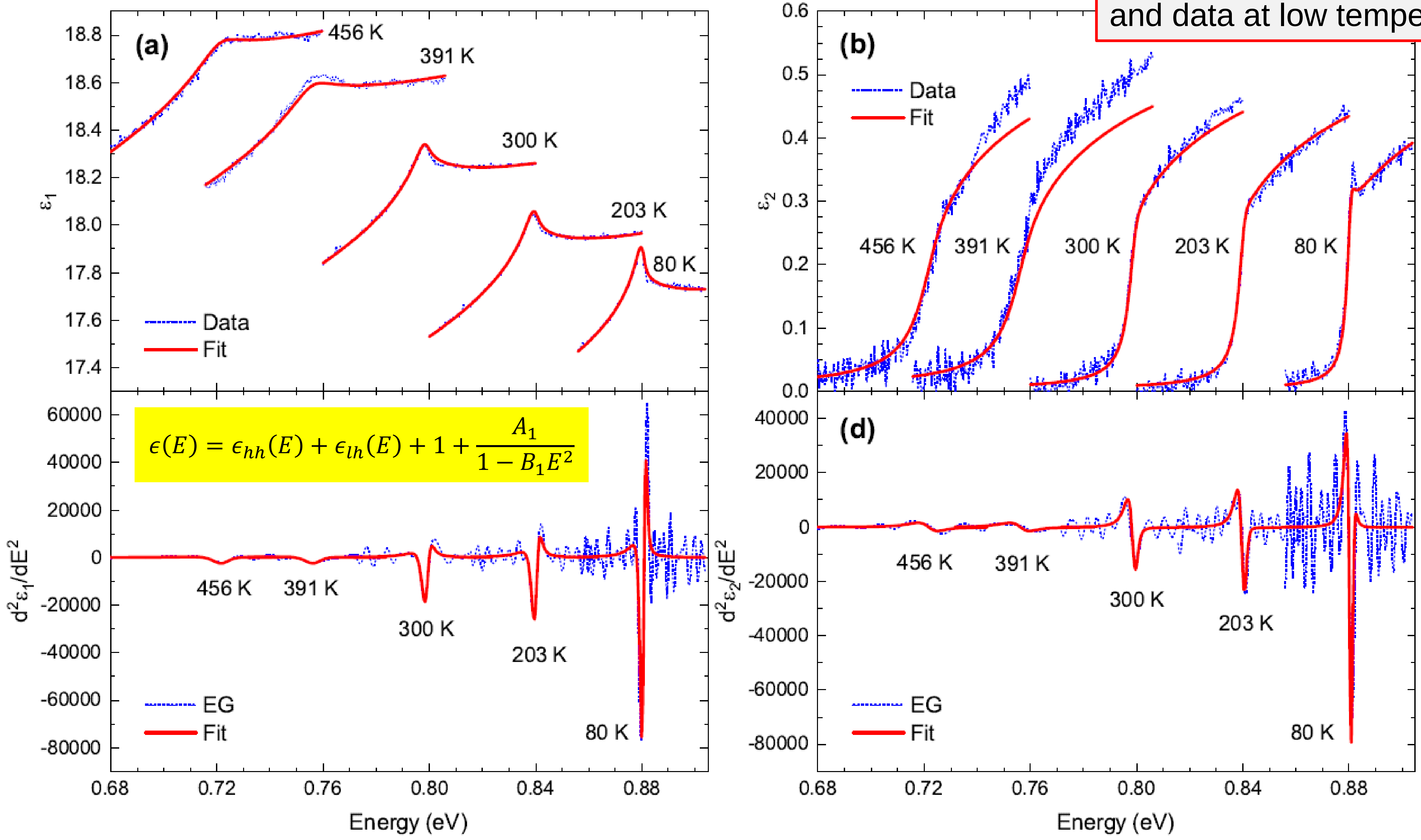
→

$$F(T) = -\frac{E_{P,4K}\left(\frac{a_{4K}}{a(T)}\right)}{E_0(T)}$$
$$M(T) = -\frac{E_{Q,4K}\left(\frac{a_{4K}}{a(T)}\right)}{E_0'(T)}$$
$$G(T) = -G_{4K}\left(\frac{a_{4K}}{a(T)}\right)$$

C. Tanguy, Phys. Rev. B **60**, 10660 (1999).
M. Cardona, J. Phys. Chem. Solids **24**, 1543 (1963).
C. Persson and U. Lindefelt, J. Appl. Phys. **82**, 5496 (1997).
G. Dresselhaus, A. F. Kip, and C. Kittel, Phys. Rev. **98**, 368 (1955).
J. Menéndez, D. J. Lockwood, J. C. Zwinkels, M. Noël, Phys. Rev. B **98**, 165207 (2018).
P. Yu and M. Cardona, *Fundamentals of Semiconductors*, (Springer, Heidelberg, 2010).

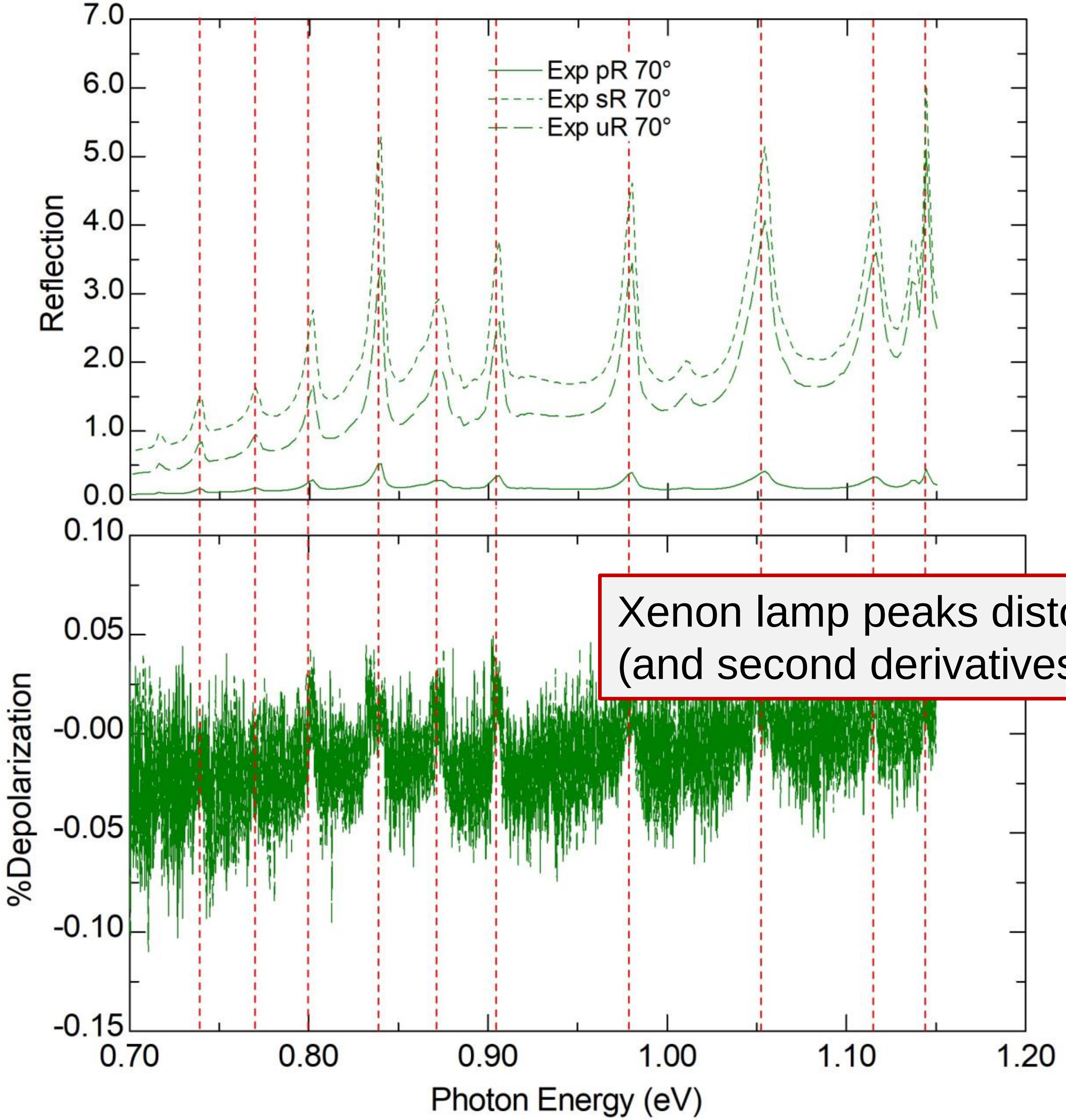
Fit results for Ge

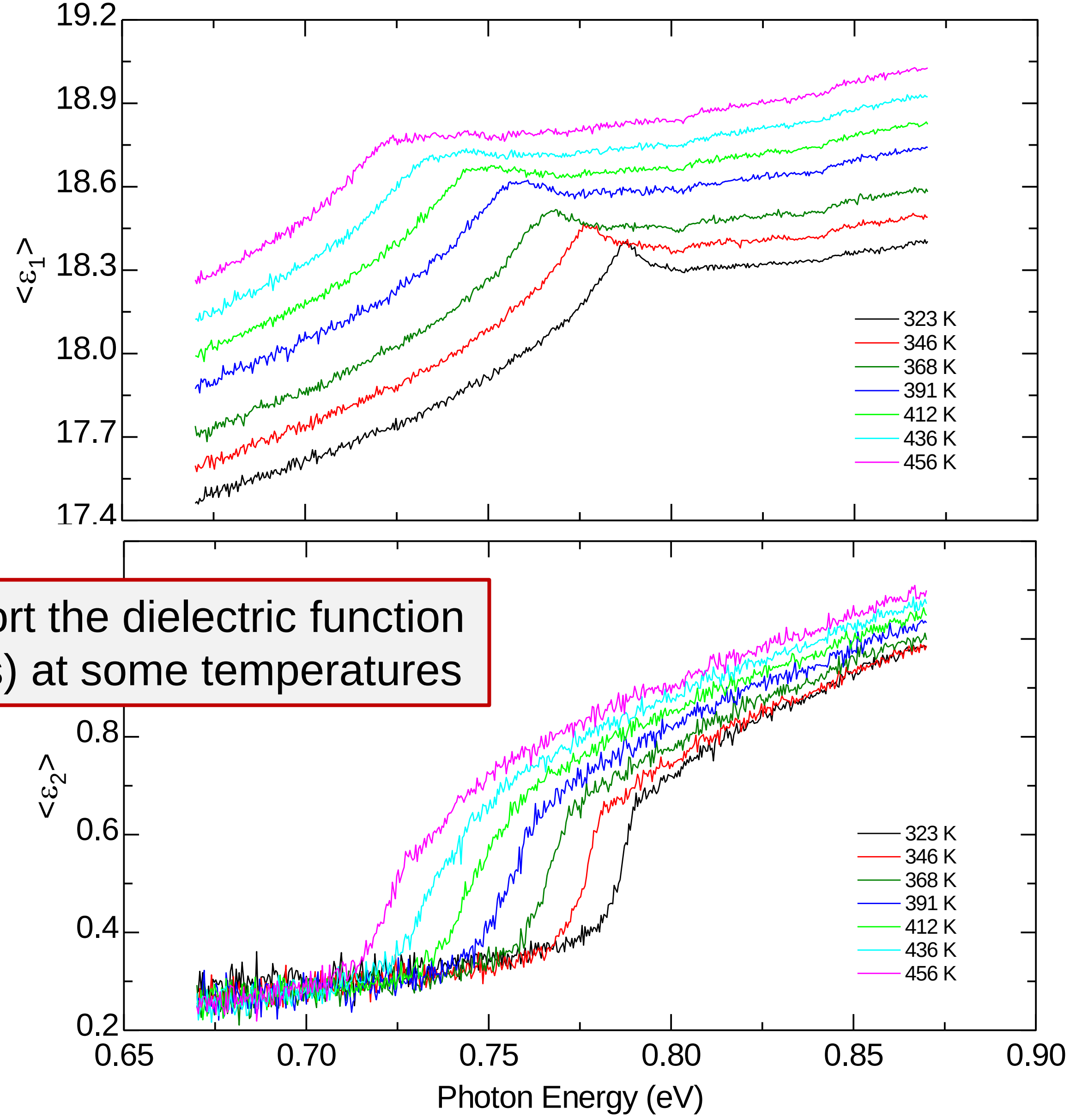
Good agreement between fit and data at low temperatures



$$\epsilon(E) = \epsilon_{hh}(E) + \epsilon_{lh}(E) + 1 + \frac{A_1}{1 - B_1 E^2}$$

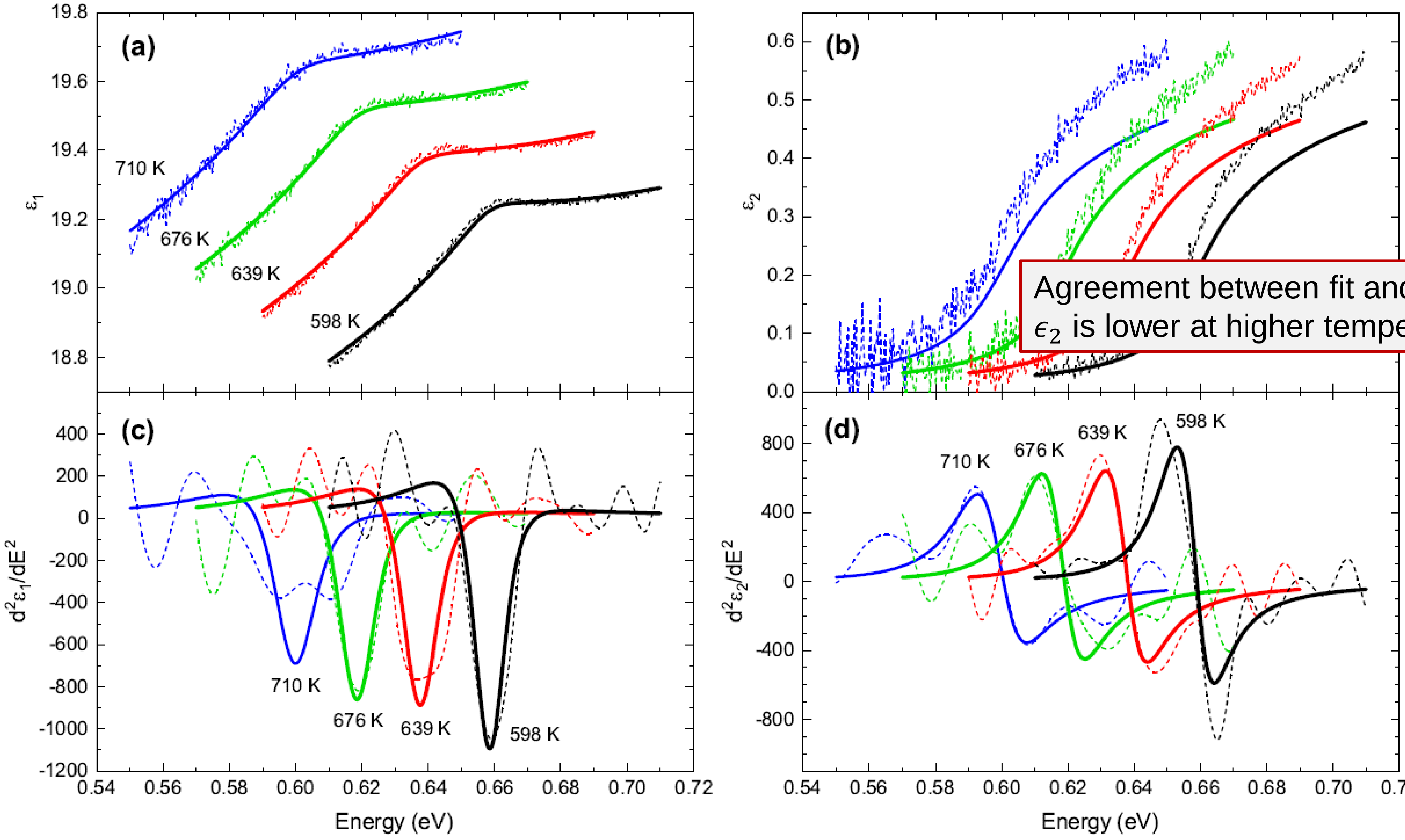
Distortions due to xenon lamp





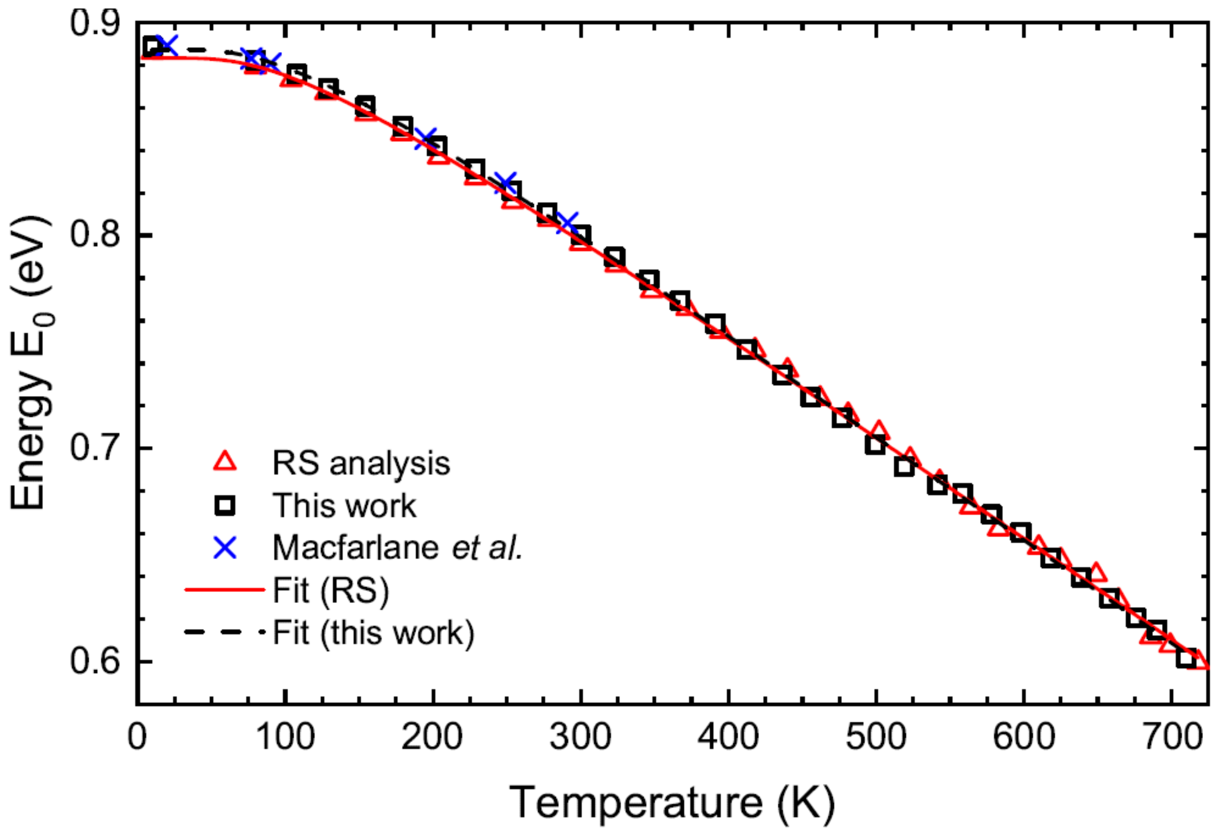
Xenon lamp peaks distort the dielectric function (and second derivatives) at some temperatures

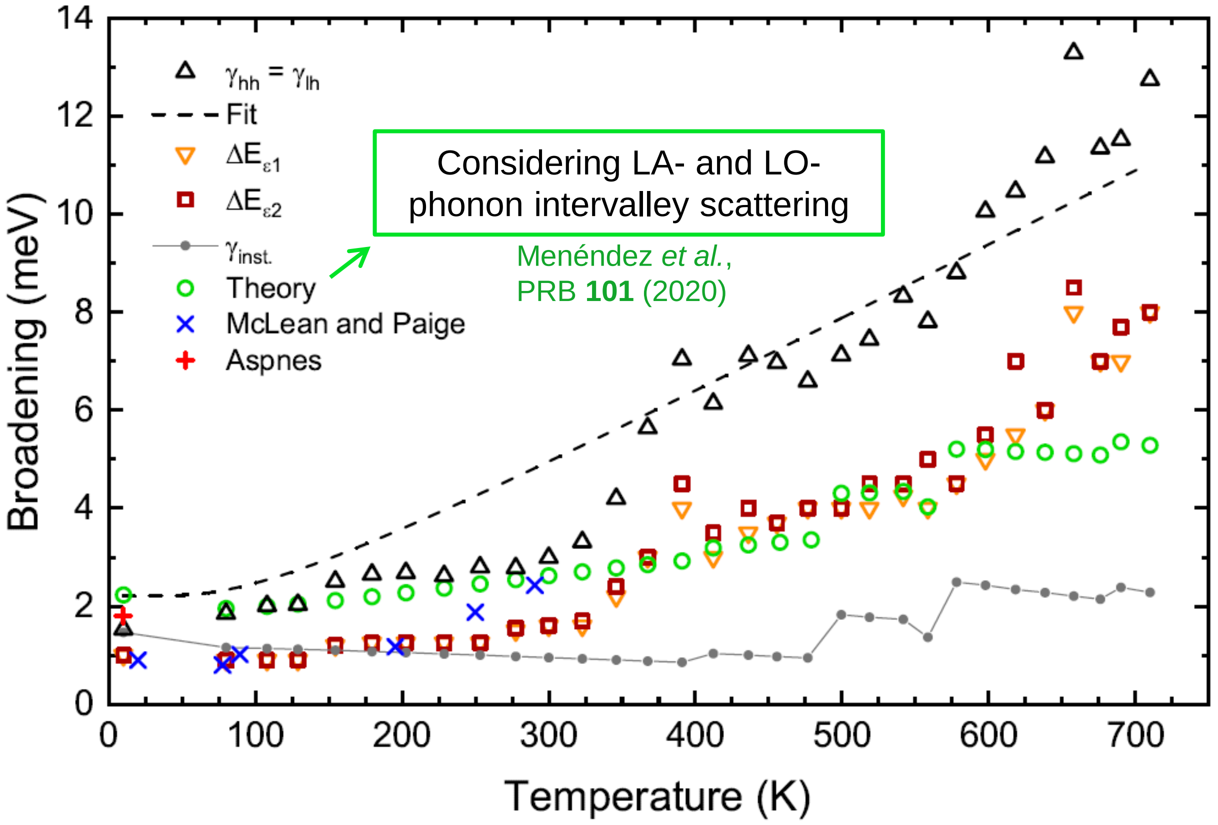
Fit results at high temperatures



Agreement between fit and data for ϵ_2 is lower at higher temperatures

Temperature dependence of the energy and broadening of E_0





$$E(T) = E_a - E_b\left[\frac{2}{e^{\frac{E_{ph}}{kT}} - 1} + 1\right]$$

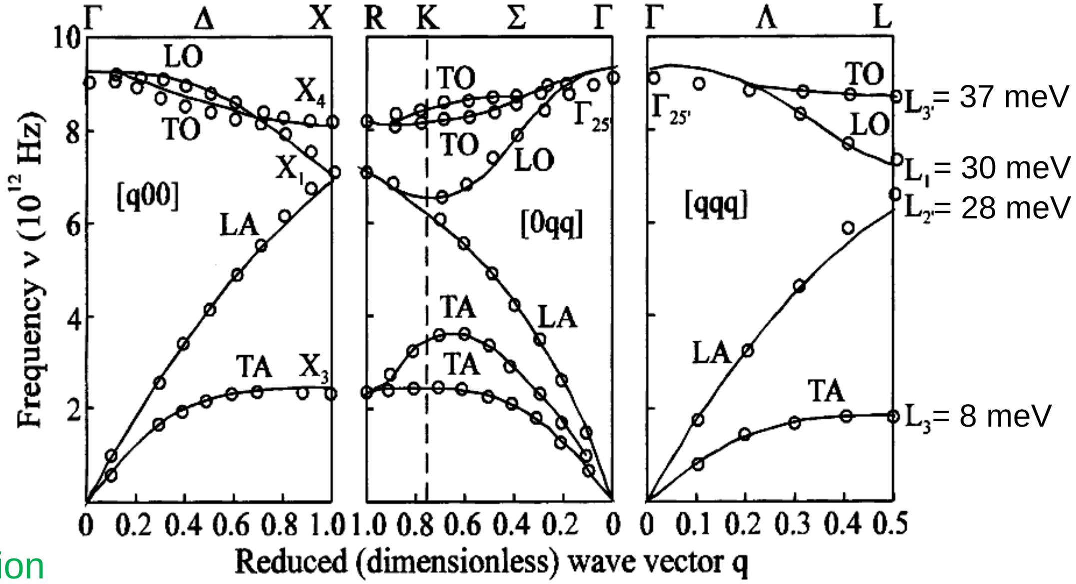
$$\Rightarrow E_{\mathrm{ph}} = 25 \pm 1\ \mathrm{meV}$$

$$\gamma(T) = \gamma_{\mathrm{a}} + \gamma_{\mathrm{b}}\left[\frac{2}{e^{\frac{E_{\mathrm{ph}}}{kT}} - 1} + 1\right]$$

L. Viña, S. Logothetidis, M. Cardona, Phys. Rev. B **30**, 1979 (1984).
C. Emminger, F. Abadizaman, N.S. Samarasingha, T.E. Tiwald, S. Zollner, J. Vac. Sci. Technol. B **38**, 012202 (2020).
G. G. Macfarlane, T. P. McLean, J. E. Quarrington, and V. Roberts, Proc. Phys. Soc. 71, 863 (1958).
T. P. McLean and E. G. S. Paige, J. Phys. Chem. Solids **23**, 822 (1962).

