Raman scattering at terahertz frequencies enabled by an infrared free electron laser

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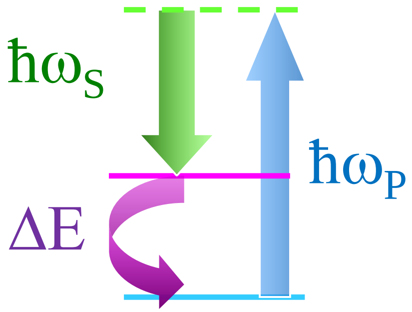
Long-wavelength (THz) Raman scattering

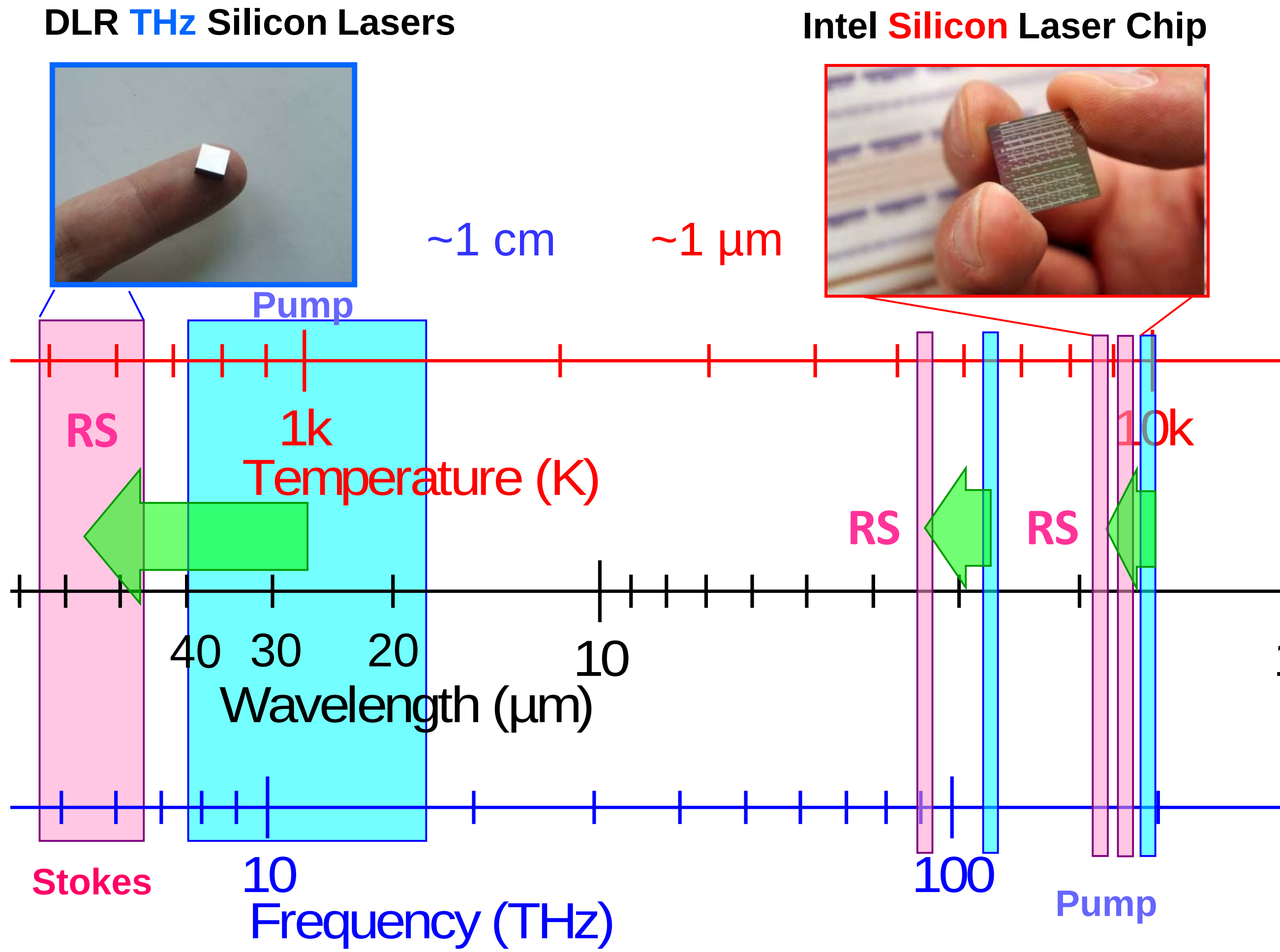


Si: 2.88 → 3.39 µm
Opt. Exp. **15**, 14355 (2007)

Ge: 5.62 → 7.60 µm
APL **102**, 011111 (2013)

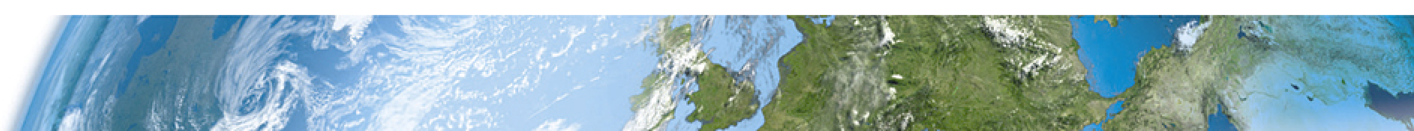
H_2: 10.6 → 16.9 µm
Opt. Lett. **3**, 144 (1978)

n-Si:
18-40µm → 47-70µm
PRL **96**, 037404 (2006)