Intravalley and intervalley scattering rates

Weber W., Phys. Rev. **B15,** 10 (1977) 4789-4803.



Electron-LA phonon intervalley (Conwell 1967):

$$\tau_{\Gamma L}^{-1} = N_V \frac{D_{\Gamma L}^2 m_{\text{eff}}^{\frac{3}{2}}}{\sqrt{2}\pi\hbar^2 \rho E_{\text{ph}}} \left[N_{\text{ph}} \sqrt{\Delta E + E_{\text{ph}}} + (1 + 2N_{\text{ph}}) \sqrt{\Delta E - E_{\text{ph}}} \right]$$

$D_{\Gamma L} = 3\text{ eV} - 6.5\text{ eV}$ … T-dep. DP (Zollner 1990)

$m_{\text{eff}} = (m_l m_t^2)^{1/3} = (1.6 \cdot 0.08^2)^{1/3}\ m_0 = 0.22\ m_0$

$N_V = 4$

N_{ph} phonon occupation factor => absorption + emission

Electron-LA phonon intravalley scattering:

$$\tau_{\text{ac}}^{-1} = \frac{\sqrt{2} E_1^2 m_{e\Gamma}^{3/2} k_B T}{\pi \hbar^4 \rho v_s^2} \sqrt{E_k}$$

→ 0 if $E_k = 0$ (we use: $E_k = 10\text{ meV}$)

$E_1 = 11.4\text{ eV}$ … deformation potential (DP)

$v_s = 5.4 \times 10^3\text{ m/s}$ … sound velocity

$\gamma = \hbar/(2\tau)$

	τ_{ac} (fs)	γ_{ac} (meV)	$\tau_{\text{op,hh}}$ (fs)	$\gamma_{\text{op,hh}}$ (meV)	$\tau_{\text{op,lh}}$ (fs)	$\gamma_{\text{op,lh}}$ (meV)	$\tau_{\Gamma L}$ (fs)	$\gamma_{\Gamma L}$ (meV)
10 K	$4\text{x}10^5$	$8\text{x}10^{-4}$	$3\text{x}10^{21}$	$1\text{x}10^{-19}$	$1\text{x}10^{23}$	$3\text{x}10^{-21}$	$1\text{x}10^3$	0.3
80 K	$5\text{x}10^4$	$6\text{x}10^{-3}$	$2\text{x}10^5$	$2\text{x}10^{-3}$	$5\text{x}10^6$	$7\text{x}10^{-5}$	600	0.6
300 K	$1\text{x}10^4$	0.02	$3\text{x}10^3$	0.1	$8\text{x}10^4$	$4\text{x}10^{-3}$	100	3
710 K	$6\text{x}10^3$	0.06	700	0.4	$3\text{x}10^4$	0.01	60	6

Hole-optical phonon intravalley scattering:

$$\tau_{\text{op},h}^{-1} = \frac{D_0^2 m_h^{3/2}}{\sqrt{2}\pi\hbar^2 \rho \sqrt{E_{\text{ph}}}} N_{\text{ph}}$$

only absorption for $E_k = 0$

$d_0 = 37\text{ eV}$ … DP (Pötz and Vogl 1981)

$D_0 = d_0/a = 7.53\text{ eV/Å}$

D. K. Ferry, *Semiconductor Transport*, (Tayler & Francis, New York, 2000).
E. M. Conwell, *High Field Transport in Semiconductors* (Academic, New York, 1967).
W. Pötz and P. Vogl, Phys. Rev. B **24**, 2025 (1981).
S. Zollner, S. Gopalan, and M. Cardona, Sol. State Comm. **76**, 877 (1990).

Excitonic binding energy

Excitonic binding (Rydberg) energy: $R_{hh/lh} = \frac{\mu_{hh/lh}}{\epsilon_{\text{st}}^2} 13.6\text{ eV}$

Temperature-dependent due to *T* dependence of ϵ_{st} and effective masses

Reduced mass: $\frac{1}{\mu_{hh/lh}} = \frac{1}{m_{hh/lh}} + \frac{1}{m_{e\Gamma}}$

T dependence of the electron effective mass through *T*-dependent band gap E_0

$$\frac{m_0}{m_{e\Gamma}} = 1 + \frac{E_P}{3}\left[\frac{2}{E_0} + \frac{1}{E_0+\Delta_0}\right]$$

Matrix element do not change much with *T*

$$E_{P(T)} = E_{P,4\text{K}}\left(\frac{a_{4\text{K}}}{a(T)}\right)$$

Hole effective masses: $\frac{m_0}{m_{hh/lh}} = \frac{1}{\hbar^2}\left[-A \pm \sqrt{B^2 + \frac{C^2}{5}}\right]$

=> *T* dependence due to DKK parameters:

$$A = \frac{1}{3}[F + 2G + 2M] + 1$$

$$B = \frac{1}{3}[F + 2G - M]$$

$$C = \frac{1}{3}[(F - G + M)^2 - +(F + 2G - M)^2]$$

$$F(T) = -\frac{E_{P,4\text{K}}\left(\frac{a_{4\text{K}}}{a(T)}\right)}{E_0(T)}$$

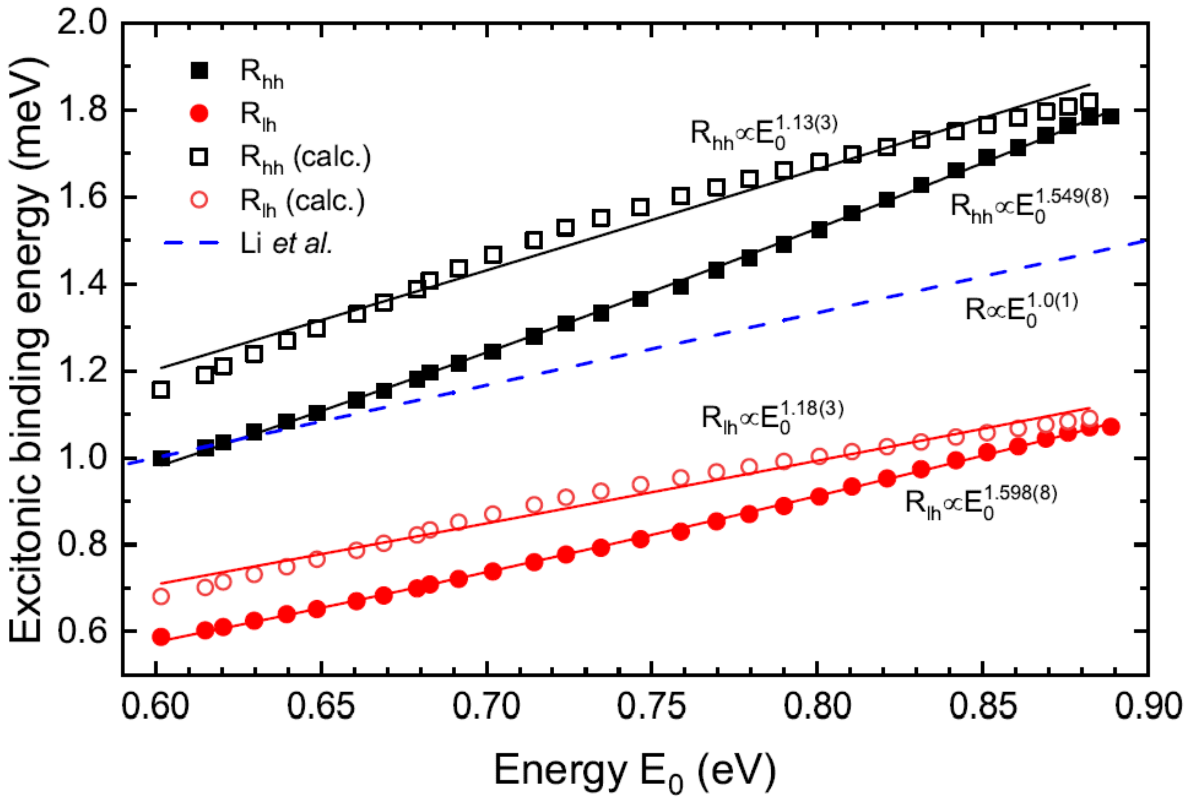
$$M(T) = -\frac{E_{Q,4\text{K}}\left(\frac{a_{4\text{K}}}{a(T)}\right)}{E_0'(T)}$$

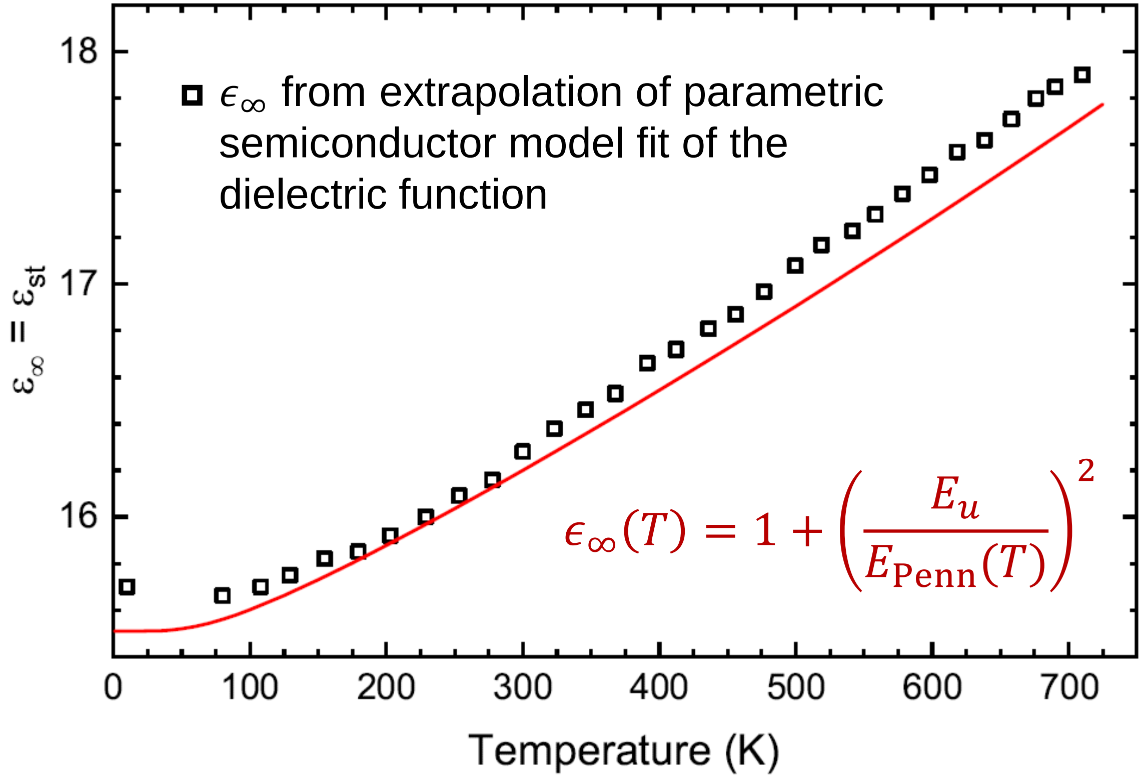
$$G(T) = -G_{4\text{K}}\left(\frac{a_{4\text{K}}}{a(T)}\right)$$

=> *T* dependence stems mainly from $E_0(T)$ and $E_0'(T)$ (and ϵ_{st})

C. Persson and U. Lindefelt, J. Appl. Phys. **82**, 5496 (1997).
M. Cardona, J. Phys. Chem. Solids **24**, 1543 (1963).

Excitonic binding energy and high-frequency dielectric constant





Excitonic binding energy depends on $\epsilon_{\mathrm{st}} = \epsilon_\infty$:

$$R_{hh/lh} \propto \epsilon_{\mathrm{st}}^{-2}$$

$E_u = \hbar\omega_u = 15.6$ eV (ω_u: Plasma frequency)

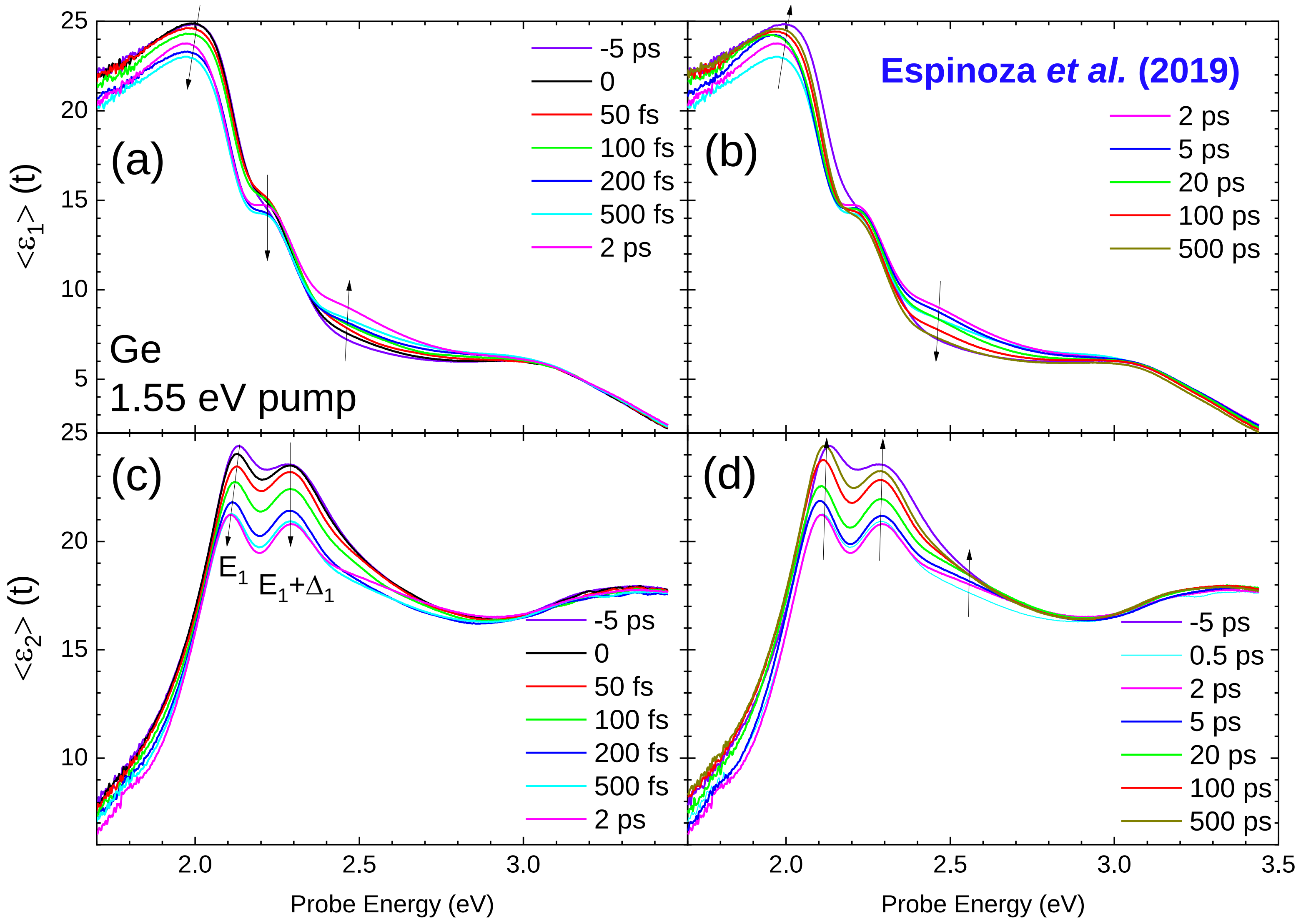
T dependence of Penn gap similar to $E_0'(T)$

$$E_{\mathrm{Penn}}(T) = 4.146 \text{ eV} - 0.05 \text{ eV} \left(\frac{2}{e^{217\,\mathrm{K}/T} - 1} + 1\right)$$

G. H. Li, A. R. Goñi, K. Syassen, and M. Cardona, Phys. Rev. B **49**, 8017 (1994).
P. Yu and M. Cardona, *Fundamentals of Semiconductors*, (Springer, Heidelberg, 2010).
N. S. Samarasingha and S. Zollner, J. Vac. Sci. Technol. B **39**, 052201 (2021).

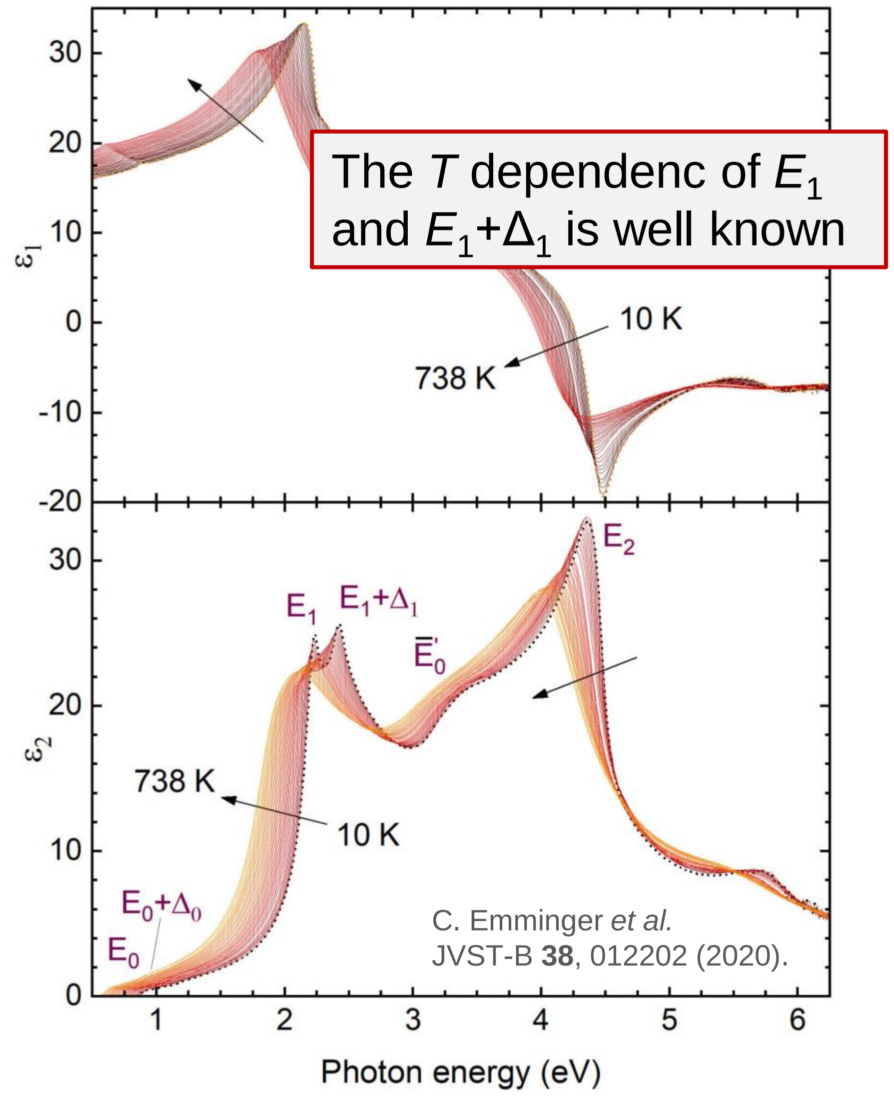
Part 2: Transient critical point parameters of Ge and Si from femtosecond pump-probe ellipsometry

Transient pseudodielectric function from pump-probe spectroscopic ellipsometry



S. Espinoza, S. Richter, M. Rebarz, O. Herrfurth, R. Schmidt-Grund, J. Andreasson, and S. Zollner, Appl. Phys. Lett. **115**, 052105 (2019).

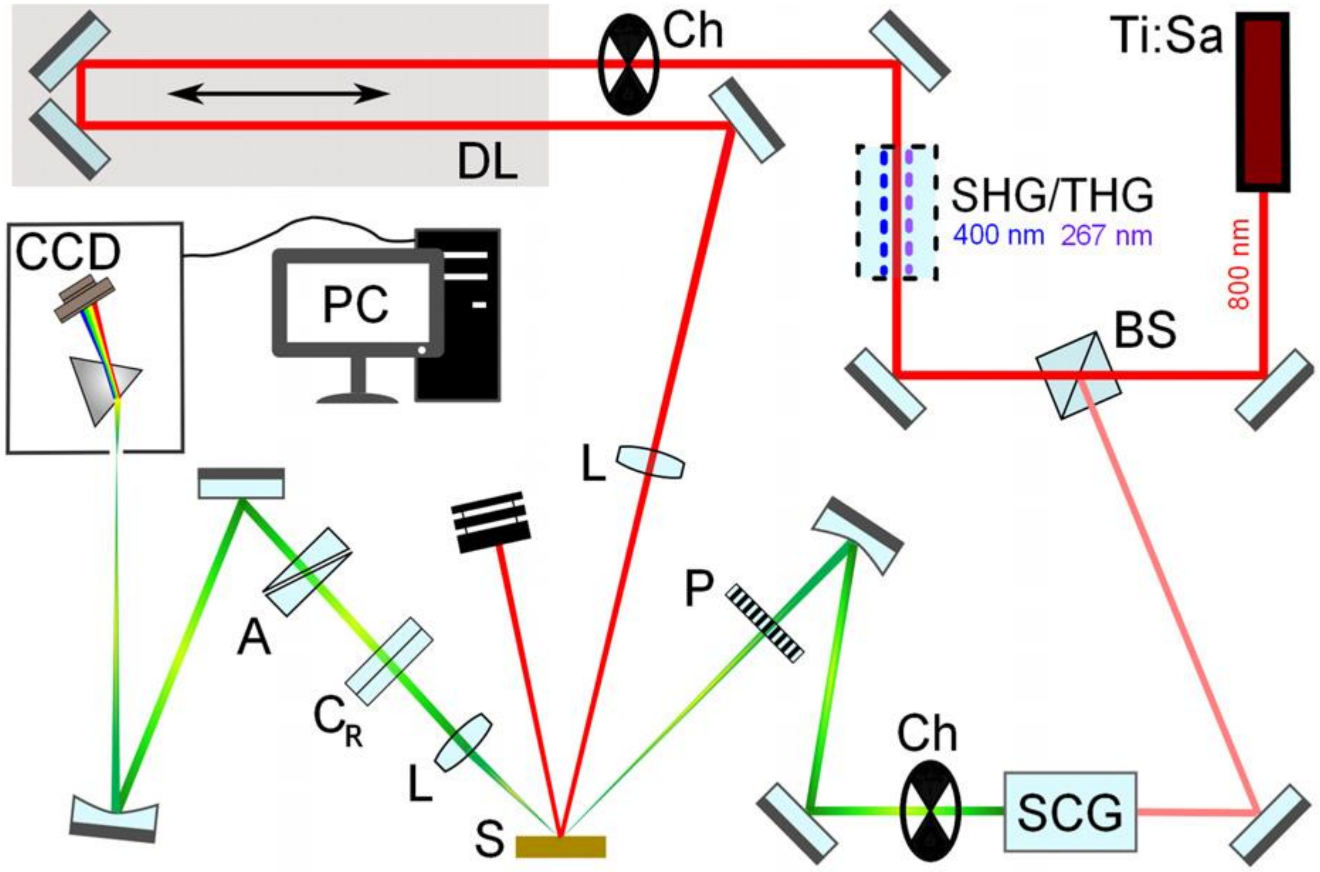
Temperature dependent dielectric function from spectroscopic ellipsometry



L. Viña, S. Logothetidis, and M. Cardona, Phys. Rev. B **30**, 1979 (1984).
N. S. Fernando *et al.*, Appl. Surf. Sci. **421**, 905 (2017).