THz Raman scattering: Challenges

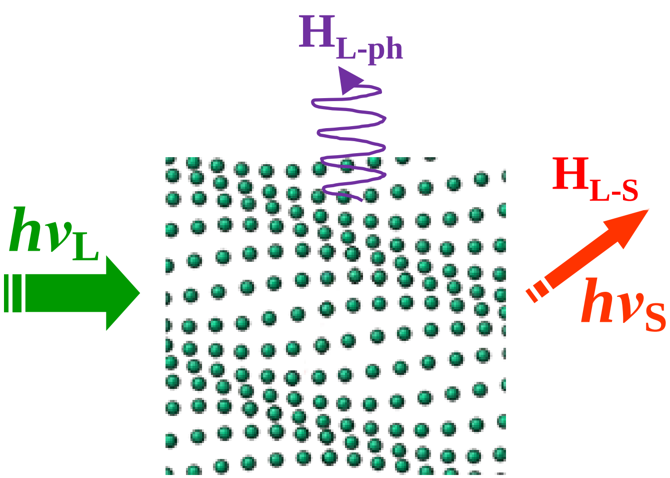
> scattering efficiency, λ^{-4}
> either free carrier absorption, λ^{2}
or excitation avoiding free electrons ?
> under optical phonon excitation
= high orders of electron-phonon interaction
> technical (filters, collecting optics)
= no commercial THz components
(notch or low pass filter, lens objective)



Interactions resulting in the Raman light scattering

(ionic Raman)

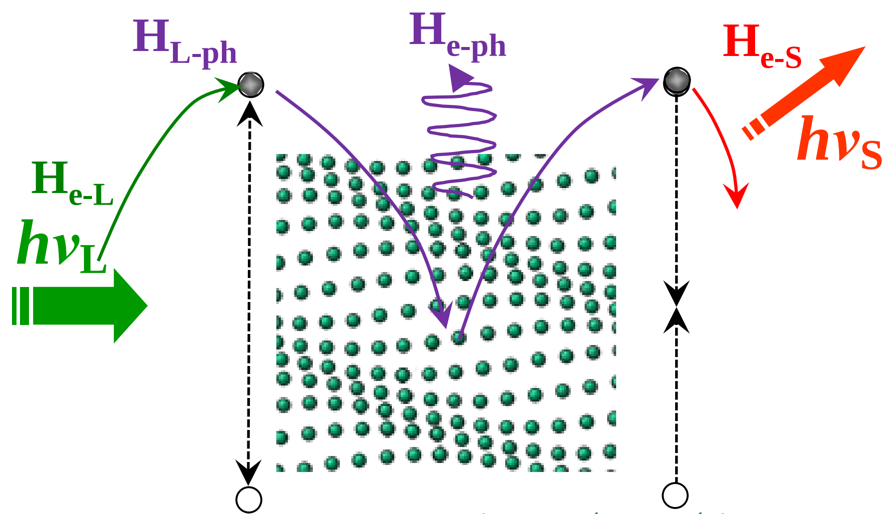
$\text{photon} \xrightarrow{|VS1\rangle} \text{RA phonon} \xrightarrow{|VS2\rangle} \text{RS photon}$



lattice anharmonicities
10^{-11}

(electronic Raman)

$\text{photon} \xrightarrow{|VS1\rangle} \text{electronic} \xrightarrow{|VS2\rangle} \text{RS photon}$



nanotherm.es/images/phonon2.png

electron-phonon interactions
zero-order 10^{-5}-10^{-7} (PRB **6**, 3886)

Interactions resulting in the Raman light scattering

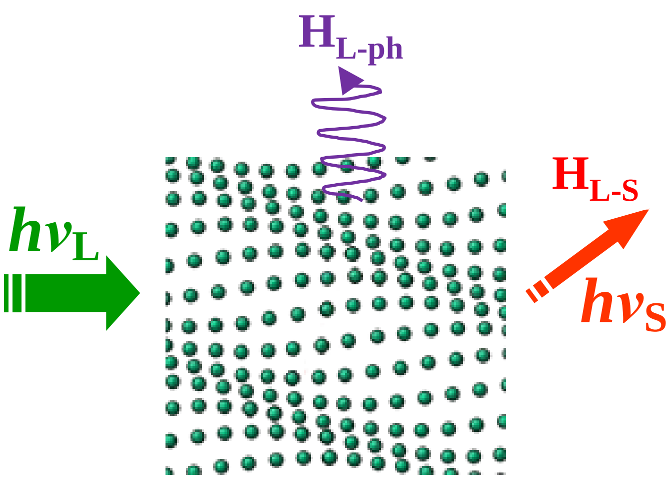
(ionic Raman)

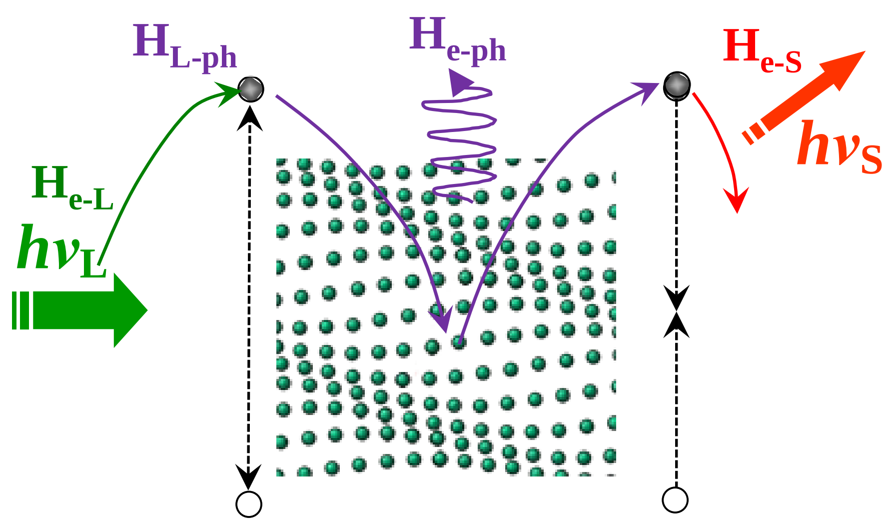
free electron – free

(electronic Raman)

free electron (e-h pair) – assisted

free carrier absorption $n\lambda^2$





lattice anharmonicities

electron-phonon interactions

nanotherm.es/images/phonon2.png

Interaction orders (N-phonon(s) scattering)

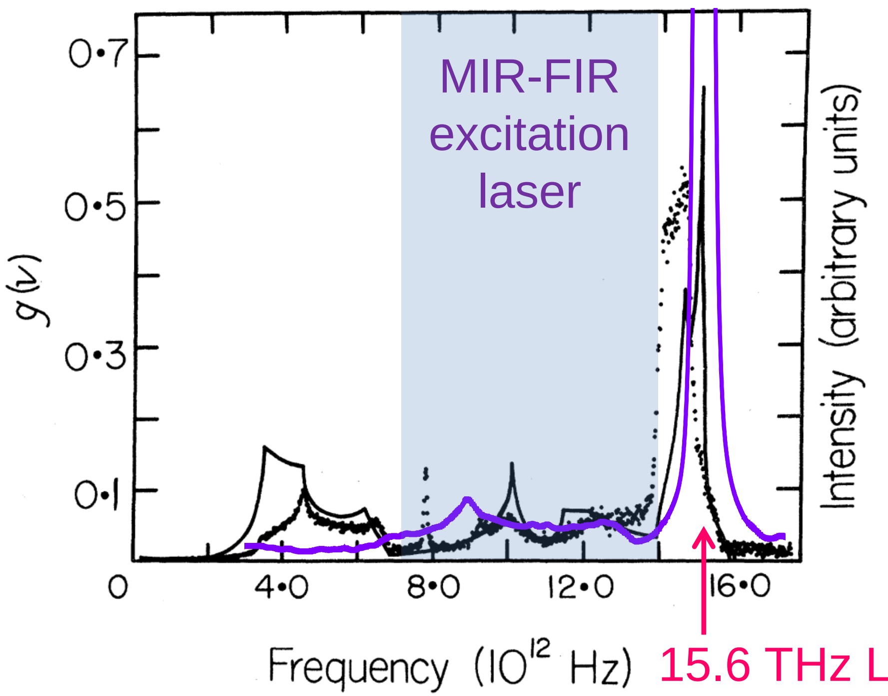


FIG. 10. The superposition of the density of states calculated by Dolling and Cowley (Ref. 9) and our two-phonon Raman data as measured at 305 °K and modified as discussed in the text.

VIS-NIR excitation laser

zero-order 10^{-5}-10^{-7}
(PRB **6**, 3886)

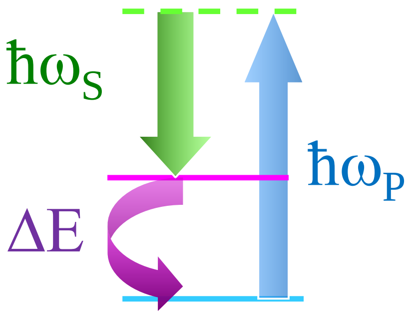
Multiphonon Raman spectrum of silicon
(Phy Rev B 7, 3685 (1973))
Second order Raman spectrum and phonon density of states of silicon
(Phys Lett 44A, 517 (1973))

Key notes:

enabling THz Raman scattering in doped silicon

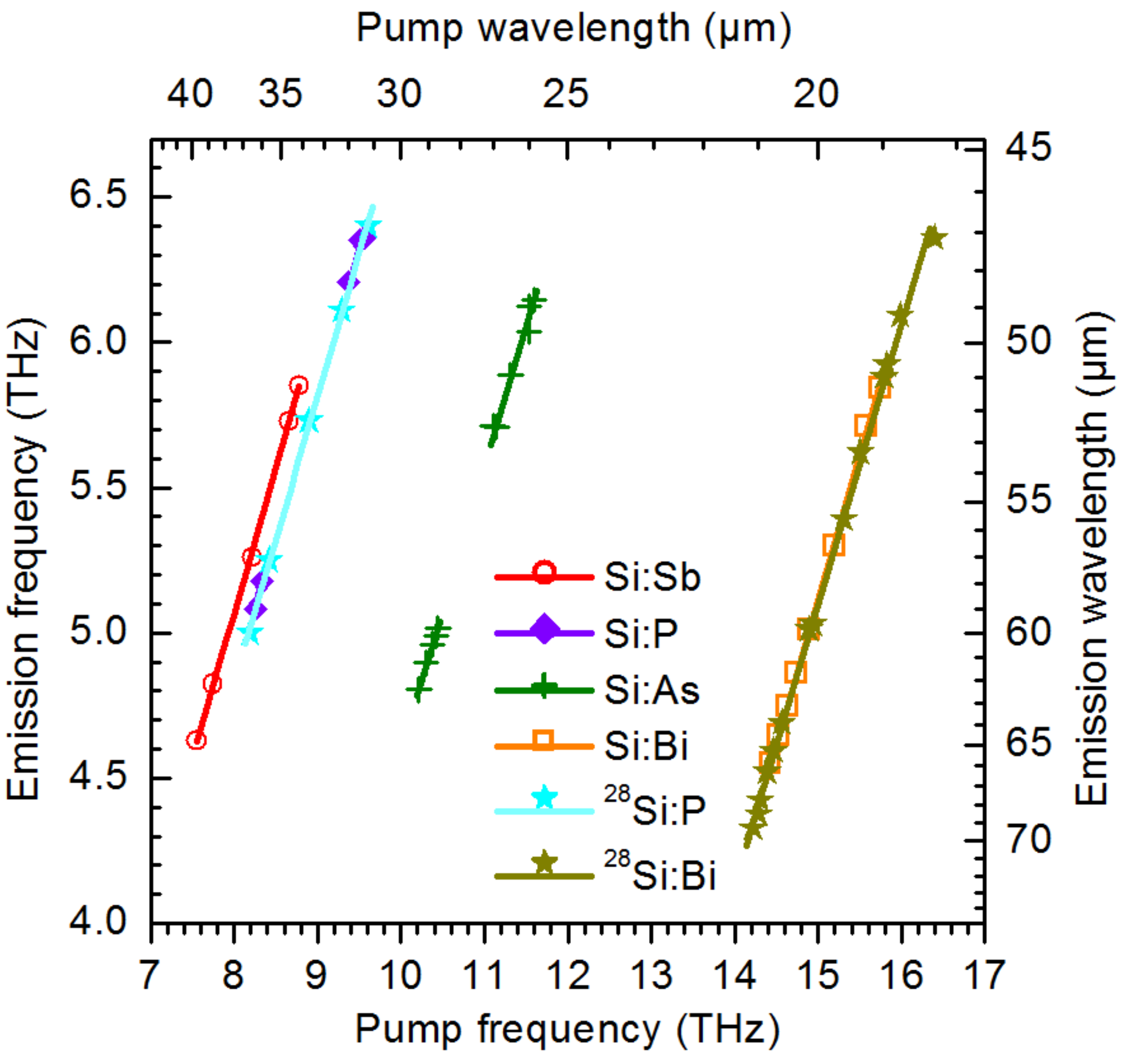
features of THz Raman scattering in doped silicon