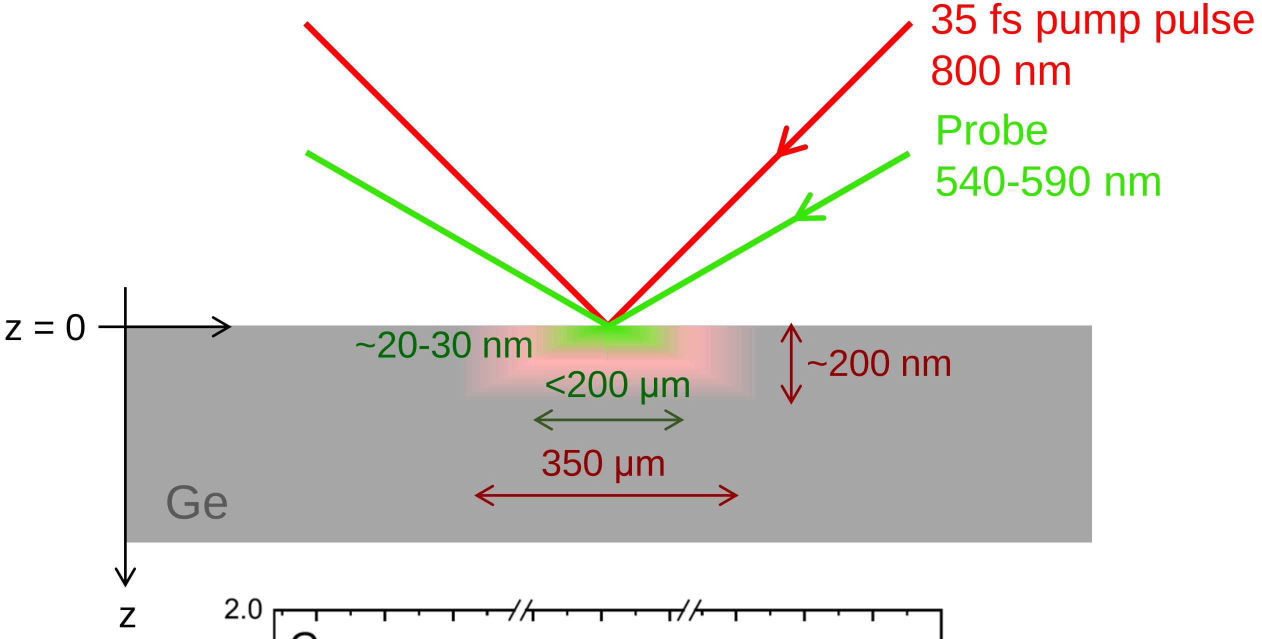
Pump-probe spectroscopic ellipsometry setup

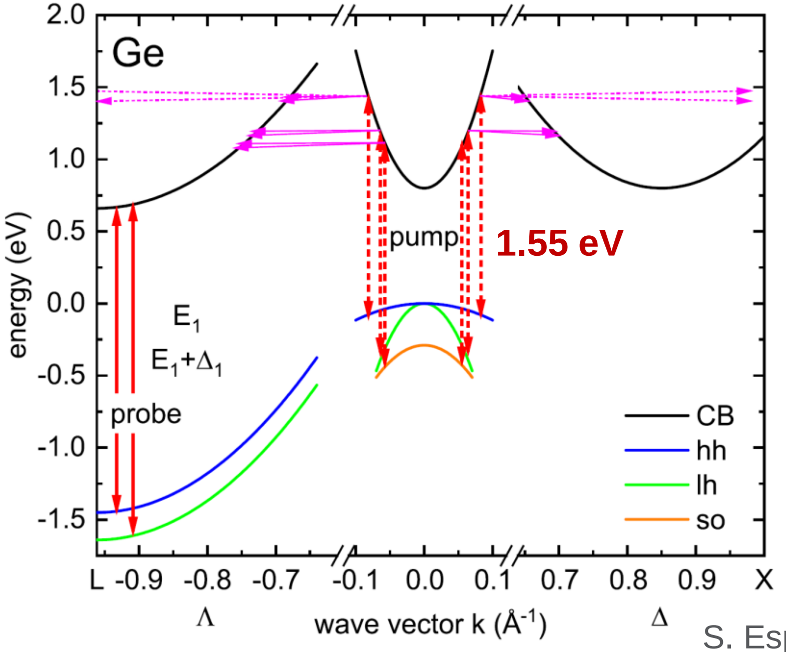
- Pump pulse: 266, 400, and 800 nm
- 35 fs laser pulses
- Repetition rate: 1 kHz
- Pulse energy: up to 6 mJ
- Carrier density: $10^{20}\ \mathrm{cm}^{-3}$
- Time resolution: 120 fs (oblique incidence)
- Spectral range: 1.7 – 3.5 eV
- Probe beam diameter <200 µm
- Pump beam diameter ~350 µm



S. Espinoza, S. Richter, M. Rebarz, O. Herrfurth, R. Schmidt-Grund, J. Andreasson, and S. Zollner, Appl. Phys. Lett. **115**, 052105 (2019)
S. Richter, M. Rebarz, O. Herrfurth, S. Espinoza, R. Schmidt-Grund, and J. Andreasson, Rev. Sci. Instrum. **92**, 033104 (2021)

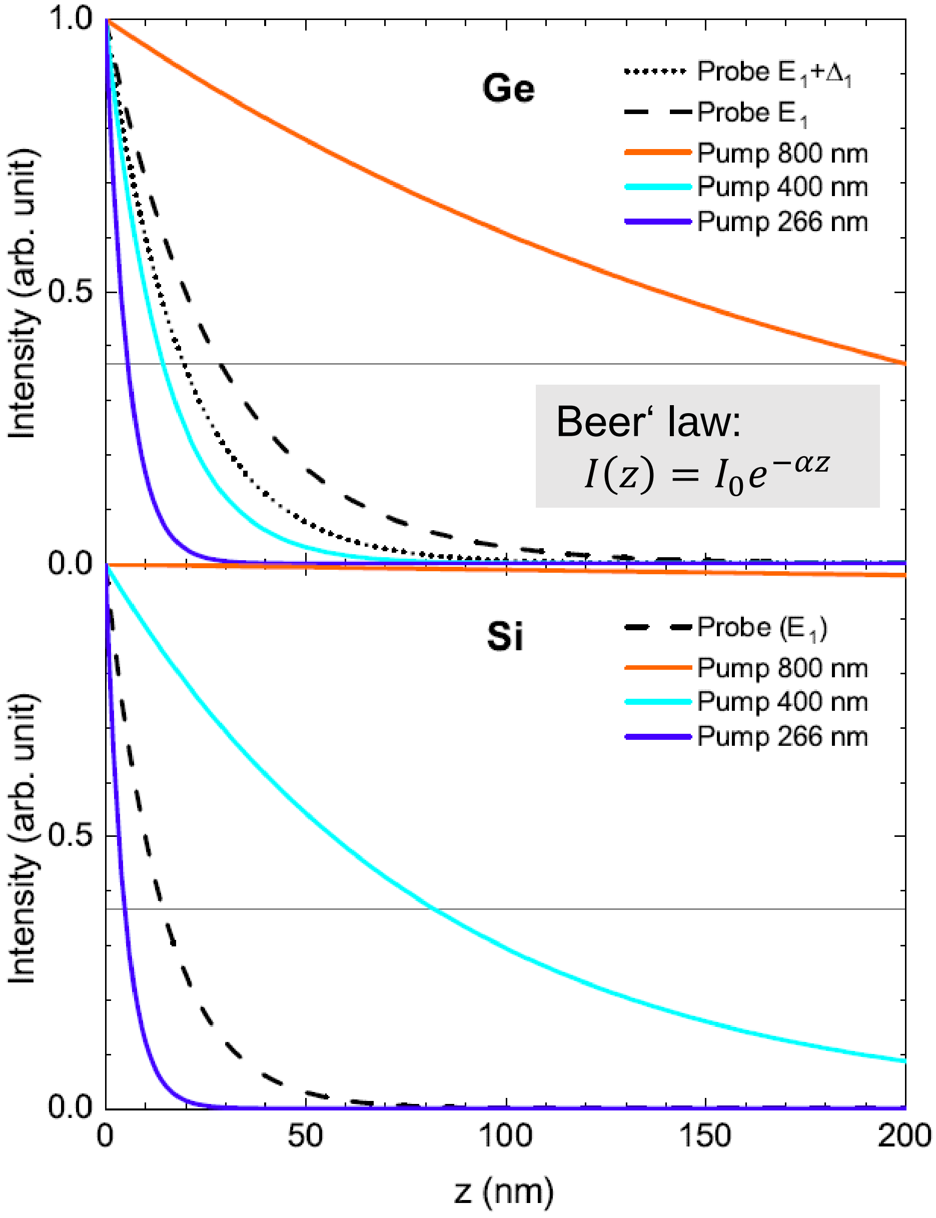
Penetration depth of probe and pump beams





Carrier density:
10^{20} cm^{-3}

Electrons scatter from Γ to X and L



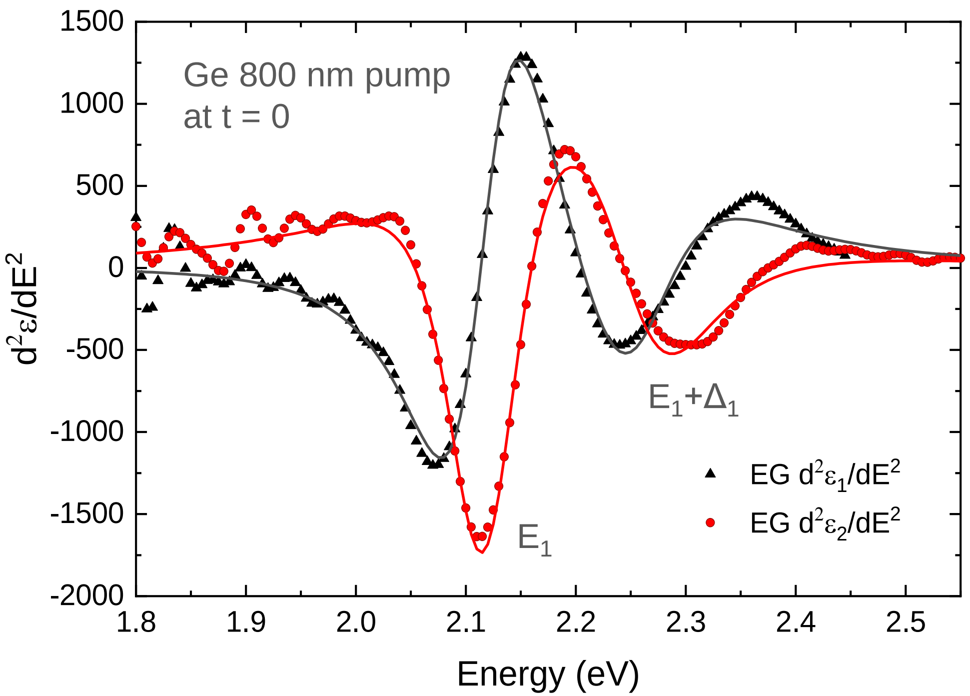
S. Espinoza *et al.*, Appl. Phys. Lett. **115**, 052105 (2019)

Calculation of the second derivatives

Second derivative of the DF using EG-filters for *M* = 4

- for data sets with equidistant energy steps Δ*E*’:

$$\frac{d^2\bar{\epsilon}(E)}{dE^2} \approx \frac{\Delta E'}{49152\sqrt{\pi}\Delta E^{13}} \sum_{j=-\infty}^{\infty} \epsilon(E_j)\Big((E-E_j)^{10} - 106(E-E_j)^8\Delta E^2 + 3608(E-E_j)^6\Delta E^4 - 45936(E-E_j)^4\Delta E^6 + 188496(E-E_j)^2\Delta E^8 - 110880\,\Delta E^{10}\Big) e^{-\frac{(E-E_j)^2}{4\Delta E^2}}$$



- for data sets with nonconstant energy steps:

$$\frac{d^2\bar{\epsilon}(E)}{dE^2} \approx \frac{1}{49152\sqrt{\pi}\Delta E^{13}} \sum_{j=-\infty}^{\infty} \epsilon(E_j)\Big((E-E_j)^{10} - 106(E-E_j)^8\Delta E^2 + 3608(E-E_j)^6\Delta E^4 - 45936(E-E_j)^4\Delta E^6 + 188496(E-E_j)^2\Delta E^8 - 110880\,\Delta E^{10}\Big) \frac{E_{j+1}-E_{j-1}}{2} e^{-\frac{(E-E_j)^2}{4\Delta E^2}}$$

V. L. Le, T. J. Kim, Y. D. Kim, D. E. Aspnes, J. Vac. Sci. Technol. B **37**, 052903 (2019)

Critical point analysis: Second derivatives from linear filters

Second derivatives calculated using a digital linear filter method (Le *et al.* 2019)

Ge: E_1 and $E_1+\Delta_1$

- EG filter width: 12-15 meV
- Fit: 2D-lineshape

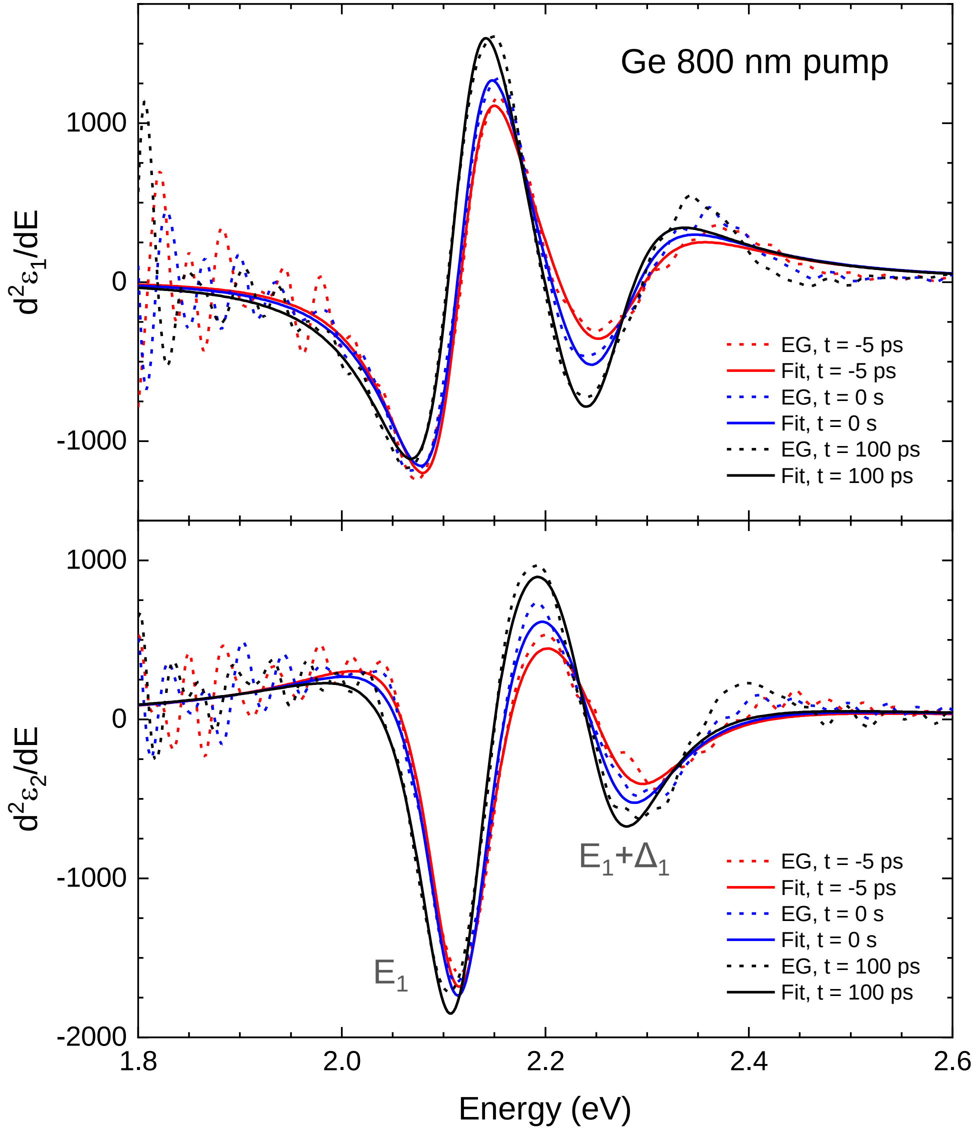
$$\epsilon_{2D}(E) = B - Ae^{i\varphi}\ln\left(E - E_g + i\Gamma\right)$$

Si: E_1

- EG filter width: 20 meV
- Fit: 0D-lineshape

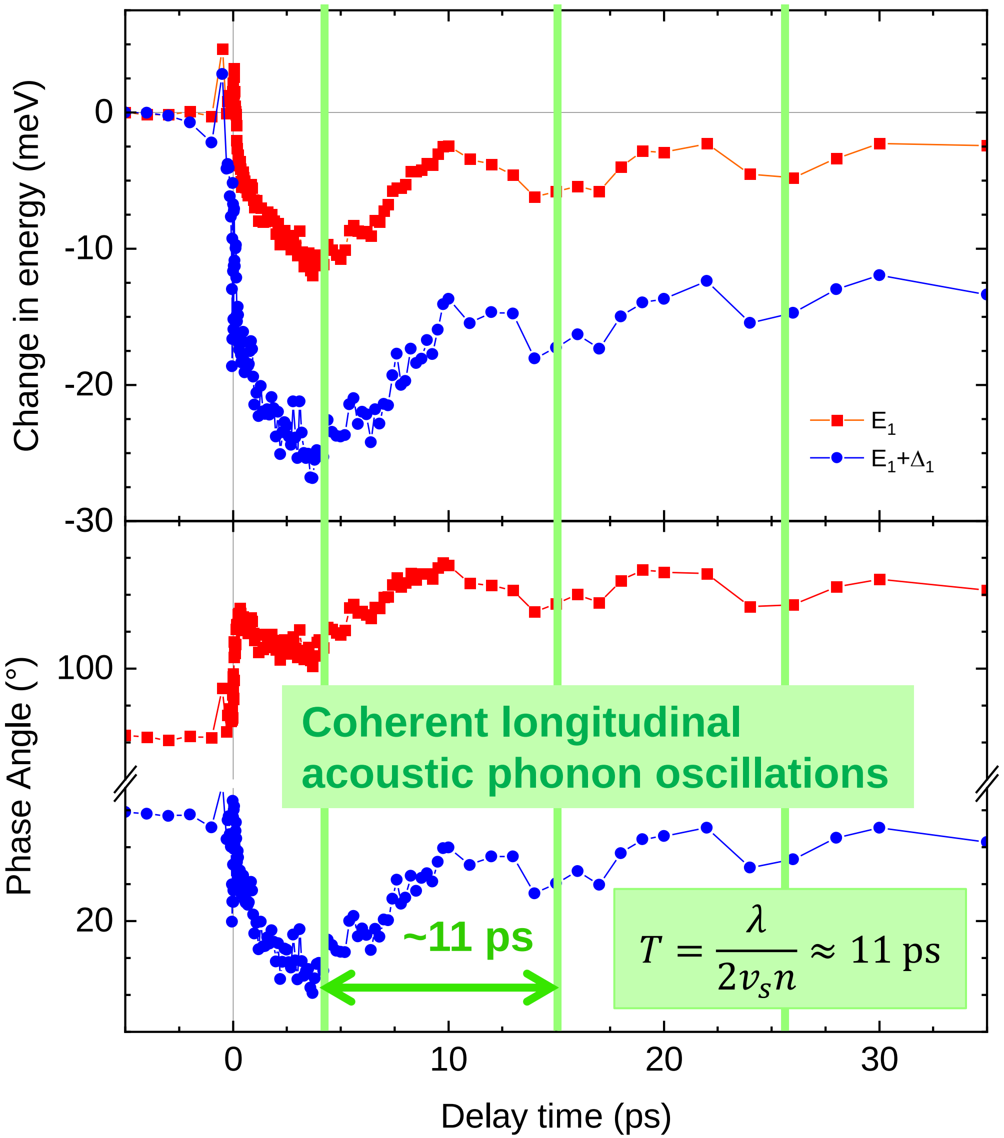
$$\epsilon_{0D}(E) = B - \frac{Ae^{i\varphi}}{E - E_g + i\Gamma}$$

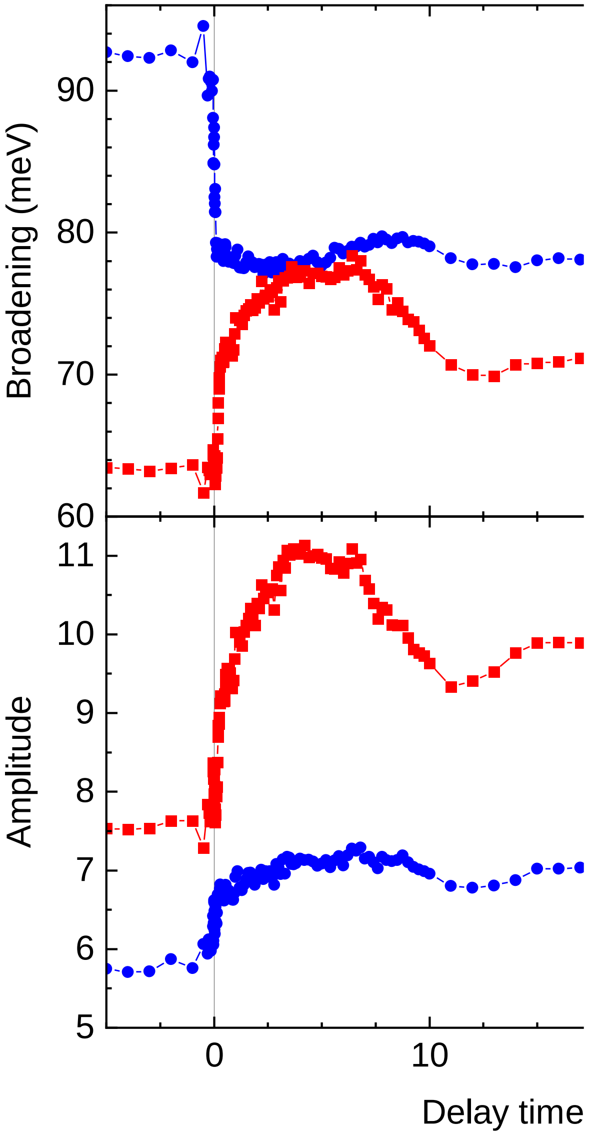
=> Better: Lineshape considering bandfilling effects