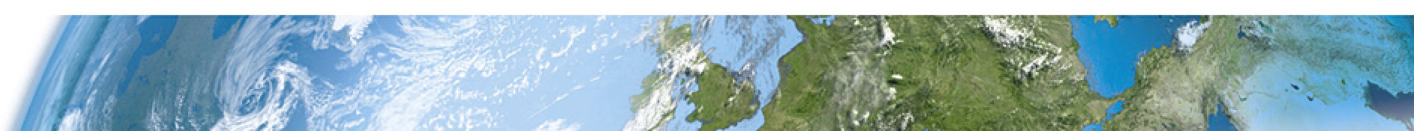
THz intracenter Raman lasing from a doped Si.



$$\hbar\omega_S = \hbar\omega_P - \Delta E,$$

PRL **96,** 037404 (2006)
APL **92**, 091111 (2008)
APL **94**, 171112 (2009)
APL **95**, 201110 (2009)
Phys B **404**, 4661 (2009)





Experimental

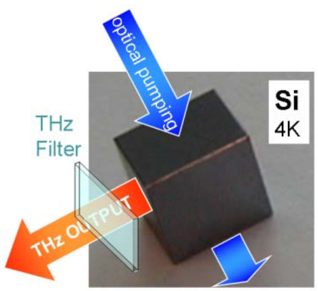
Float-zone grown

^{nat}Si: P, Sb, As, Bi

^{28}Si: P, Bi

^{nat}Si: Sb+P,

$N_D \sim (2\text{-}14)\times10^{15}$ cm^{-3}



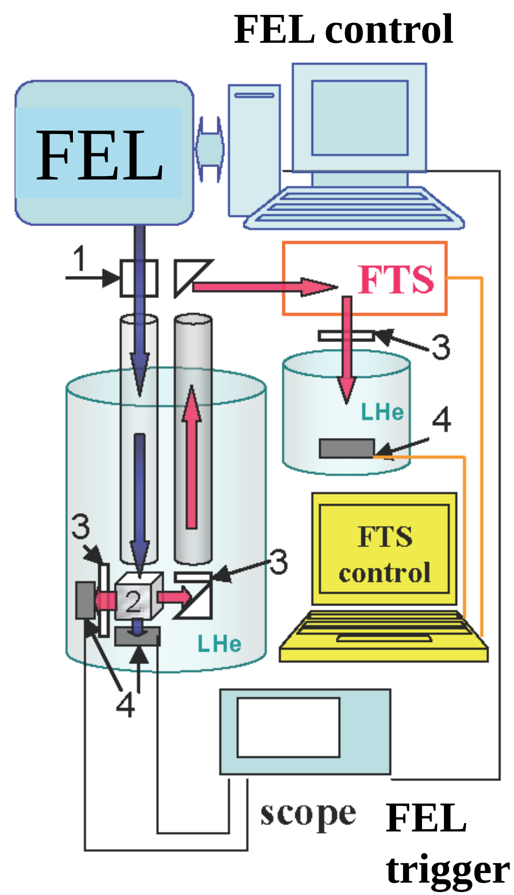
resonator on total internal reflection

IR free electron laser

(FELIX, NL)
5-10 ps pulses
1/20/40 ns separation
up to 1 MW peak
λ~16-42 µm

(CLIO, F)
5-10 ps pulses
16 ns separation
up to 10 MW peak
λ~16-42 µm

(FELBE, D)
5-10 ps pulses
77 ns separation
up to 0.1 MW peak
λ~16-22 µm

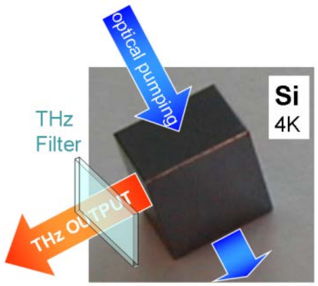


Volume enhanced ?