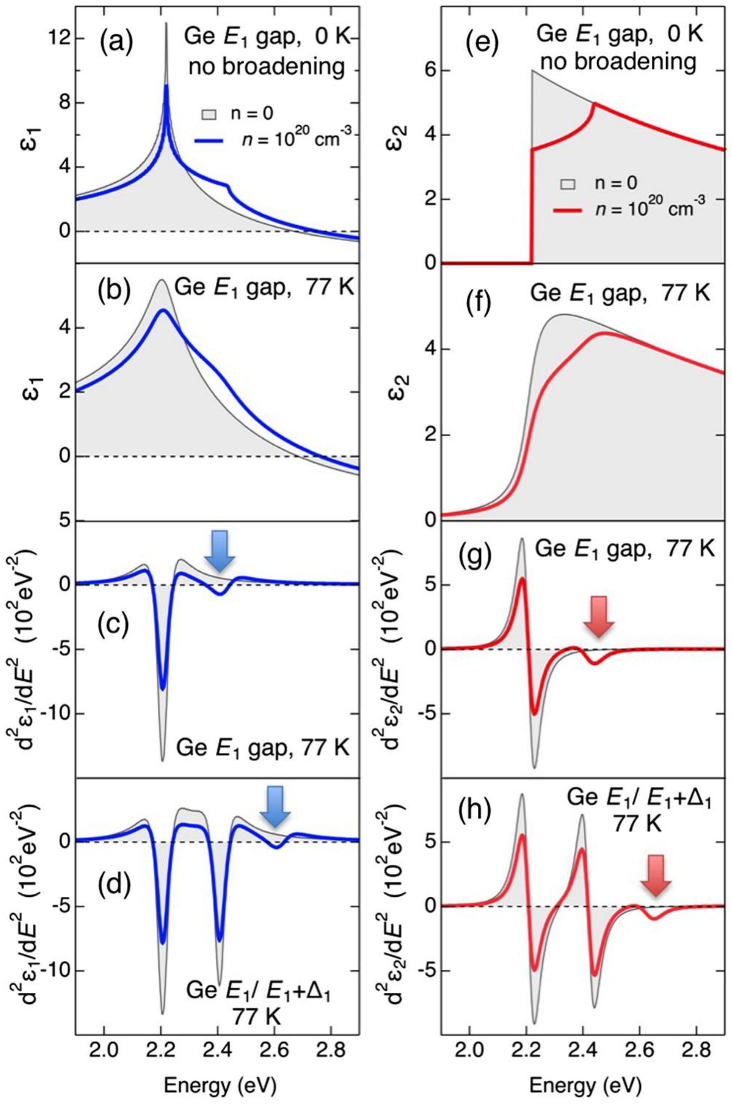


D. E. Aspnes, *Handbook on Semiconductors*, edited by M. Balkanski (North-Holland, Amsterdam, 1980).

Critical point parameters as functions of delay time – Ge 800 nm pump







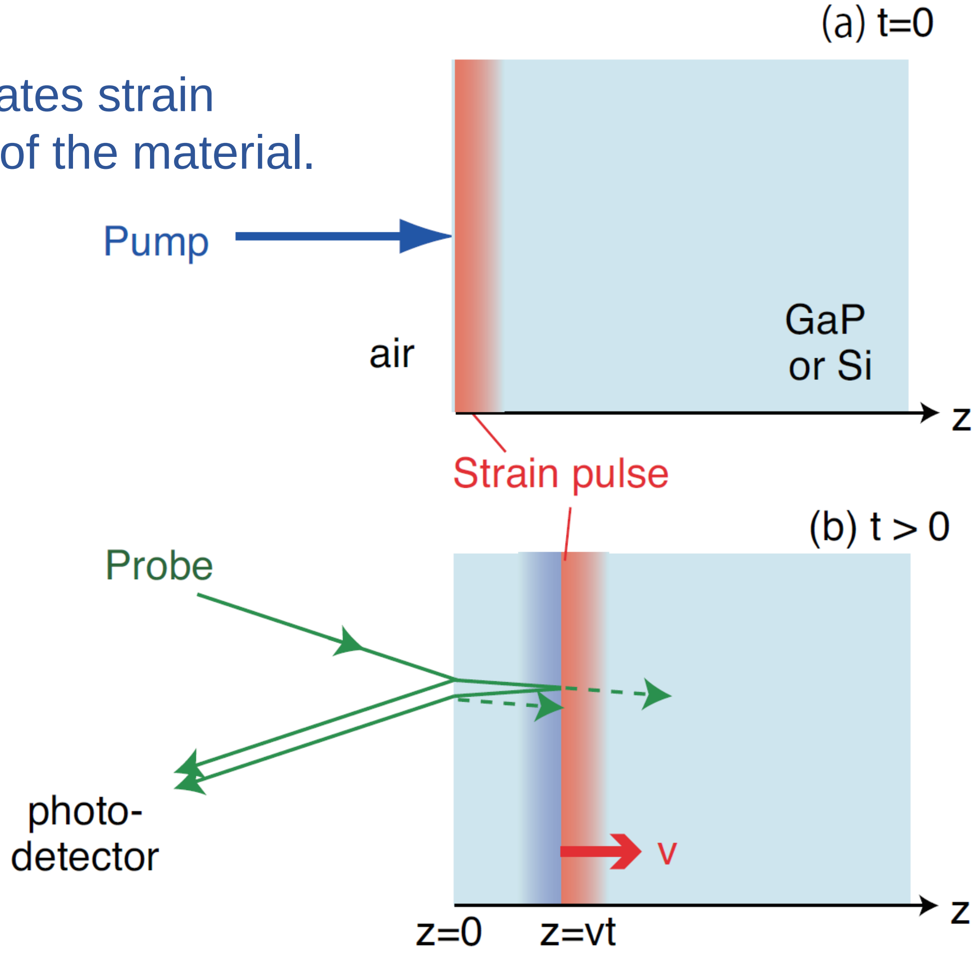
C. Xu, N. S. Fernando, S. Zollner, J. Kouvetakis, and J. Menéndez, Phys. Rev. B **118**, 267402 (2017).

C. Thomsen, H. T. Grahn, H. J. Maris, and J. Tauc, Phys. Rev. B 34, 4129 (1986)

Creation and propagation of a strain pulse

The pump pulse creates strain close to the surface of the material.

The probe pulse gets reflected by the strain pulse, which moves through the crystal.



Period: $T = \dfrac{\lambda}{2 v_s n}$

λ …. probe wavelength
v_s … longitudinal sound velocity
n …. refractive index

K. Ishioka, V. Rustagi, U. Höfer, H. Petek, and C. J. Stanton, Phys. Rev. B **95**, 035205 (2017).

Coherent phonon oscillations – energy shifts (Ge 800 nm pump)

The oscillations can be fitted with a damped oscillator and an exponential decay:

$$\Delta E(t) = \Delta E_a(t) + \Delta E_b(t) = -E_a \cos\left(\frac{2\pi t}{T} - \delta\right) e^{\frac{t}{\tau_a}} - E_b e^{\frac{t}{\tau_b}}$$

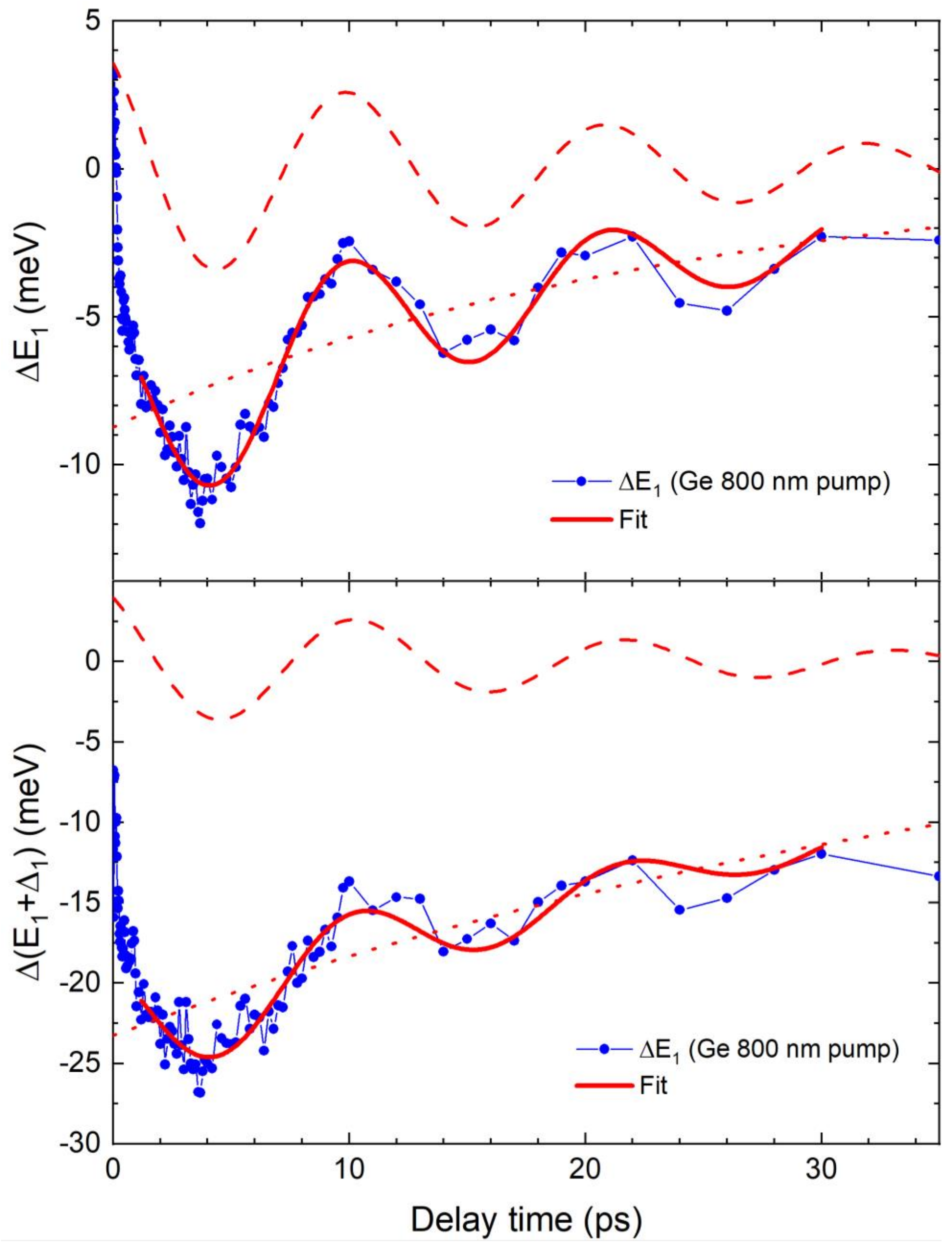
Fit result:

	$\Delta(E_1)$	$\Delta(E_1 + \Delta_1)$
E_a (meV)	4.2 ± 0.4	4.7 ± 0.8
E_b (meV)	8.7 ± 0.3	23.3 ± 0.5
T (ps)	11.0 ± 0.2	11.4 ± 0.4
δ	2.58 ± 0.06	2.6 ± 0.1
τ_a (ps)	20 ± 4	18 ± 7
τ_b (ps)	23 ± 2	42 ± 3

Temperature increase:

$$\Delta T \approx \frac{E(1-R)}{cV\rho} \approx 25\ \mathrm{K}$$

$\Delta E_1 \approx -10$ meV => $\Delta T \approx 20$ K
$\Delta(E_1 + \Delta_1) \approx -25$ meV => $\Delta T \approx 40$ K



L. Viña, S. Logothetidis, M. Cardona, Phys. Rev. B **30**, 1979 (1984).

Calculating strain and energy shifts (Ge 800nm pump)

Stress-strain relation: $\epsilon_3 = (S_{11} + 2S_{12})\sigma = \frac{\sigma}{C_{11}+2C_{12}}$ (assuming: $\sigma_{ii} = \sigma$ for all i and $\sigma_{ij} = 0$ for $i \neq j$)

$(C_{11} + 2C_{12})^{-1} = 4.44 \times 10^{-8} \frac{\text{cm}^2}{\text{N}}$

Electron contribution: $\sigma_{\text{el}} = -B \frac{\partial E_g}{\partial P} N$ => $\epsilon_{\text{el}} = (C_{11} + 2C_{12})^{-1}\sigma_{\text{el}} = -6.4 \times 10^{-4}$

Phonon contribution: $\sigma_{\text{ph}} = -\frac{3B\beta}{c}(E - E_g)N$ => $\epsilon_{\text{ph}} = (C_{11} + 2C_{12})^{-1}\sigma_{\text{ph}} = -1.2 \times 10^{-4}$

Total stress/strain: $\sigma_{33} = \sigma_{\text{el}} + \sigma_{\text{ph}}$ => $\epsilon_{33} = (C_{11} + 2C_{12})^{-1}\sigma_{33} = -7.6 \times 10^{-4}$

In-plane and out-of-plane strain: $\epsilon_{\perp} = \epsilon_{33}$ and $\epsilon_{\parallel} = 0$

Hydrostatic and shear strain: $\epsilon_H = \epsilon_S = 2.5 \times 10^{-4}$

Hydrostatic shift: $\Delta E_H = \sqrt{3} D_1^1 \epsilon_H = -3.4 \text{ meV}$

Shear splitting: $\Delta E_S = \sqrt{6} D_3^3 \epsilon_S = 1.6 \text{ meV}$

Critical point energy shift: $\Delta E_1 = \frac{\Delta_1}{2} + \Delta E_H - \sqrt{\frac{(\Delta_1)^2}{4} + (\Delta E_S)^2} = -3.4 \text{ meV}$

$$\Delta(E_1 + \Delta_1) = -\frac{\Delta_1}{2} + \Delta E_H + \sqrt{\frac{(\Delta_1)^2}{4} + (\Delta E_S)^2} = -3.4 \text{ meV}$$

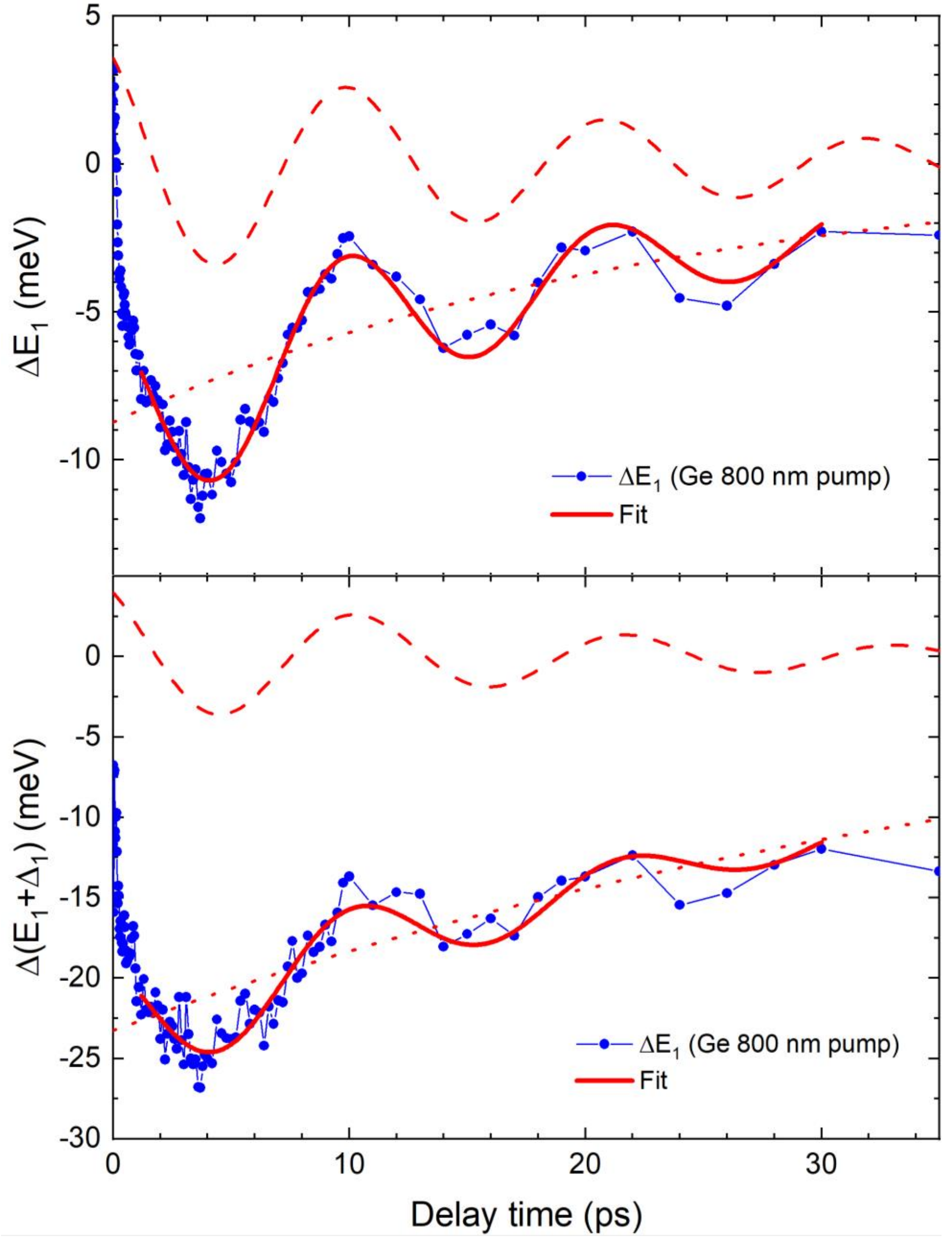
C. Thomsen, H. T. Grahn, H. J. Maris, and J. Tauc, Phys. Rev. B 34, 4129 (1986).
P. Yu and M. Cardona, *Fundamentals of Semiconductors*, (Springer, Heidelberg, 2010).
S. Zollner, in: S.T. Pantelides, S. Zollner (Eds.), *Silicon-Germanium Carbon Alloys: Growth, Properties and Applications*, (Taylor & Francis, New York, 2002).

Calculating strain from observed oscillations (Ge 800 nm pump)

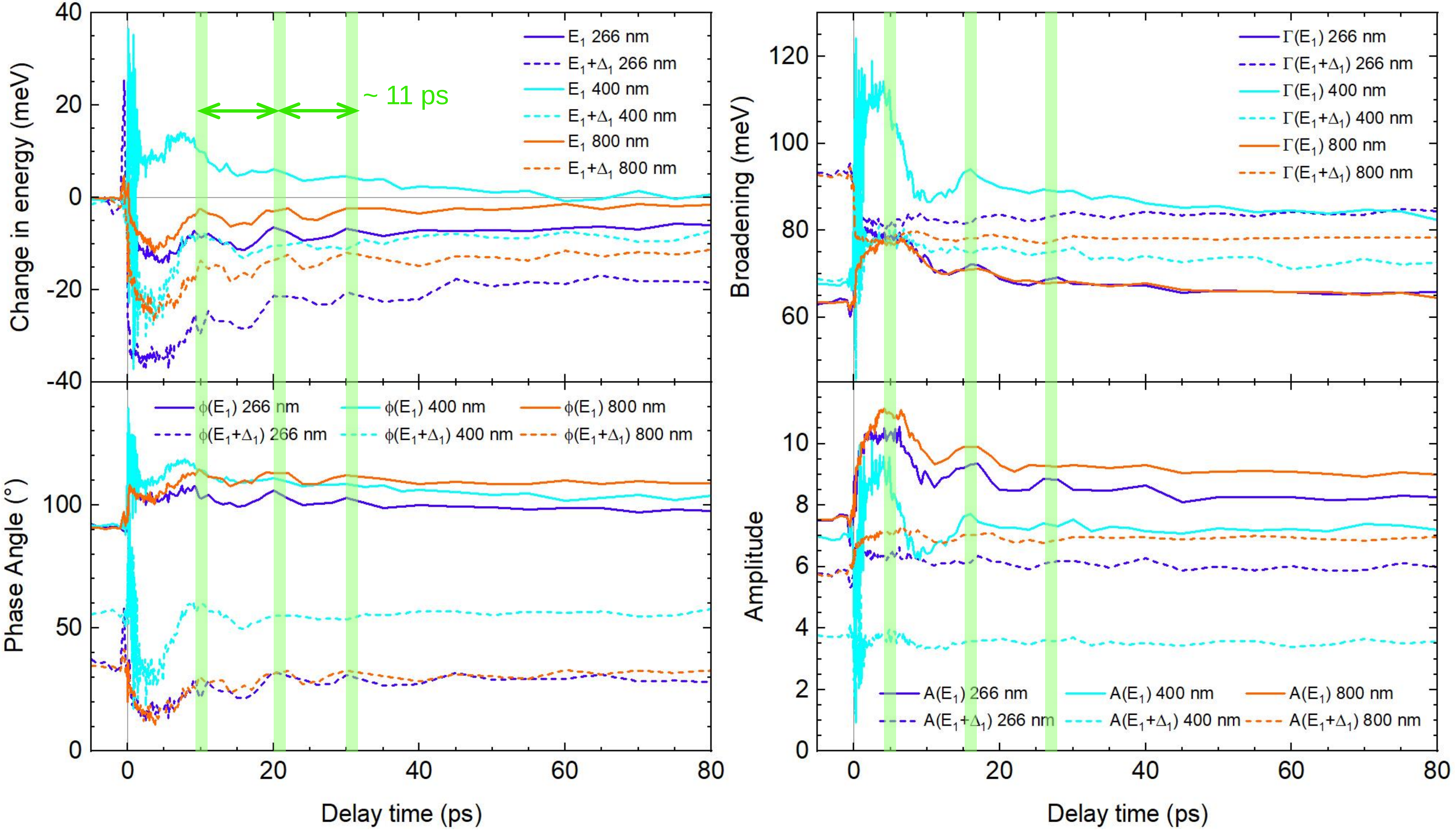
	$\Delta(E_1)$	$\Delta(E_1+\Delta_1)$
E_a (meV)	4.2 ± 0.4	4.7 ± 0.8
E_b (meV)	8.7 ± 0.3	23.3 ± 0.5
T (ps)	11.0 ± 0.2	11.4 ± 0.4
δ	2.58 ± 0.06	2.6 ± 0.1
τ_a (ps)	20 ± 4	18 ± 7
τ_b (ps)	23 ± 2	42 ± 3

- **Adding energy shifts (data):**
  $\Delta E_1 + \Delta(E_1+\Delta_1) = 2\Delta E_H \approx 9$ meV
  => $\Delta E_H \approx 4.5$ meV
  => $\epsilon_H = \frac{\Delta E_H}{\sqrt{3}D_1^1} \approx -3.3 \times 10^{-4}$
  => $\epsilon_\perp = 3\epsilon_H \approx -1 \times 10^{-3}$
- **Subtracting energy shifts (data):**
  $|\Delta E_1^S - \Delta(E_1+\Delta_1)^S| = \left|\Delta_1 - 2\sqrt{\frac{(\Delta_1)^2}{4} + (\Delta E_S)^2}\right| \approx 0.5$ meV
  => $|\Delta E_S| \approx 7.0$ meV
  => $|\epsilon_S| = \frac{|\Delta E_S|}{\sqrt{6}D_3^3} \approx 1.1 \times 10^{-3}$
  =>$\epsilon_\perp = 3\epsilon_S \approx -3.3 \times 10^{-3}$