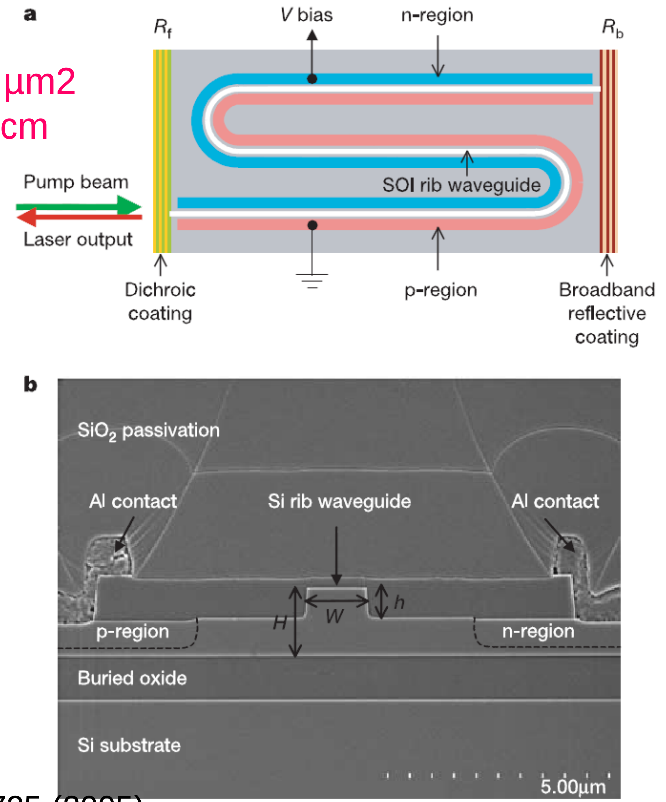
Transparent in THz	band-gap laser	under band-gap
up to ~1e14 centers	*up to ~1e10 centers*	*up to ~1e14 centers*

core 1.6 µm2
length 4.8 cm

skin depth
under 1 µm



resonator on total internal reflection



Nature 433, 725 (2005)

Figure 1 Silicon waveguide used in the Raman laser experiment. **a**, Schematic layout of the silicon waveguide laser cavity with optical coatings applied to the facets and a p-i-n structure along the waveguide. **b**, Scanning electron microscope cross-section image of a silicon rib waveguide with a p-i-n diode structure.

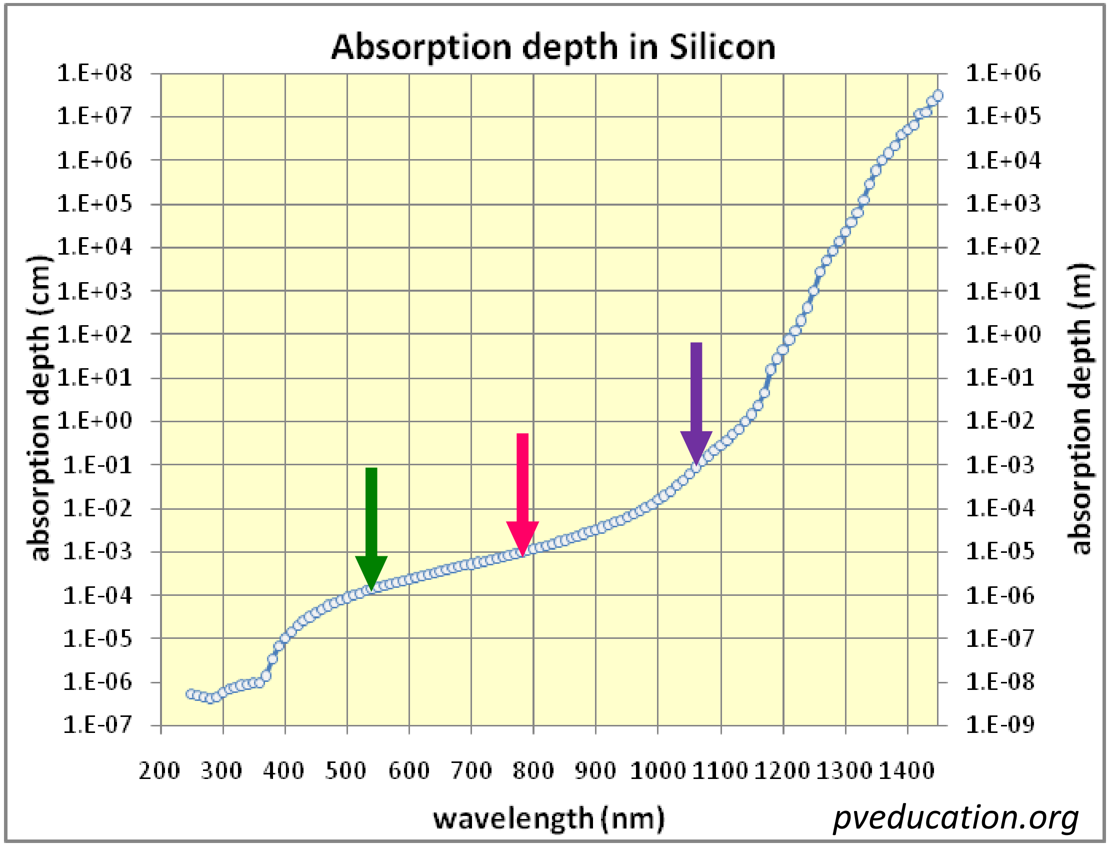
DLR

Volume enhanced !