Compares well with calculated strain:
$\epsilon_{33} = -7.6 \times 10^{-4}$

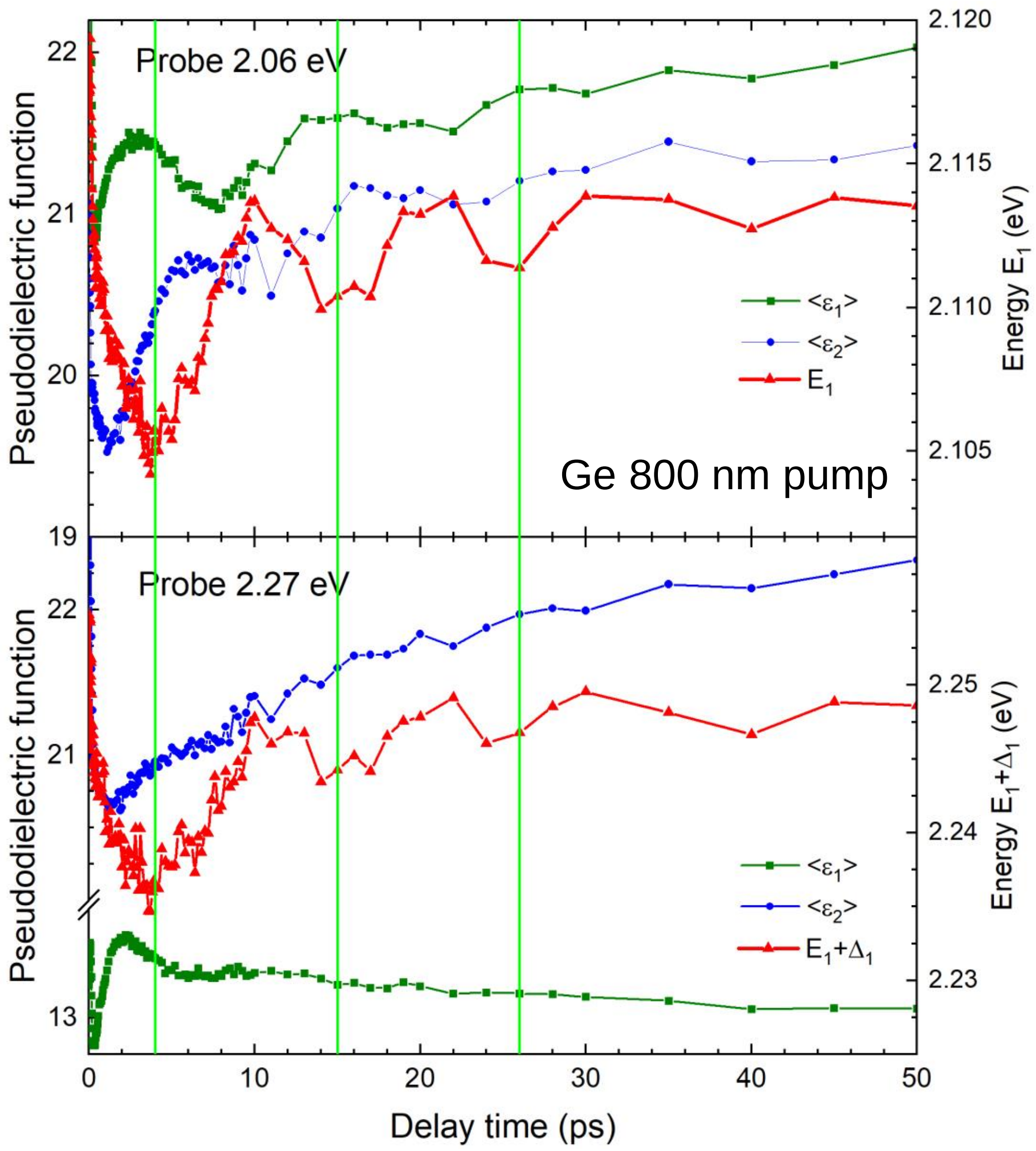


Critical point parameters of Ge (266, 400, and 800 nm pump)



=> Oscillations present in all Ge data sets (266, 400, and 800 nm pump)

Coherent longitudinal acoustic phonon oscillations



→ Oscillations in the CP parameters more pronounced than in the dielectric function

Ge

E_1

$\lambda = 585$ nm
$n = 5.65$
$v_s = 4.87 \times 10^5$ cm/s
$T = \frac{\lambda}{2 v_s n} \approx 11$ ps

$E_1+\Delta_1$

$\lambda = 550$ nm
$n = 5.16$
$v_s = 4.87 \times 10^5$ cm/s
$T = \frac{\lambda}{2 v_s n} \approx 11$ ps

Si

$\lambda = 365$ nm
$n = 6.52$
$v_s = 8.43 \times 10^5$ cm/s
$T = \frac{\lambda}{2 v_s n} \approx 3.3$ ps

InP

$\lambda = 390$ nm
$n = 3.98$
$v_s = 4.58 \times 10^5$ cm/s
$T = \frac{\lambda}{2 v_s n} \approx 11$ ps

GaSb

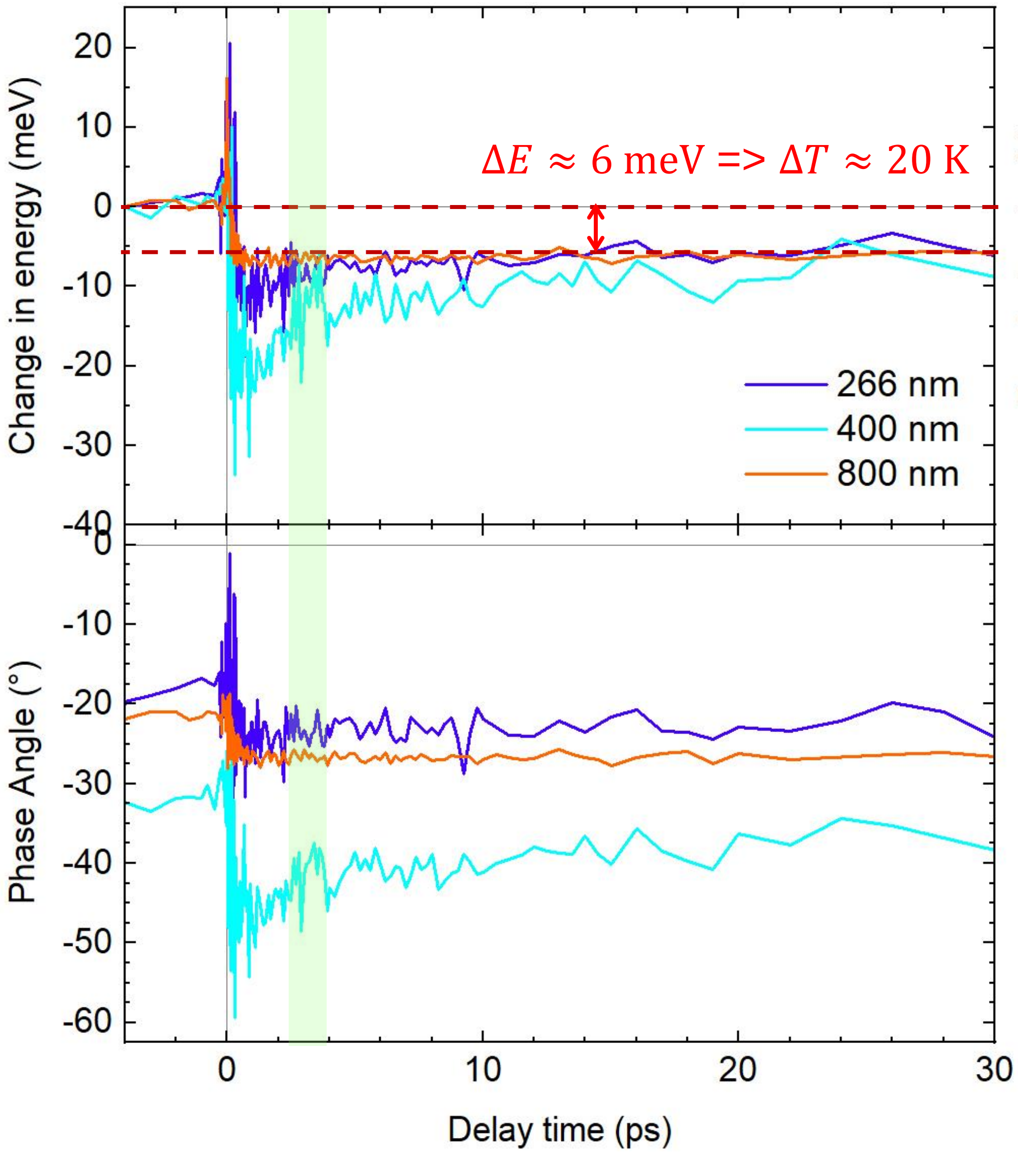
E_1

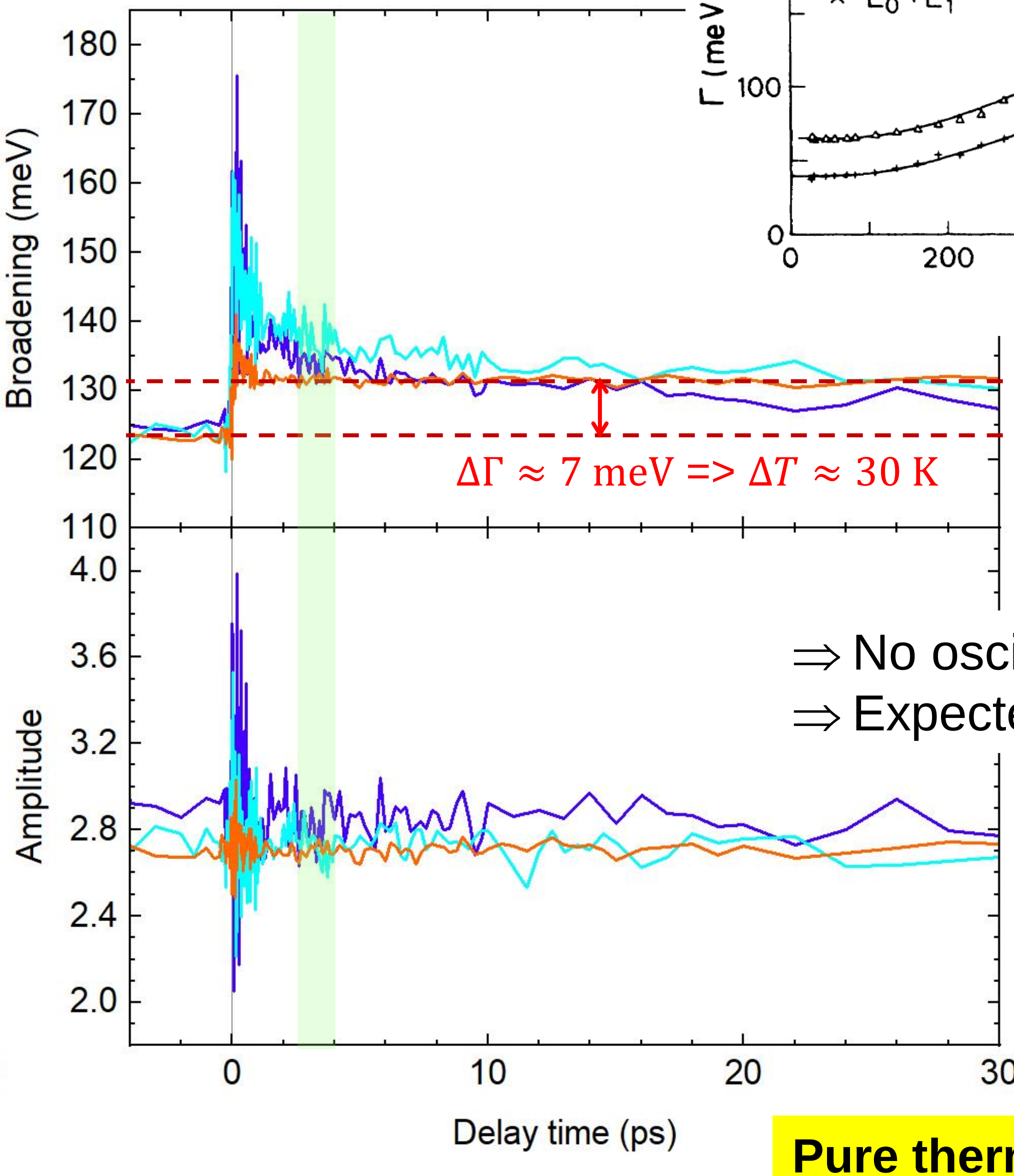
$\lambda = 620$ nm
$n = 5.24$
$v_s = 4 \times 10^5$ cm/s
$T = \frac{\lambda}{2 v_s n} \approx 15$ ps

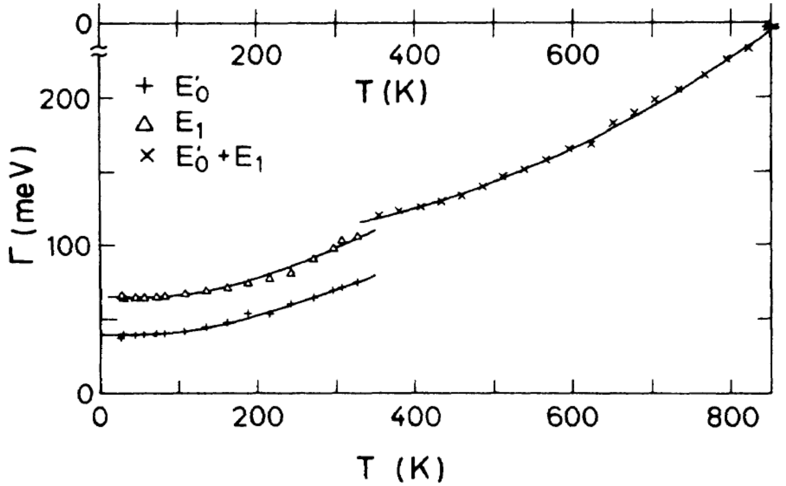
$E_1+\Delta_1$

$\lambda = 510$ nm
$n = 4.45$
$v_s = 4 \times 10^5$ cm/s
$T = \frac{\lambda}{2 v_s n} \approx 14$ ps

Critical point parameters of Si (266, 400, and 800 nm pun







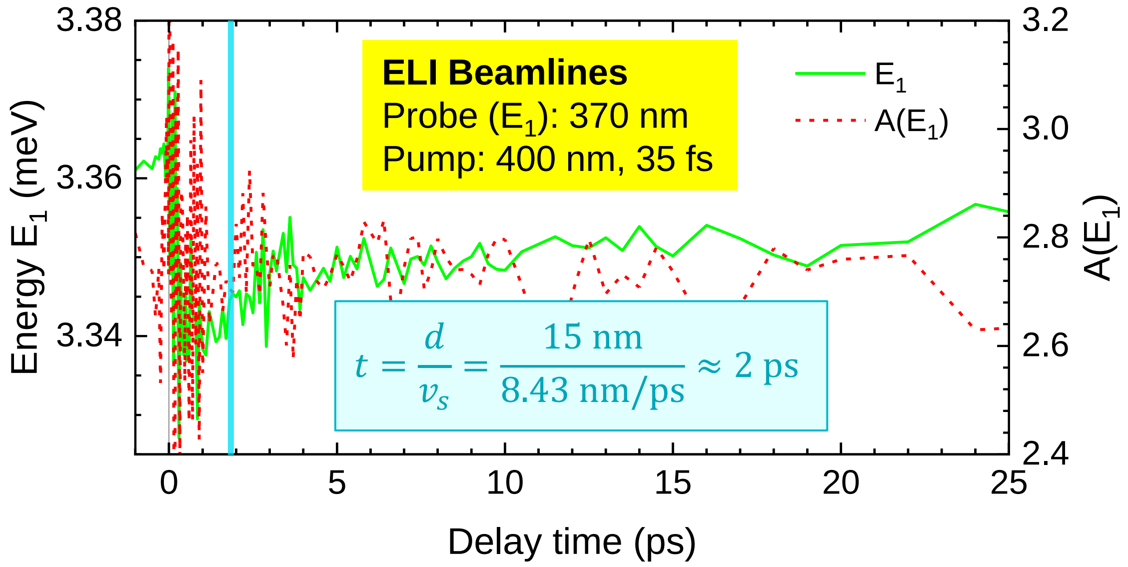
⇒ No oscillations detected
⇒ Expected period:

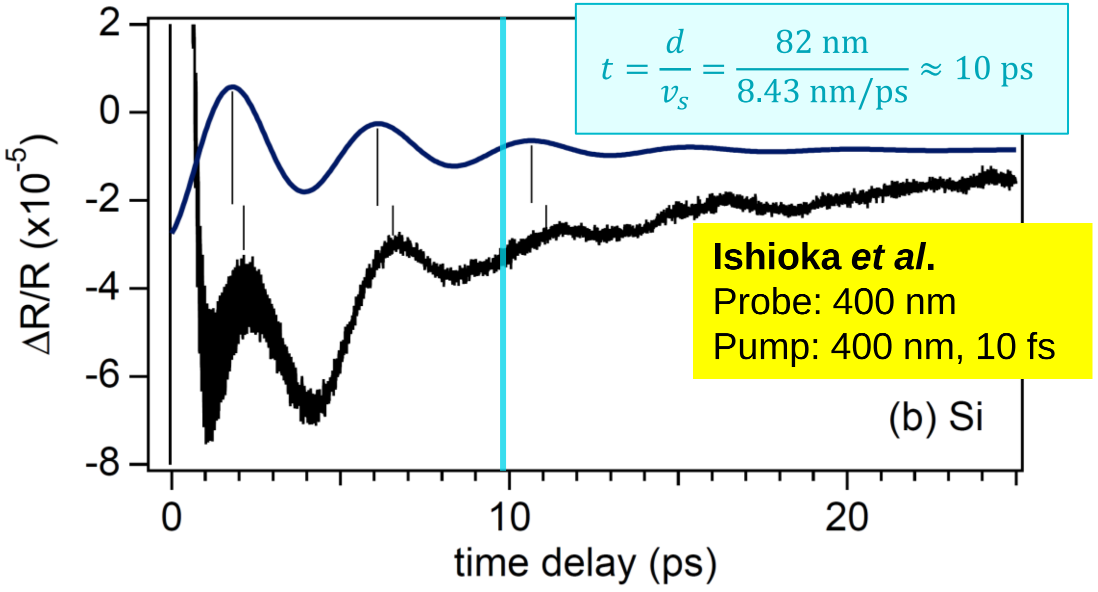
$$T = \frac{\lambda}{2 v_s n} \approx 3\ \text{ps}$$

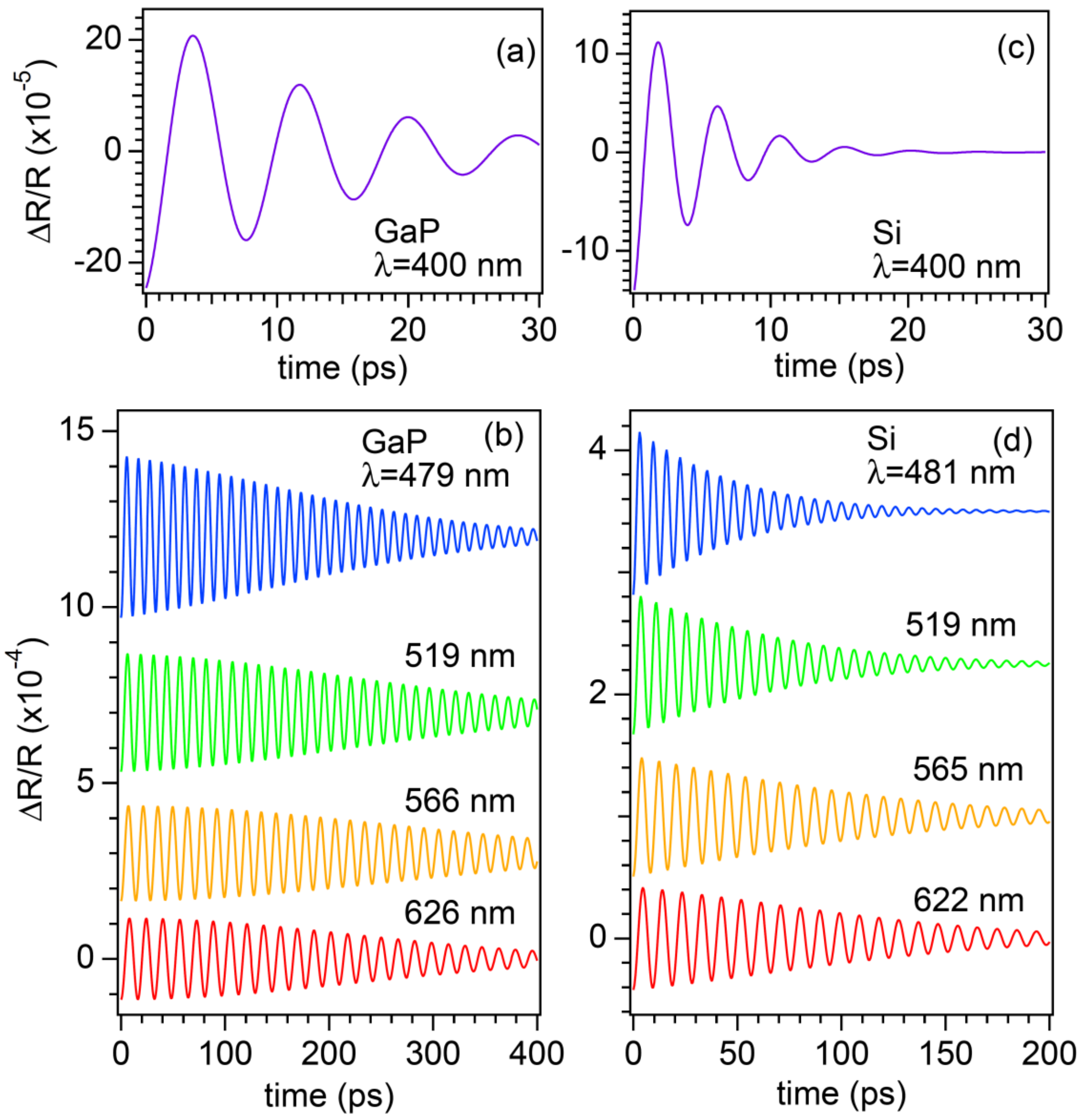
Pure thermal effect

P. Lautenschlager, M. Garriga, L. Viña, and M. Cardona, Phys. Rev. B **36**, 4821 (1987).

Coherent longitudinal acoustic phonon oscillations in Si







K. Ishioka, V. Rustagi, U. Höfer, H. Petek, and C. J. Stanton, Phys. Rev. B **95**, 035205 (2017).

SUMMARY & OUTLOOK

Part 1: Excitonic effects at the direct band gap E_0 of Ge

- **Temperature dependence of E_0 obtained from spectroscopic ellipsometry**
  - Good agreement between model and data despite having only two fit parameters (energy and broadening).
- **Outlook & future work**
  - Application to other semiconductors
  - Consider non-parabolicity and warping

Part 2: Analysis of femtosecond pump-probe ellipsometry data

- **Temporal evolution of E_1 and $E_1+\Delta_1$ in Ge**
  - Oscillations in CP parameters due to coherent longitudinal acoustic phonons.
- **Temporal evolution of CP parameters in Si**
  - No phonon oscillations detected.
- **Outlook & future work**
  - Taking new data with time steps targeted to resolve phonon oscillations.
  - Investigating bandfilling effects.

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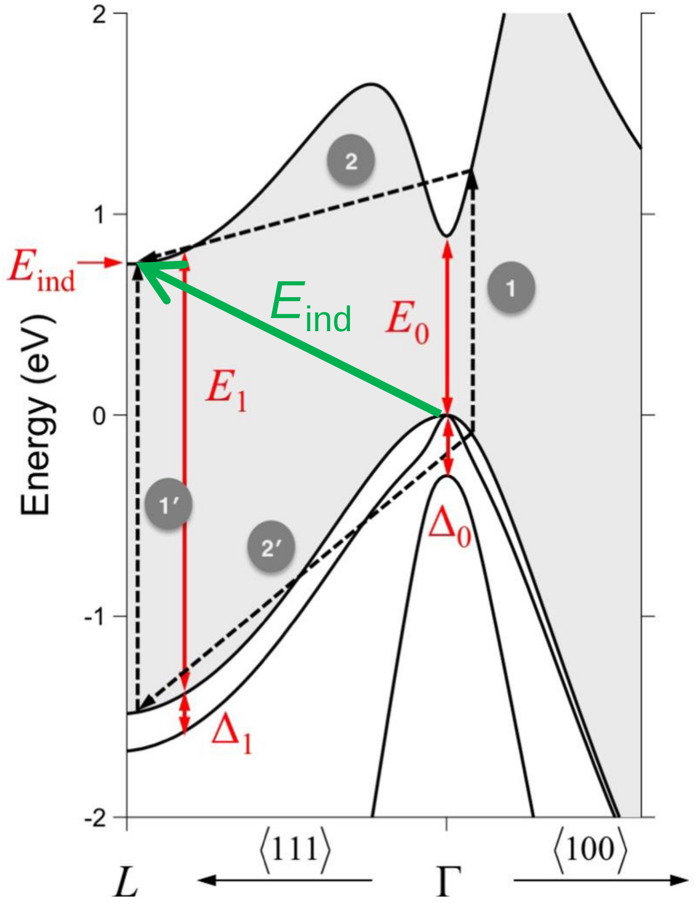
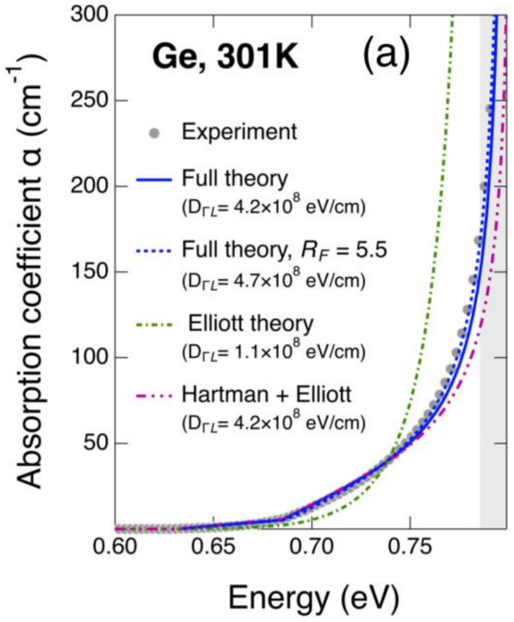
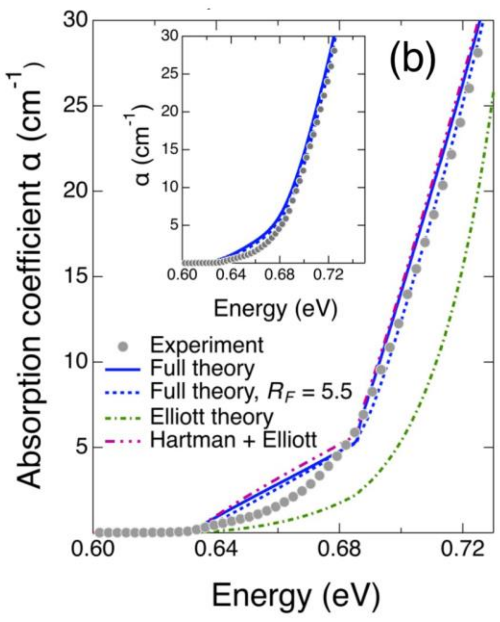
2017 AVS 64th International Symposium and Exhibition
Left to right: Cesy Zamarripa, Rigo Carrasco, Carola Emminger, Prof. John Woollam, Dr. Stefan Zollner, Farzin Abadizaman, Nuwanjula Samarasingha and Pablo Paradis

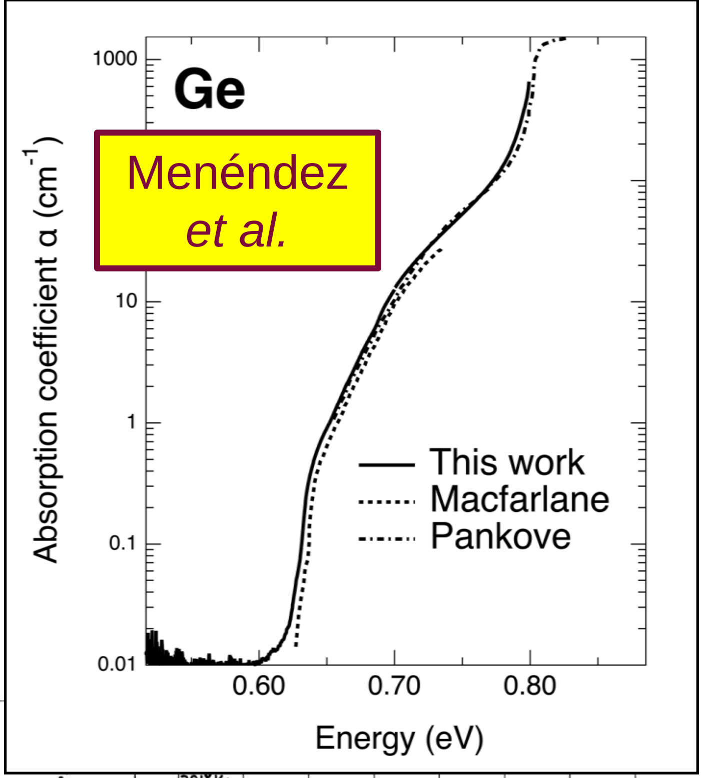
Backup slides

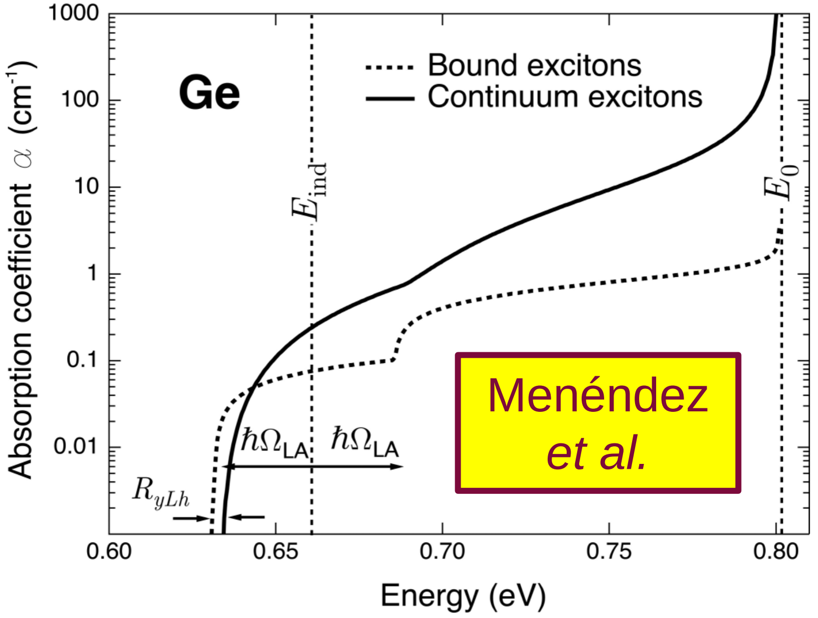
Indirect and direct band gap of Ge

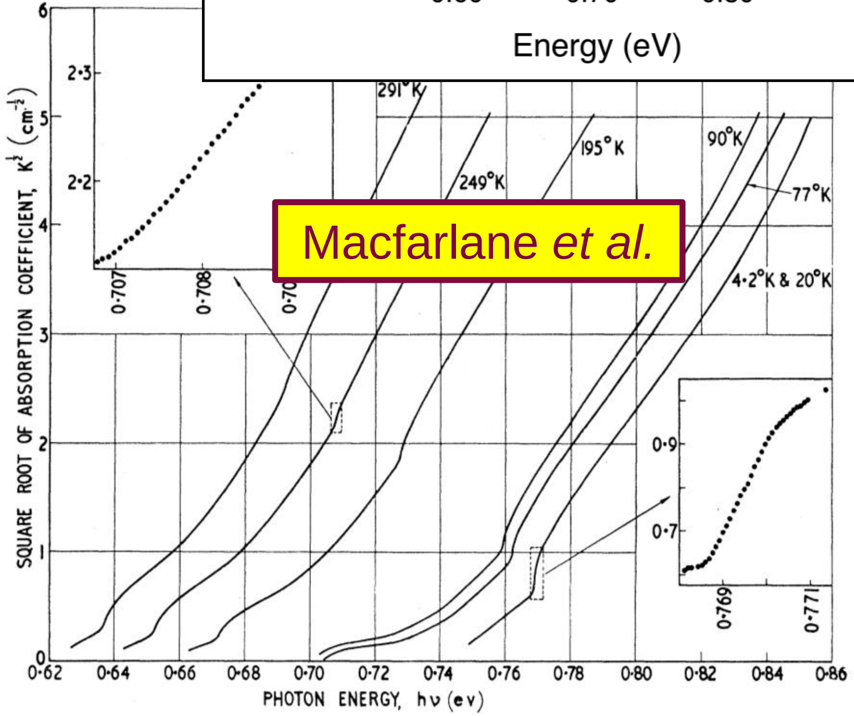




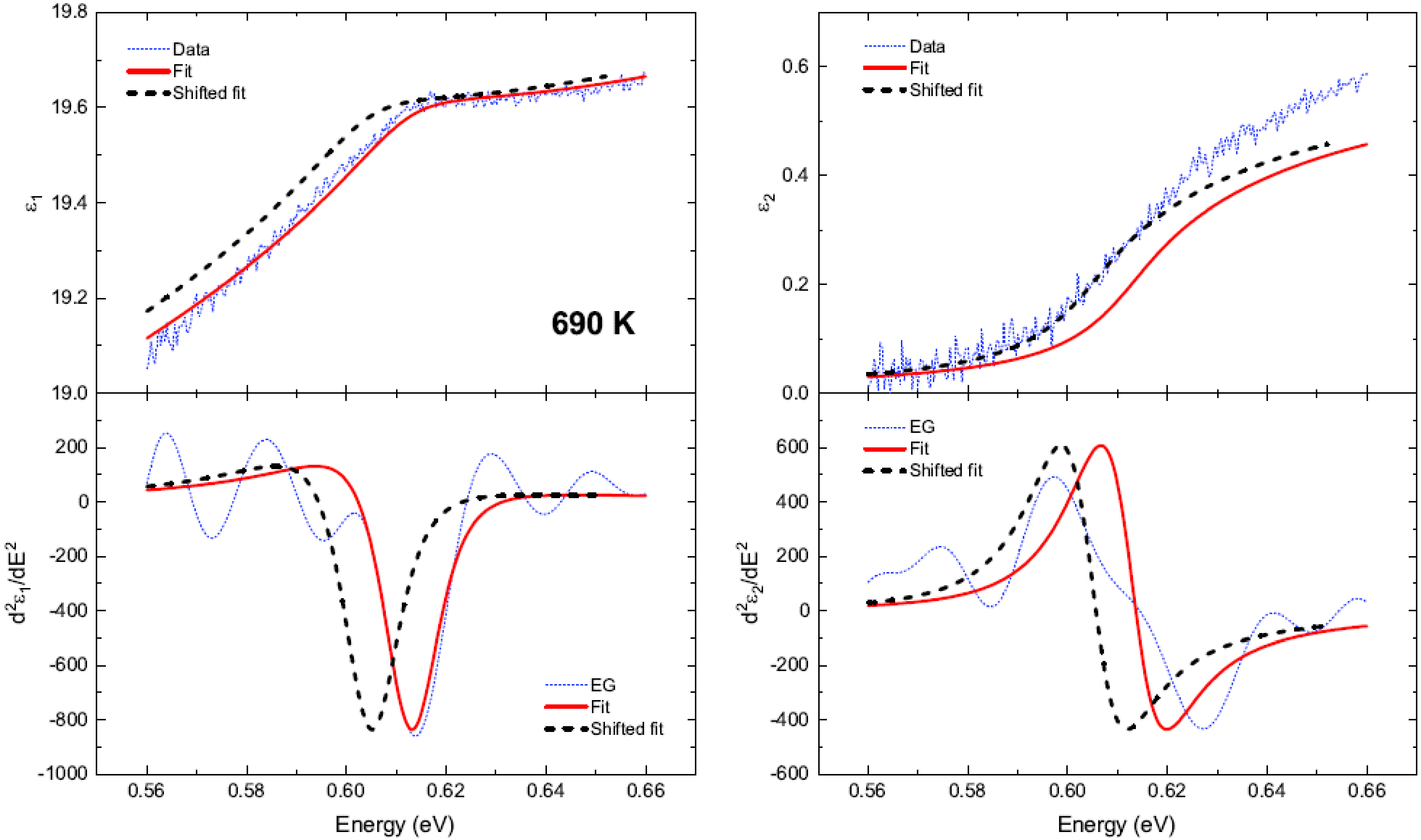








J. Menéndez, D.J. Lockwood, J.C. Zwinkels, and M. Noël, Phys. Rev. B **98**, 165207 (2018).
J. Menéndez, D.C. Poweleit, and S.E. Tilton, Phys. Rev. B **101**, 195204 (2020).
G.G. Macfarlane, T.P. McLean, J.E. Quarrington, and V. Roberts, Phys. Rev. **108**, 1377 (1957).


Data
Fit
Shifted fit
690 K
ε1
d²ε1/dE²
EG
ε2
d²ε2/dE²
Energy (eV)
0.56
0.58
0.60
0.62
0.64
0.66

Broadening theory

$$\eta_c(\epsilon) = \eta_c^+(\epsilon) + \eta_c^-(\epsilon)$$

Conwell's expression
(LA-phonon intervalley scattering)

$$\eta_c^\pm(\epsilon)$$

$$= \frac{2}{\sqrt{2}\pi\rho}\left(\frac{D_{\mathrm{LA}}^2}{\hbar^2\Omega_{\mathrm{LA}}}\right) m_\perp\sqrt{m_\parallel}\left(n_{\mathrm{LA}} + \frac{1}{2} \pm \frac{1}{2}\right)\sqrt{E_0 + \epsilon - E_{\mathrm{ind}} \mp \hbar\Omega_{\mathrm{LA}}}$$

$$+ \frac{2\sqrt{2}}{3\pi\rho}\left(\frac{d_{\mathrm{LO}}'^2}{\hbar^4\Omega_{\mathrm{LO}}}\right) m_\perp m_\parallel^{\frac{3}{2}}\left(n_{\mathrm{LO}} + \frac{1}{2} \pm \frac{1}{2}\right)(E_0 + \epsilon - E_{\mathrm{ind}} \mp \hbar\Omega_{\mathrm{LO}})^{3/2}$$

→ Forbidden LO scattering from "around L to Γ"

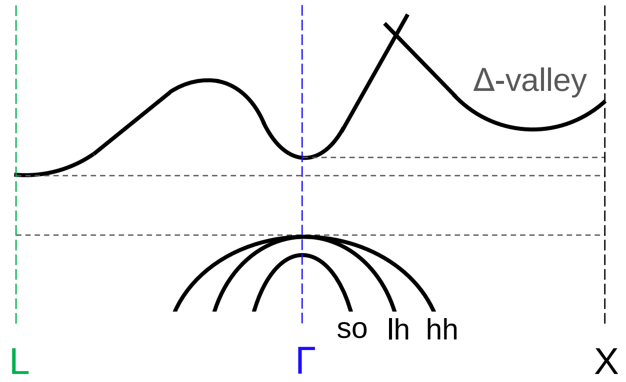
$$+ \frac{4\sqrt{2}}{3\pi\rho}\left(\frac{d_{\mathrm{TA}}'^2}{\hbar^4\Omega_{\mathrm{TA}}}\right) m_\perp m_\parallel^{\frac{3}{2}}\left(n_{\mathrm{TA}} + \frac{1}{2} \pm \frac{1}{2}\right)(E_0 + \epsilon - E_{\mathrm{ind}} \mp \hbar\Omega_{\mathrm{TA}})^{3/2}$$

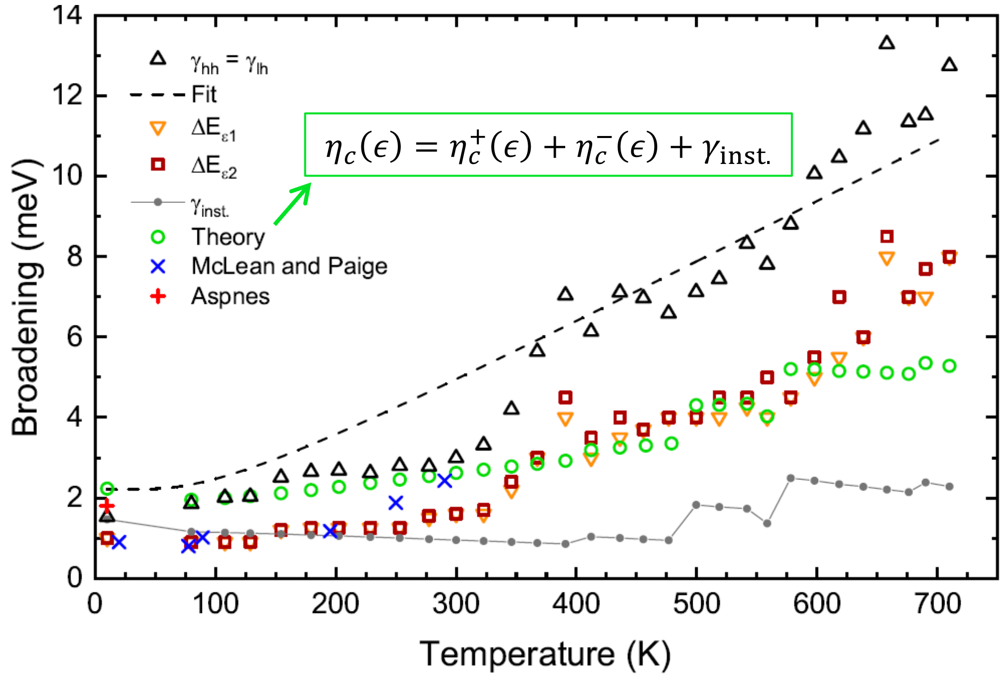
$$+ \frac{2\sqrt{2}}{\pi\rho}\left(\frac{d_{\mathrm{TA}}'^2}{\hbar^4\Omega_{\mathrm{TA}}}\right) m_c m_\perp\sqrt{m_\parallel}\left(n_{\mathrm{TA}} + \frac{1}{2} \pm \frac{1}{2}\right)\epsilon\sqrt{E_0 + \epsilon - E_{\mathrm{ind}} \mp \hbar\Omega_{\mathrm{TA}}}$$

Forbidden TA scattering

$$+ \frac{2}{\sqrt{2}\pi\rho}\left(\frac{D_{\Delta\Gamma}^2}{\hbar^2\Omega_\Delta}\right) m_\Delta^{\frac{3}{2}}\left(n_\Delta + \frac{1}{2} \pm \frac{1}{2}\right)\sqrt{E_0 + \epsilon - E_\Delta \mp \hbar\Omega_\Delta}$$

= 0, because conduction band minimum at Γ is below the Δ minimum

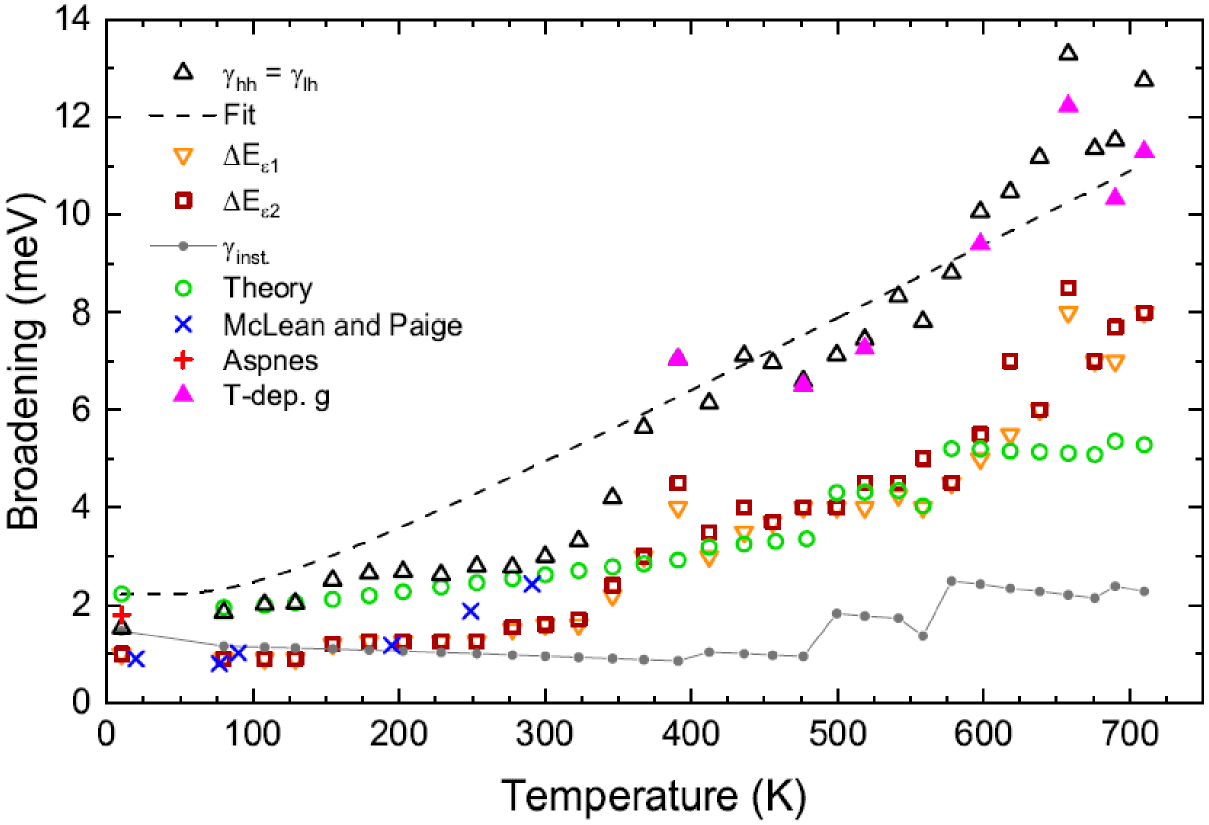


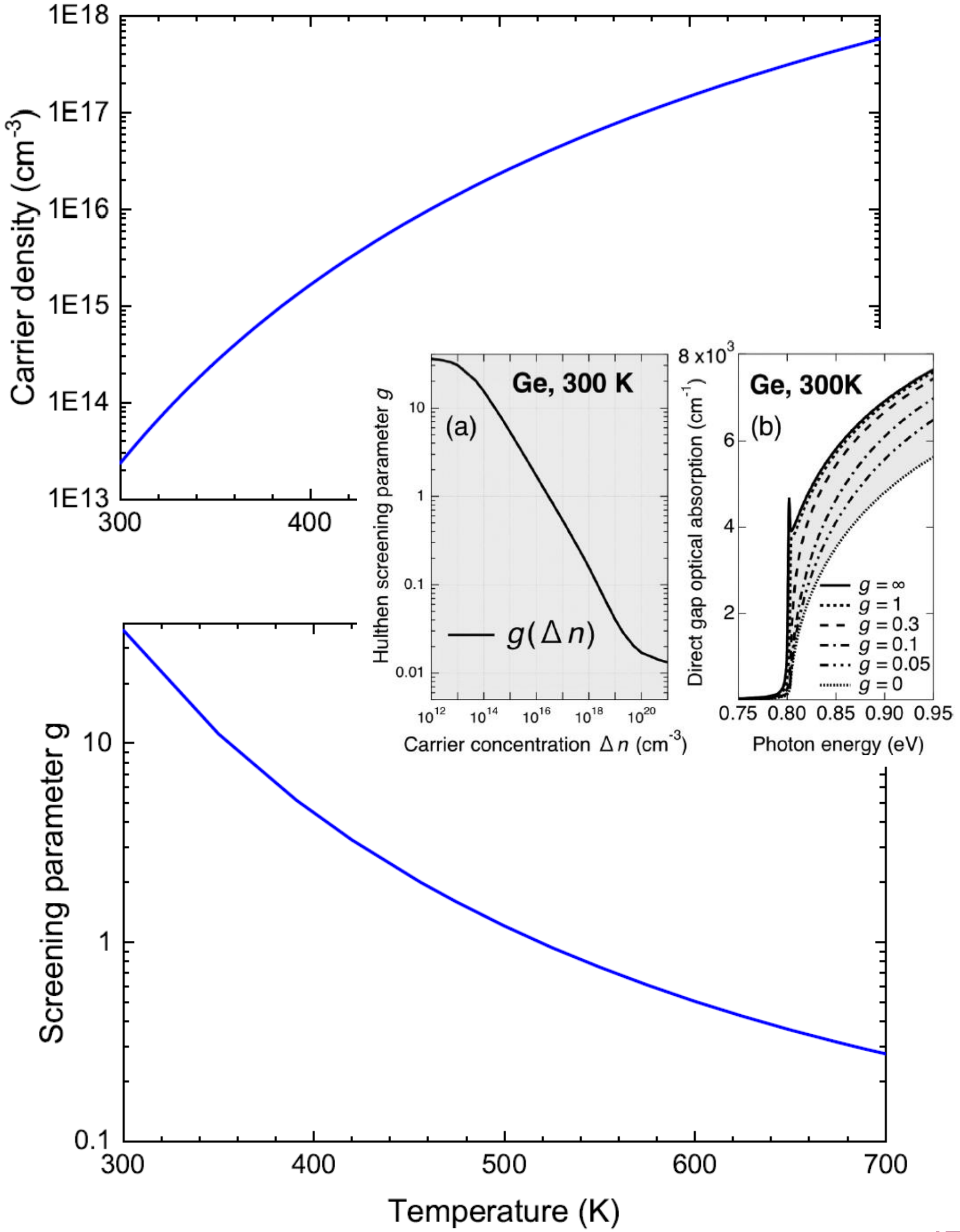


J. Menéndez, C. D. Poweleit, and S. E. Tilton, Phys. Rev. B **101**, 195204 (2020).
E. M. Conwell, *High Field Transport in Semiconductors* (Academic, New York, 1967).