532 nm band-gap laser *skin depth 1 µm* ~~*electronic Raman in Si*~~

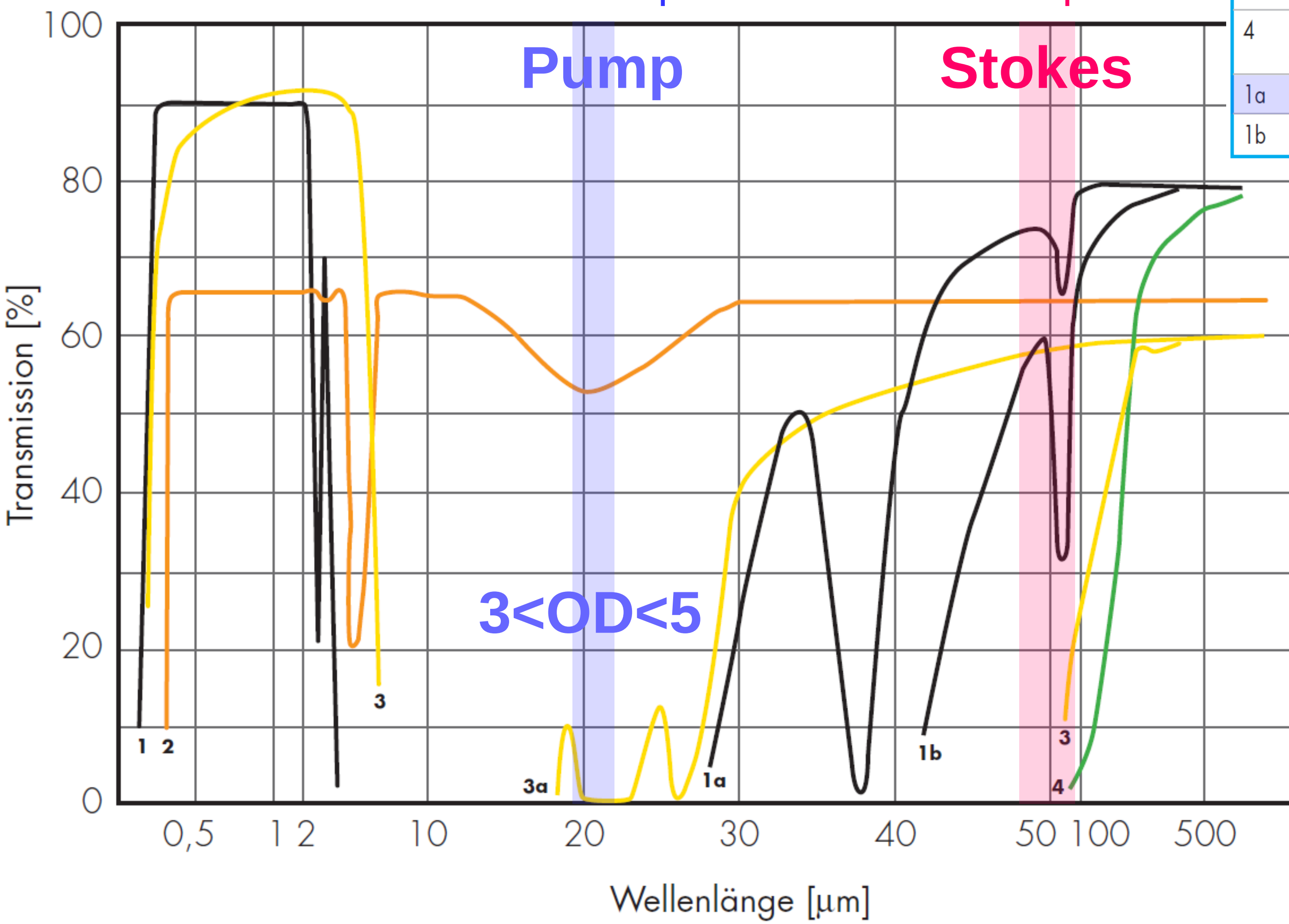
785 nm almost BG *skin depth 10 µm* *e-Raman in Si:Bi (40.8meV=30µm)*

1064 nm below BG *skin depth 1 mm* *e-Raman in n-Si (~23meV=54µm)*



Filtering with solids (lattice absorption)