Screening

Considering the temperature dependence of the screening parameter affects the broadening at higher temperatures (▲):





J. Menéndez, C. D. Poweleit, and S. E. Tilton, PRB **101**, 195204 (2020)

$$A = 1 - 1/3\left[\frac{E_P}{E_0} + \frac{2E_Q}{E_0'}\right]$$

$$B = 1/3\left[-\frac{E_P}{E_0} + \frac{E_Q}{E_0'}\right]$$

$$C^2 = \frac{4E_PE_Q}{3E_0E_0'} + \Delta$$

DKK parameters and effective masses

M. Cardona, J. Phys. Chem. Solids **24**, 1543 (1963):

$$A = \tfrac{1}{3}(F+2G+2M)+1$$
$$B = \tfrac{1}{3}(F+2G-M)$$
$$C^2 = \tfrac{1}{3}[(F-G+M)^2-(F+2G-M)^2]$$

$$F(T) = -\frac{E_{P,4\mathrm{K}}\left(\frac{a_{4\mathrm{K}}}{a(T)}\right)}{E_0(T)}$$

$$M(T) = -\frac{E_{Q,4\mathrm{K}}\left(\frac{a_{4\mathrm{K}}}{a(T)}\right)}{E_0'(T)}$$

$$G(T) = -G_{4\mathrm{K}}\left(\frac{a_{4\mathrm{K}}}{a(T)}\right)$$

For A = -13.38, B = -8.48, and |C| = 13.14 at 4 K (also used in J. Menendez et al., Phys. Rev. B **98**, 165207 (2018)):

$$E_{P,4\mathrm{K}} = 26.0\ \mathrm{eV}$$
$$E_{Q,4\mathrm{K}} = 18.5\ \mathrm{eV}$$
$$G_{4\mathrm{K}} = -1.04$$

$$m_{hh,4\mathrm{K}} = 0.326\ m_0$$
$$m_{lh,4\mathrm{K}} = 0.0422\ m_0$$

Generation of strain pulse

Thomsen *et al.* (1986): $\sigma_{33} = -B\frac{\partial E_g}{\partial P}N - \frac{3B\beta}{c}(E - E_g)N$

Wright *et al.* (2002): $\sigma_{33} = -B\frac{\partial E_g}{\partial P}N - 3B\beta\Delta T$

$$\epsilon_{ij} = \sum_{kl} S_{ijkl}\sigma_{kl}$$

$$\hat{S} = \begin{pmatrix} S_{11} & S_{12} & S_{12} & 0 & 0 & 0 \\ S_{21} & S_{11} & S_{12} & 0 & 0 & 0 \\ S_{21} & S_{21} & S_{11} & 0 & 0 & 0 \\ 0 & 0 & 0 & S_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & S_{44} & 0 \\ 0 & 0 & 0 & 0 & 0 & S_{44} \end{pmatrix}$$

$$\epsilon_3 = S_{21}(\sigma_1 + \sigma_2) + S_{11}\sigma_3$$

$$\sigma_1 = \sigma_2 = \sigma_3 = \sigma$$

$$\epsilon_3 = (S_{11} + 2S_{12})\sigma$$

$$S_{11} + 2S_{12} = \frac{1}{C_{11} + 2C_{12}} = 0.444\ \frac{\mathrm{cm}^2}{\mathrm{N}}$$

$$\epsilon_3 = \left(4.44 \times 10^{-8}\ \frac{\mathrm{cm}^2}{\mathrm{N}}\right)\sigma.$$

Calculating strain and energy shifts (Ge 800nm pump)

$\frac{\partial E_g}{\partial P} = 5\ \mathrm{eV}$ (indirect gap)

Stress-strain relation: $\epsilon_3 = (S_{11} + 2S_{12})\sigma = \frac{\sigma}{C_{11}+2C_{12}}$ (assuming: $\sigma_{ii} = \sigma$ for all i and $\sigma_{ij} = 0$ for $i \neq j$)

$(C_{11} + 2C_{12})^{-1} = 4.44 \times 10^{-8}\ \frac{\mathrm{cm}^2}{\mathrm{N}}$

Electron contribution: $\sigma_{\mathrm{el}} = -B\frac{\partial E_g}{\partial P}N$ => $\epsilon_{\mathrm{el}} = (C_{11} + 2C_{12})^{-1}\sigma_{\mathrm{el}} = -6.4 \times 10^{-4}$

Phonon contribution: $\sigma_{\mathrm{ph}} = -\frac{3B\beta}{c}(E - E_g)N$ => $\epsilon_{\mathrm{ph}} = (C_{11} + 2C_{12})^{-1}\sigma_{\mathrm{ph}} = -1.2 \times 10^{-4}$

Total stress/strain: $\sigma_{33} = \sigma_{\mathrm{el}} + \sigma_{\mathrm{ph}}$ => $\epsilon_{33} = (C_{11} + 2C_{12})^{-1}\sigma_{33} = -7.6 \times 10^{-4}$

In-plane and out-of-plane strain: $\epsilon_{\perp} = \epsilon_{33}$ and $\epsilon_{\parallel} = 0$

Hydrostatic and shear strain: $\epsilon_H = \epsilon_S = 2.5 \times 10^{-4}$

Hydrostatic shift: $\Delta E_H = \sqrt{3}D_1^1\epsilon_H = -3.4\ \mathrm{meV}$

Shear splitting: $\Delta E_S = \sqrt{6}D_3^3\epsilon_S = 1.6\ \mathrm{meV}$

Critical point energy shift: $\Delta E_1 = \frac{\Delta_1}{2} + \Delta E_H - \sqrt{\frac{(\Delta_1)^2}{4} + (\Delta E_S)^2} = -3.4\ \mathrm{meV}$

$$\Delta(E_1 + \Delta_1) = -\frac{\Delta_1}{2} + \Delta E_H + \sqrt{\frac{(\Delta_1)^2}{4} + (\Delta E_S)^2} = -3.4\ \mathrm{meV}$$

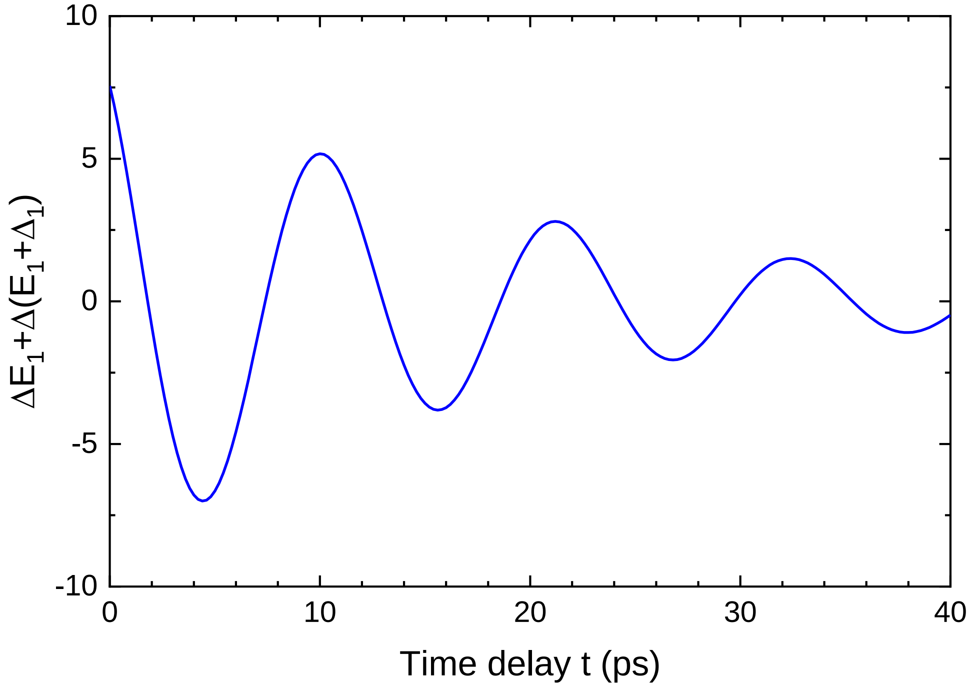
- Hydrostatic strain: $\epsilon_H = \frac{\epsilon_\perp + 2\epsilon_\parallel}{3}$
- Shear strain: $\epsilon_S = \frac{\epsilon_\perp - \epsilon_\parallel}{3}$
- Hydrostatic shift: $\Delta E_H = \sqrt{3} D_1^1 \epsilon_H$
- Shear splitting: $\Delta E_S = \sqrt{6} D_3^3 \epsilon_S$
- Shift: $\Delta E_1 = E_1^s - E_1^0 = \frac{\Delta_1}{2} + \Delta E_H - \sqrt{\frac{(\Delta_1)^2}{4} + (\Delta E_S)^2}$

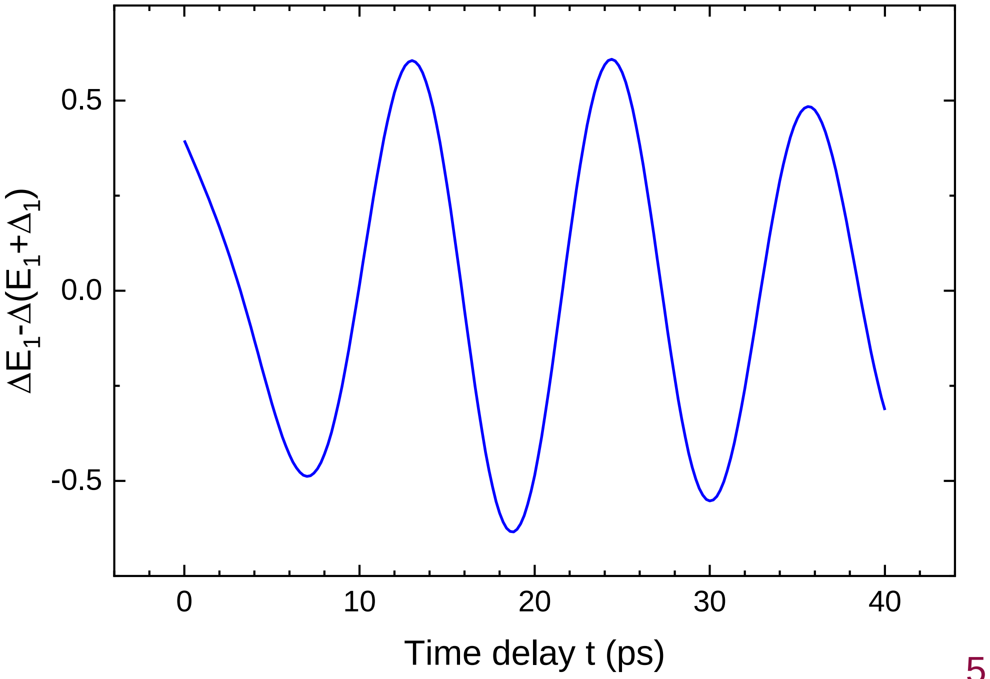
$$\Delta(E_1 + \Delta_1) = (E_1 + \Delta_1)^s - (E_1^0 + \Delta_1) = -\frac{\Delta_1}{2} + \Delta E_H + \sqrt{\frac{(\Delta_1)^2}{4} + (\Delta E_S)^2}$$

- **Adding energy shifts (data):**
  $\Delta E_1 + \Delta(E_1 + \Delta_1) = 2\Delta E_H \approx 9 \text{ meV}$
  => $\Delta E_H \approx 4.5 \text{ meV}$
  => $\epsilon_H = \frac{\Delta E_H}{\sqrt{3} D_1^1} \approx -3.3 \times 10^{-4}$
  => $\epsilon_\perp = 3\epsilon_H \approx -1 \times 10^{-3}$
- **Subtracting energy shifts (data):**

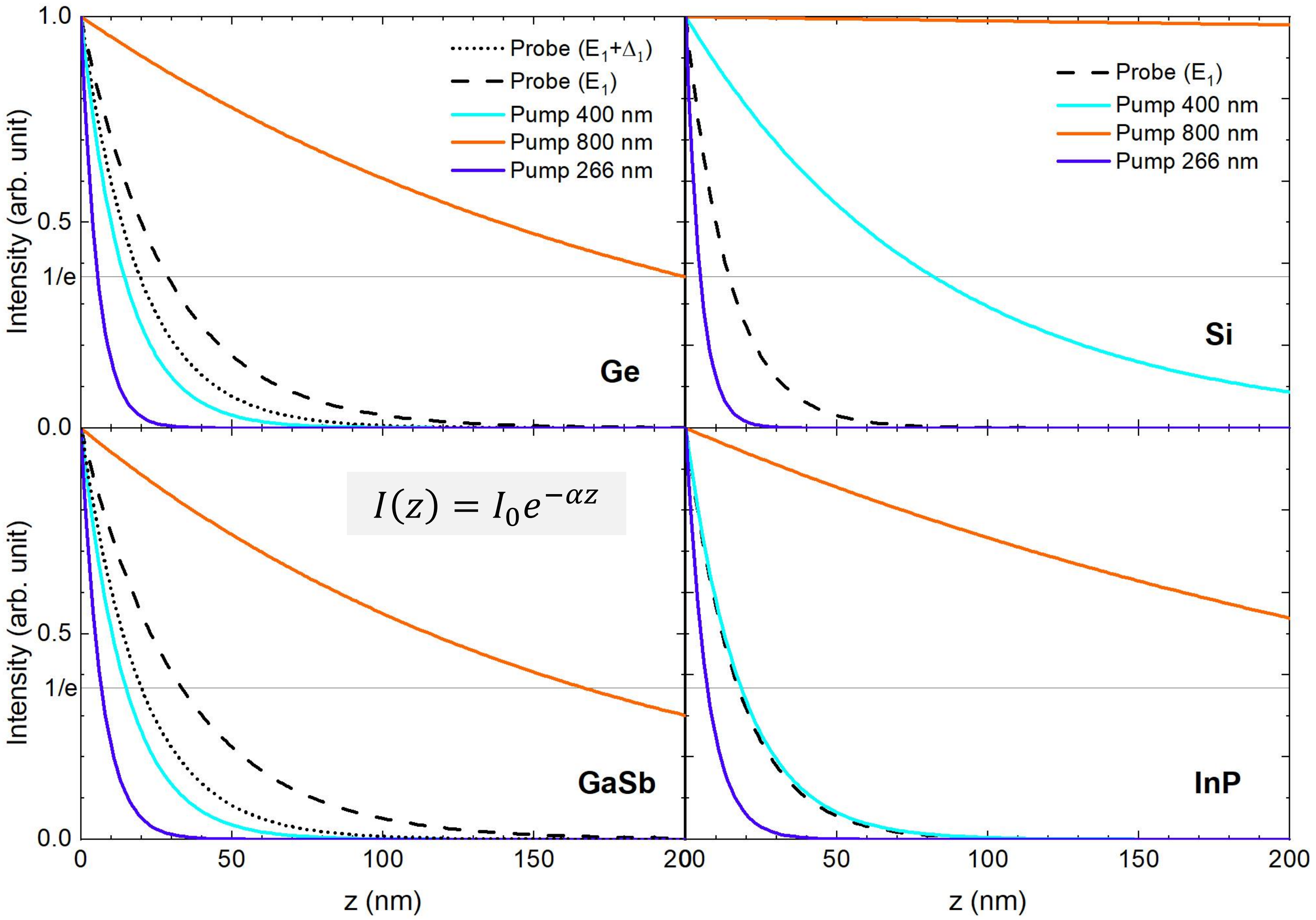
$$|\Delta E_1^s - \Delta(E_1 + \Delta_1)^s| = \left|\Delta_1 - 2\sqrt{\frac{(\Delta_1)^2}{4} + (\Delta E_S)^2}\right| \approx 0.5 \text{ meV}$$

  => $|\Delta E_S| \approx 7.0 \text{ meV}$
  => $|\epsilon_S| = \frac{|\Delta E_S|}{\sqrt{6} D_3^3} \approx 1.1 \times 10^{-3}$
  => $|\epsilon_\perp| = 3|\epsilon_S| \approx 3.3 \times 10^{-3}$



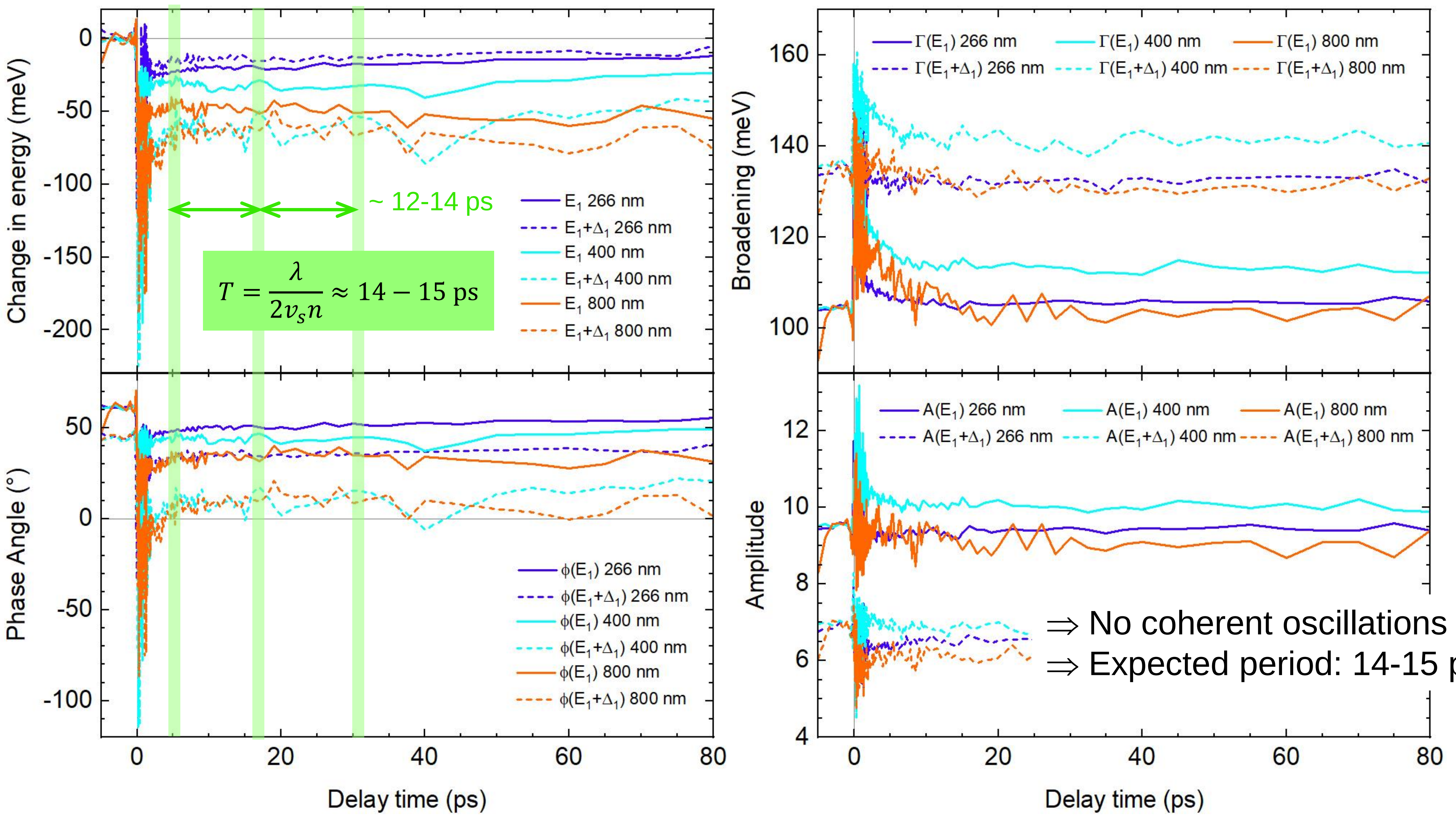


Beer's law and penetration depth for different pump wavelengths



$$I(z) = I_0 e^{-\alpha z}$$

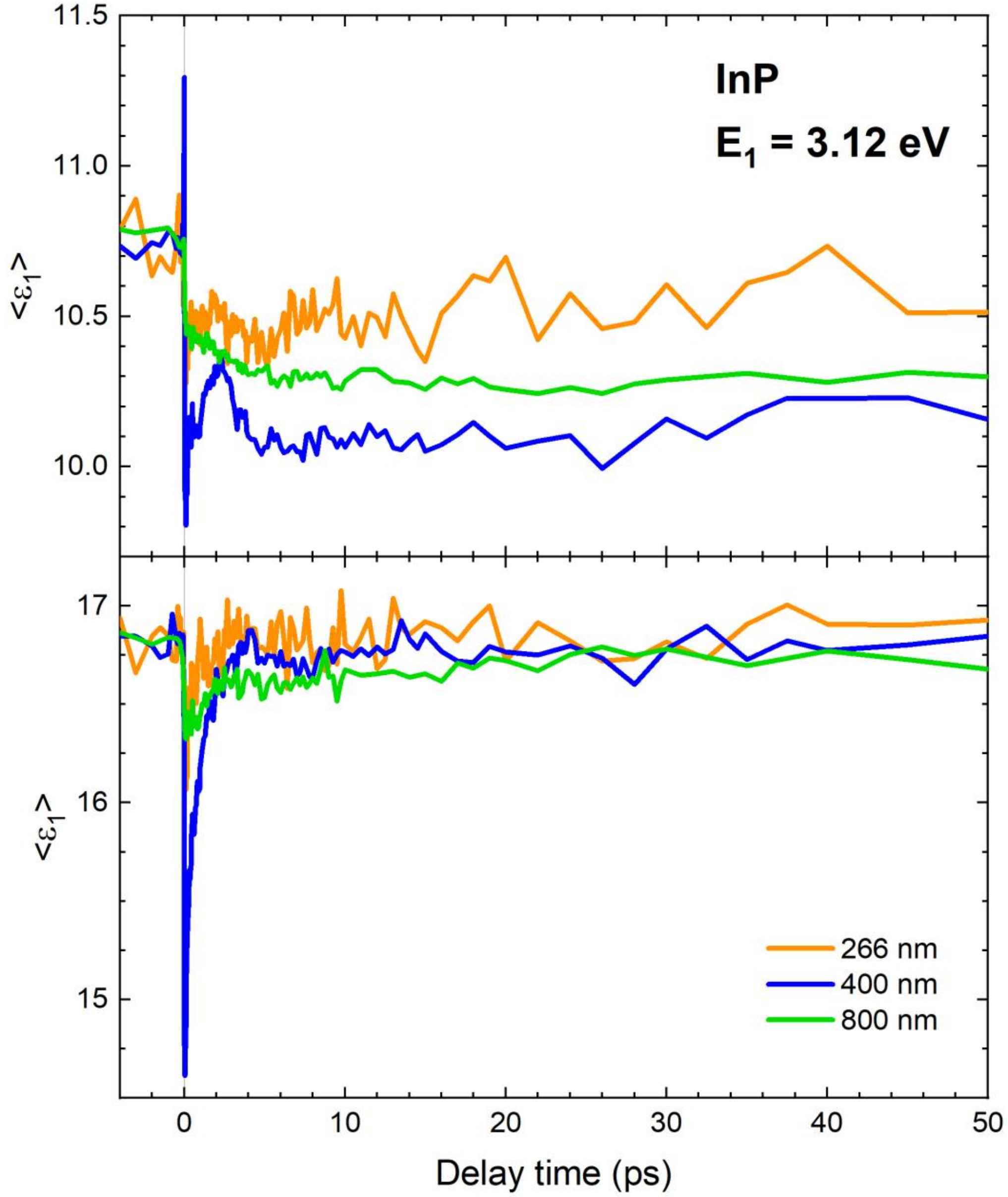
Critical point parameters of GaSb (266, 400, and 800 nm pump)

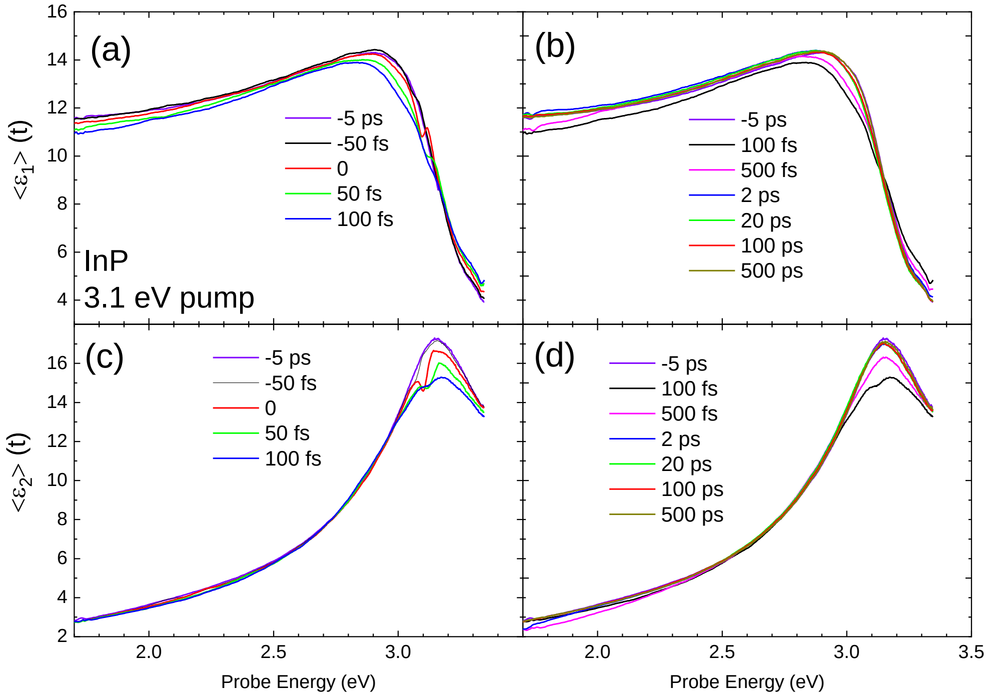


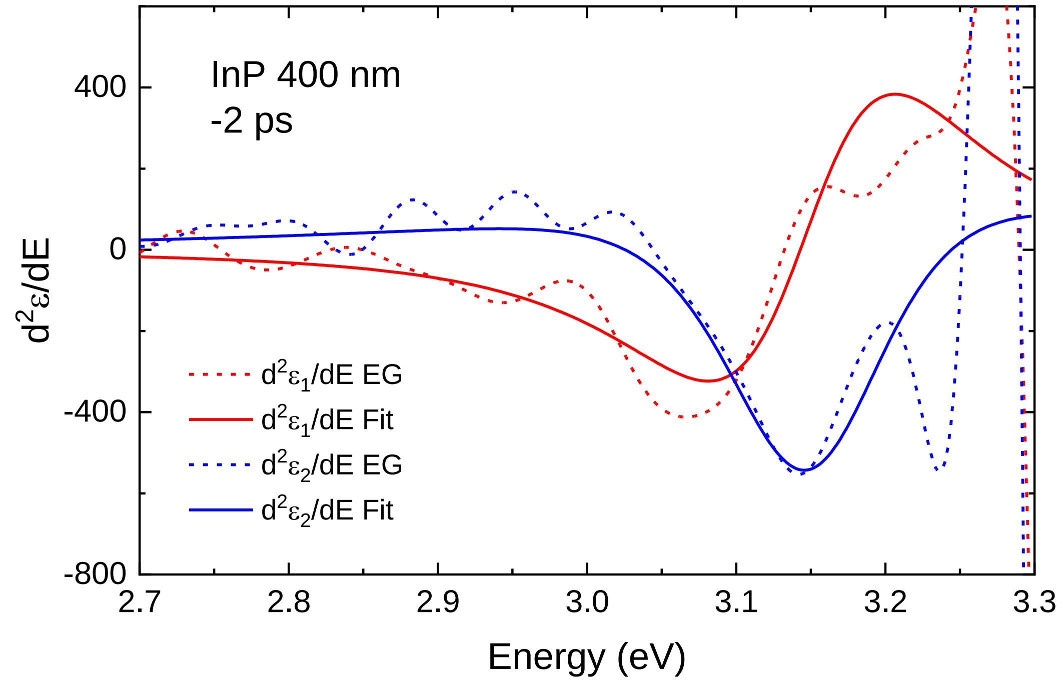
$\Rightarrow$ No coherent oscillations detected

$\Rightarrow$ Expected period: 14-15 ps

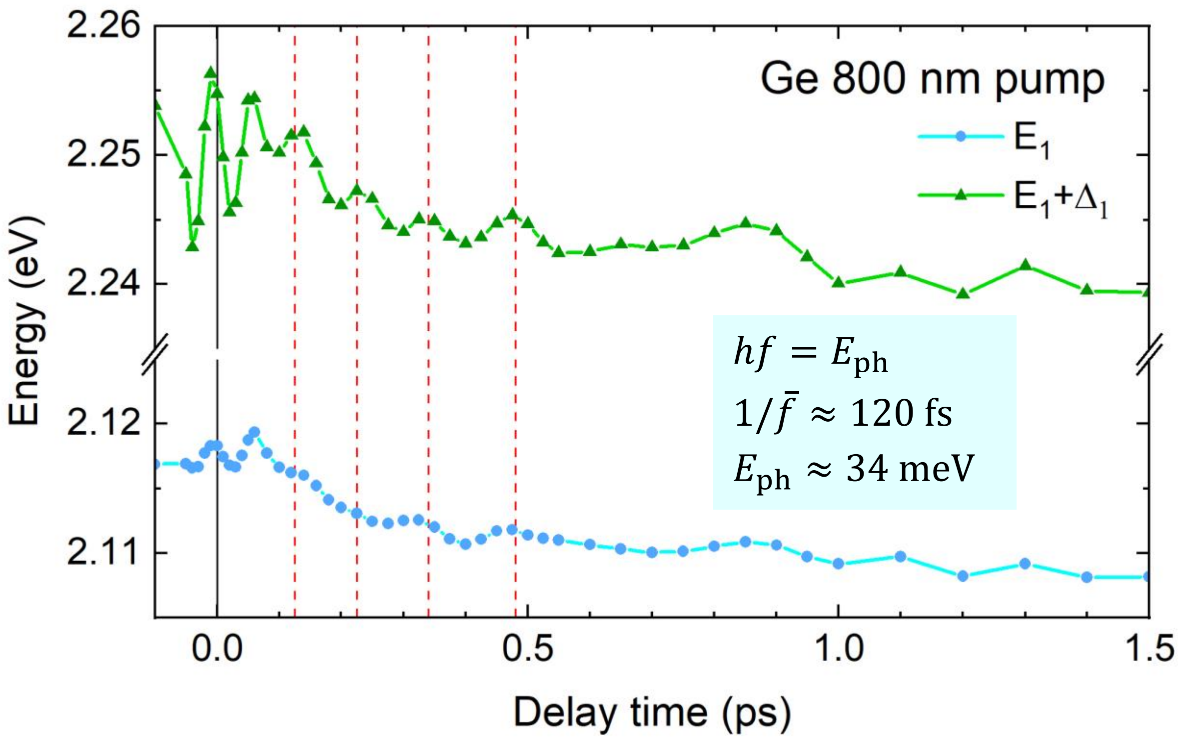
Problem with InP: Data only up to 3.34 eV

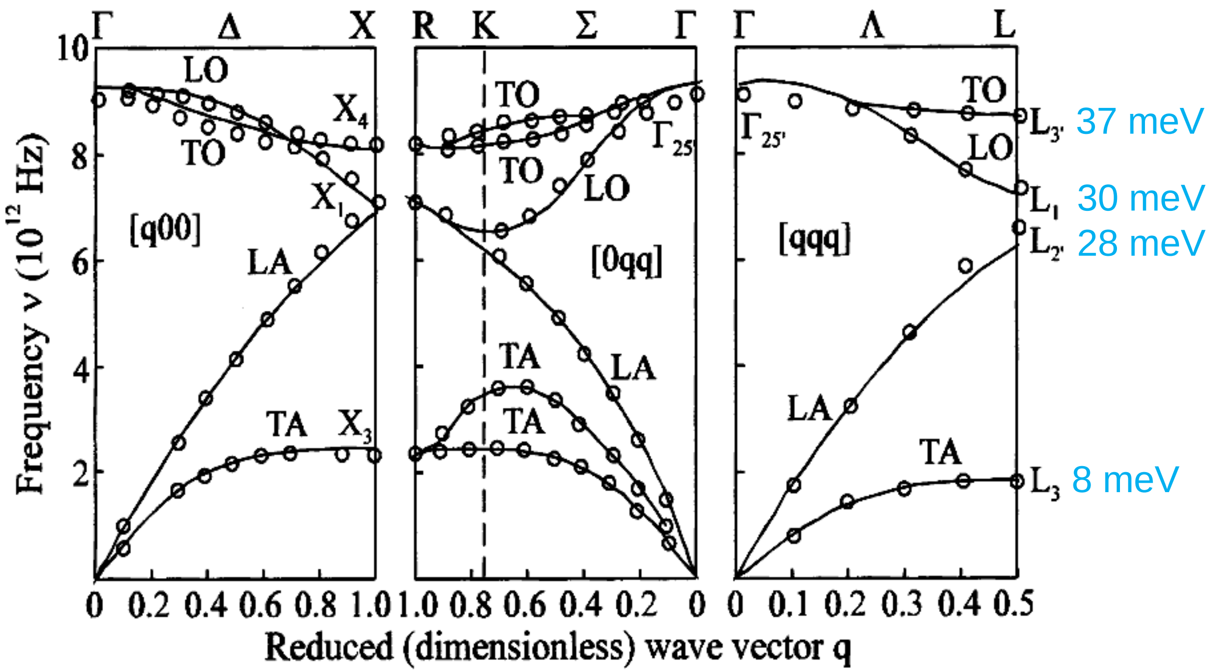






Coherent optical phonon oscillations?

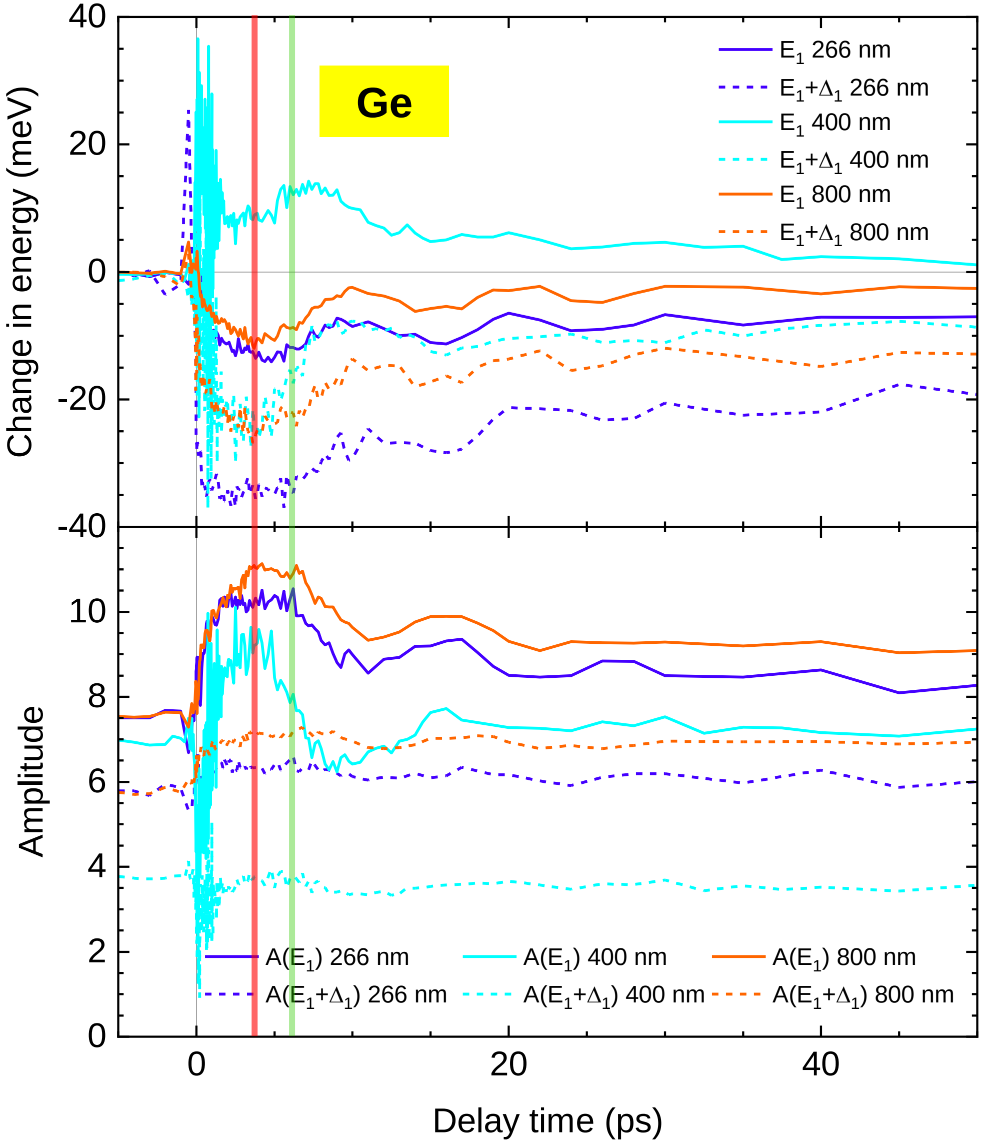




Weber W., Phys. Rev. **B15,** 10 (1977) 4789-4803.

- Are these oscillations due to coherent optical phonons?
- Period about 100-150 fs ($E_{\mathrm{ph}} \approx 28 - 40$ meV)
- The temporal bandwidth is ~120 fs (limited by oblique incidence and probe spot size)

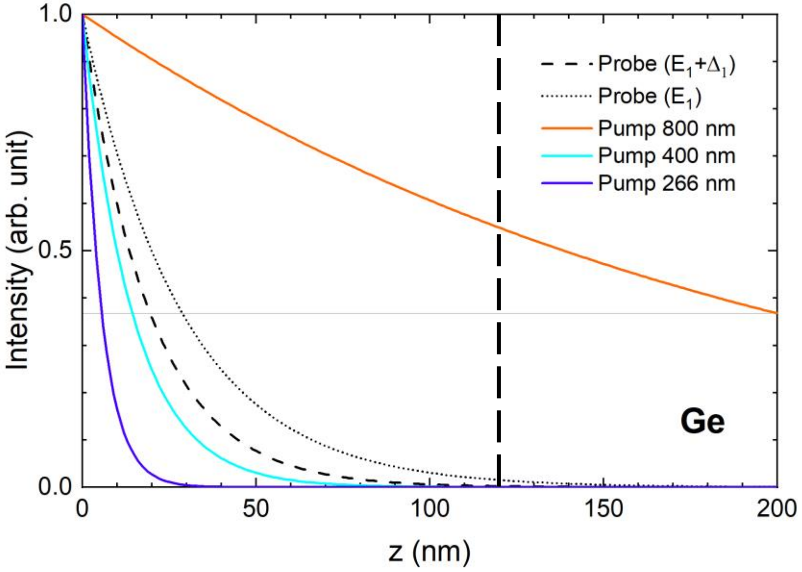
Propagation of strain pulses in Ge



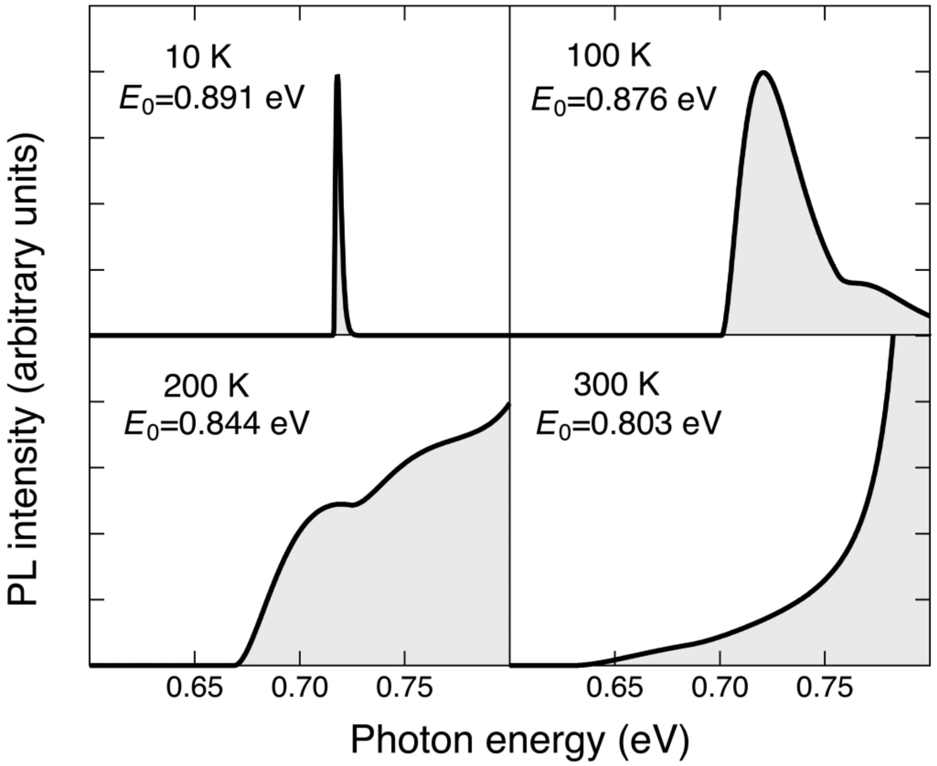
$$t = \frac{d}{v_s} = \frac{18 \text{ nm}}{4.87 \text{ nm/ps}} \approx 4 \text{ ps}$$

$$t = \frac{d}{v_s} = \frac{29 \text{ nm}}{4.87 \text{ nm/ps}} \approx 6 \text{ ps}$$

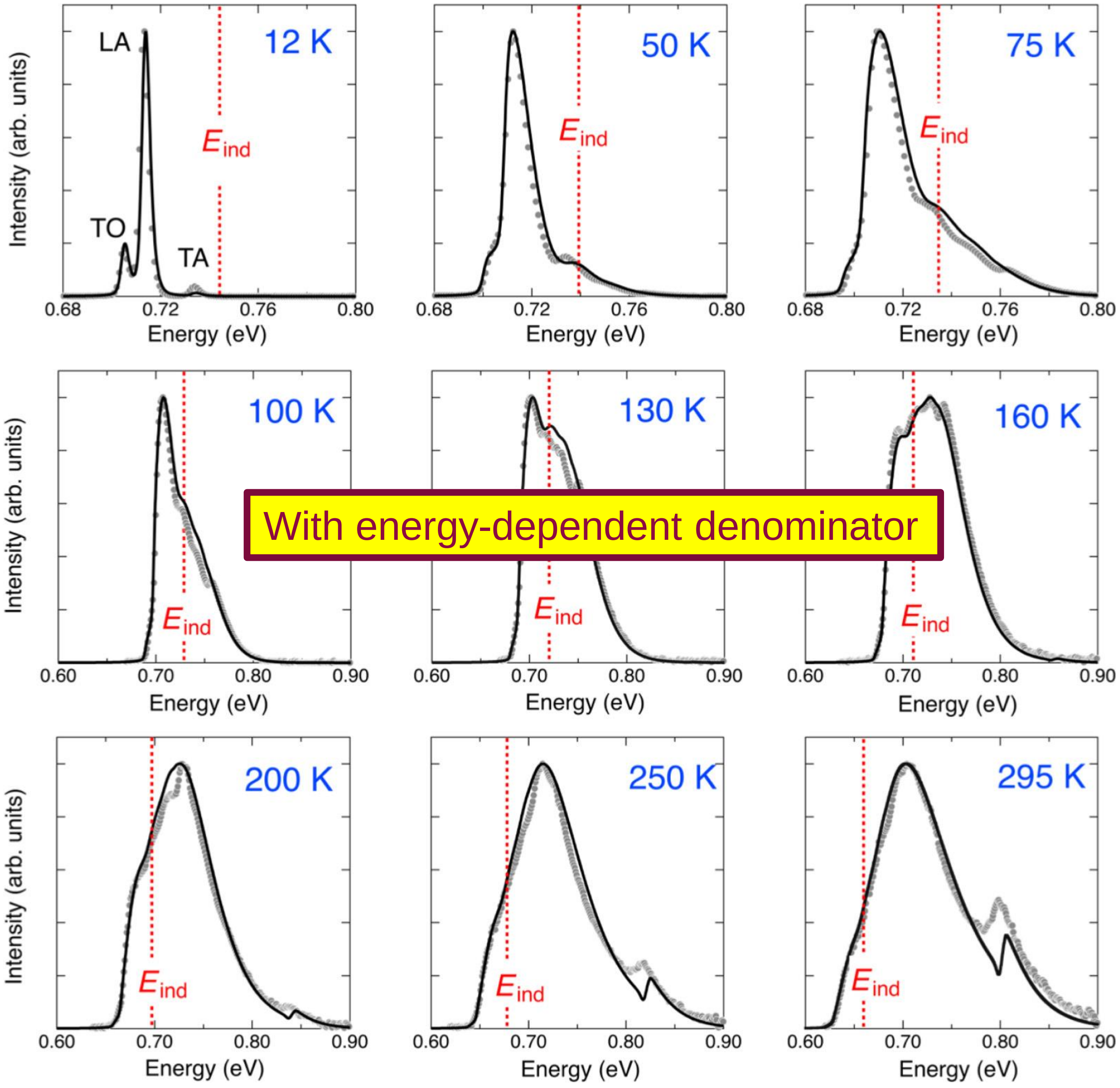
$$d = 25 \text{ ps} \cdot 4.87 \frac{\text{nm}}{\text{ps}} \approx 120 \text{ nm}$$



Temperature-dependent photoluminescence in Ge



Conventional theory:
W. Van Roosbroeck and W. Shockley,
PR **94**, 1558 (1954).



With energy-dependent denominator

Menéndez *et al.*

J. Menéndez, C. D. Poweleit, and S. E. Tilton, Phys. Rev. B **101**, 195204 (2020)