n-Si: 18.3-21µm → 47-69µm

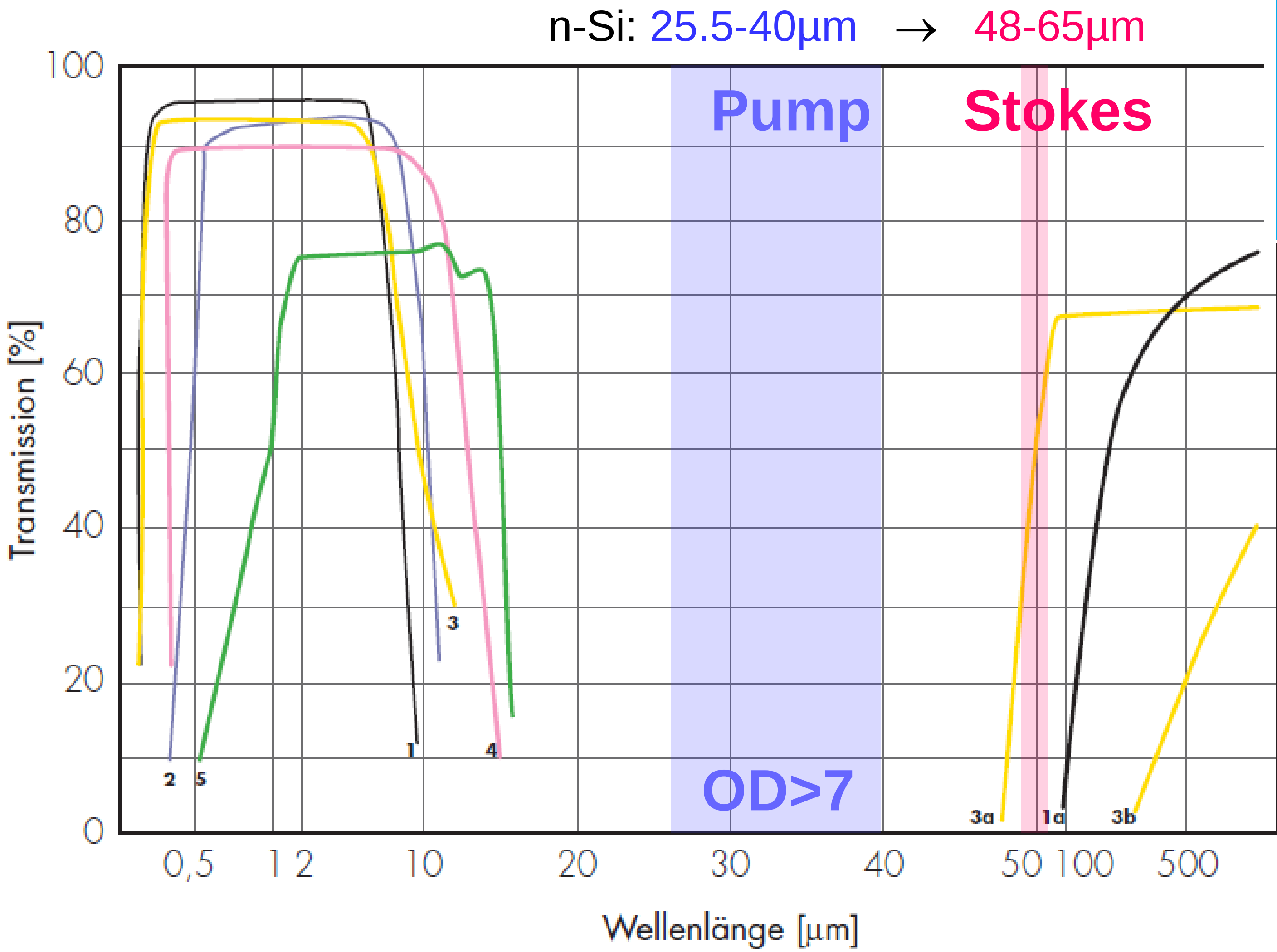


1	Quarz	kristallin	10 mm		
2	Diamant IIa		1 mm		
3	Al_2O_3	Saphir	1 mm		
3a	Al_2O_3	Saphir	1 mm	4°K	
4	Quarz-glas		1 mm		
1a	Quarz	kristallin	1 mm	4°K	Z-cut
1b	Quarz	kristallin	RT		

LOT ORIEL GRUPPE EUROPA

DLR

Filtering with solids (lattice absorption)



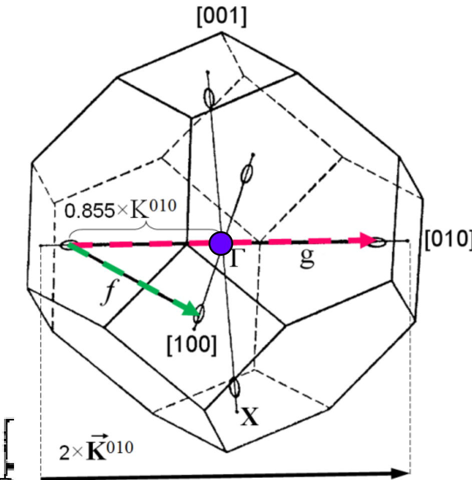
1	MgF_2	2 mm	
1a	MgF_2	1 mm	
2	CdF_2	5 mm	
3	CaF_2	1 mm	
3a	CaF_2	1 mm,	4°K
3b	CaF_2	3,5 mm	
4	PbF_2	2 mm	
5	CdS	2 mm	

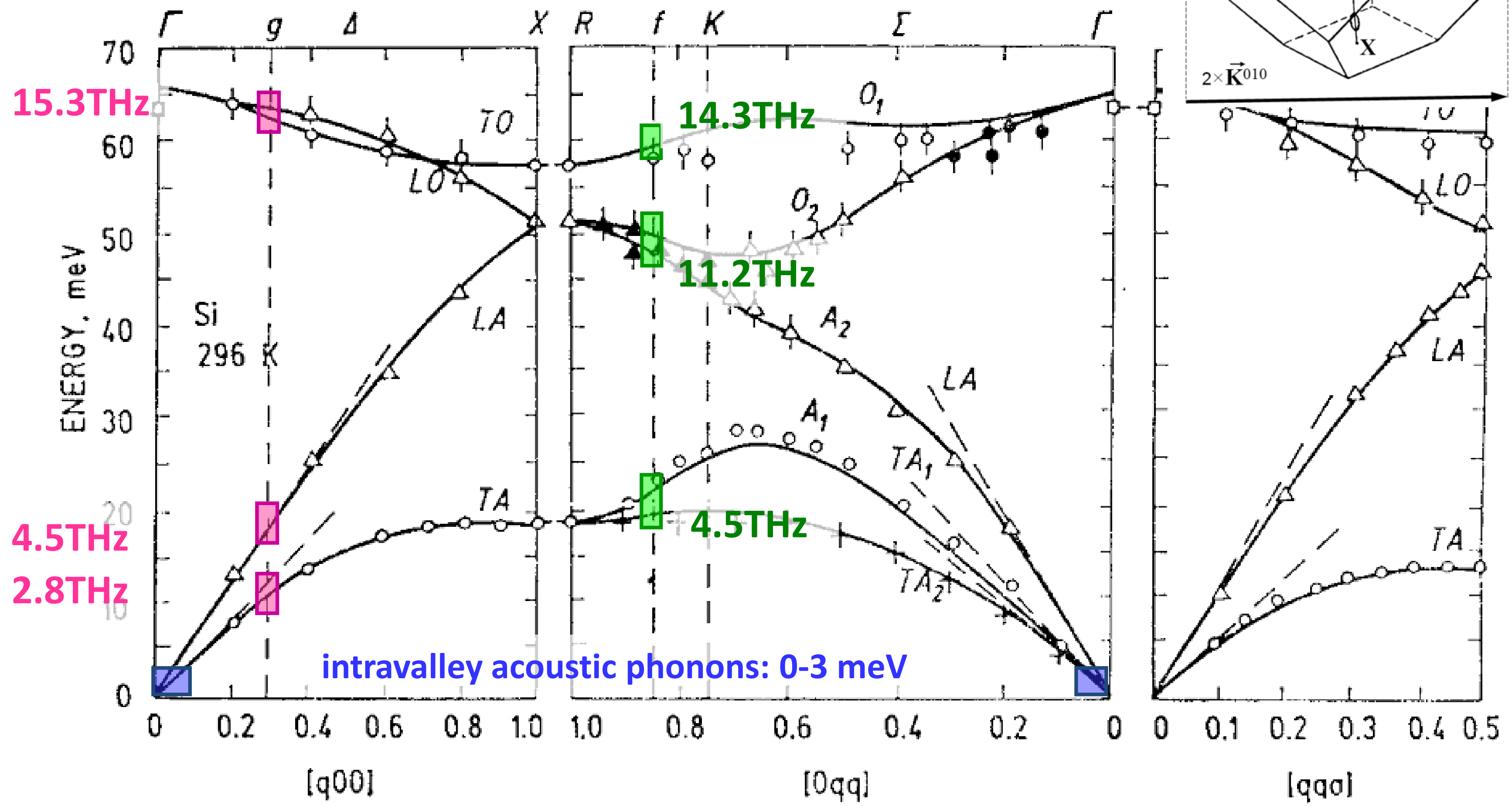
LOT ORIEL GRUPPE EUROPA

DLR

intra- and inter-valley phonons (a multi-valley semiconductor)

- Zone-centered optical phonons $f_{LTO}(\Gamma) \approx 15.6$ THz
- Phonons related to the critical points of the Brillouin zone
- Intervalley phonons, 3-15 THz



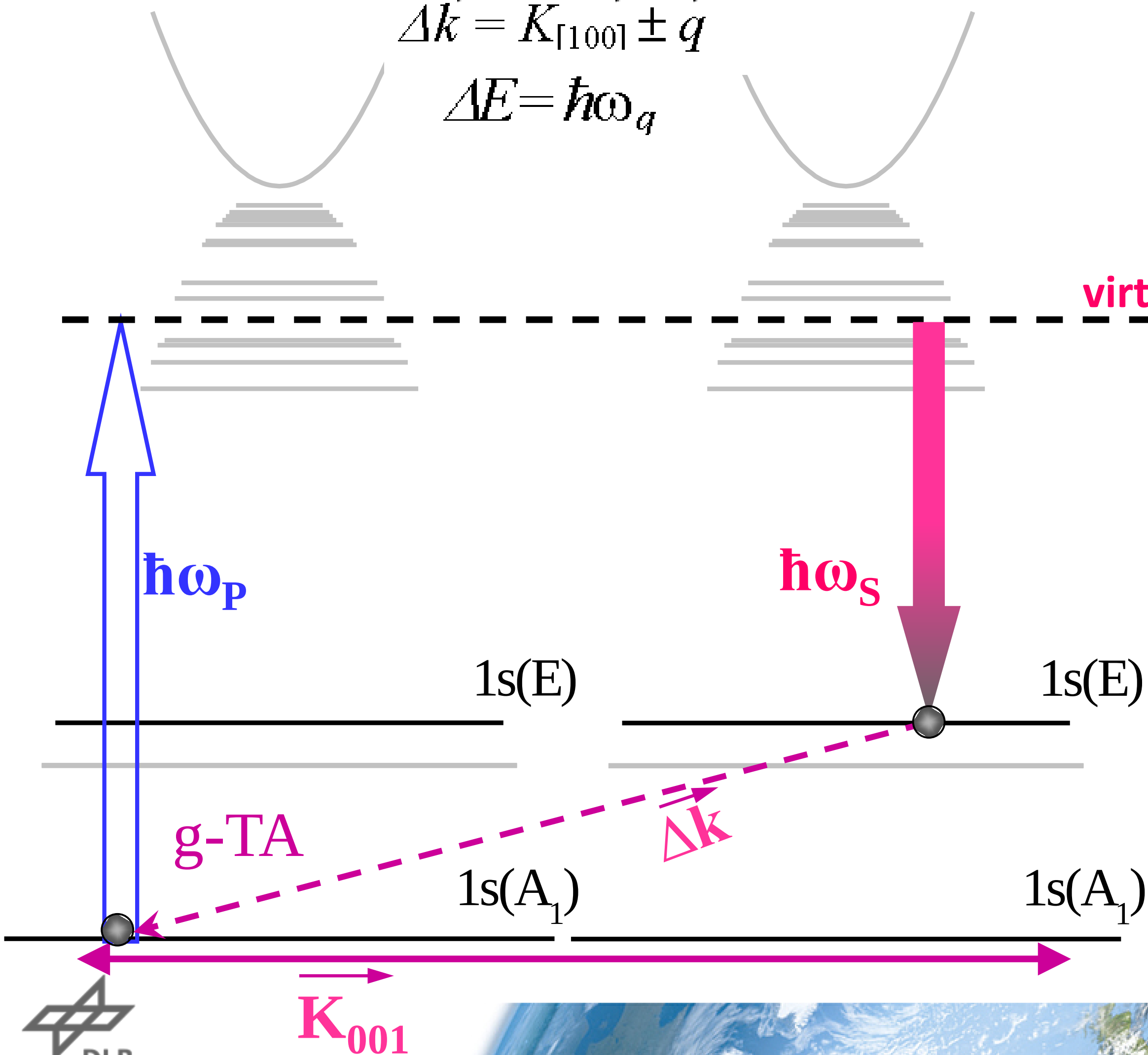


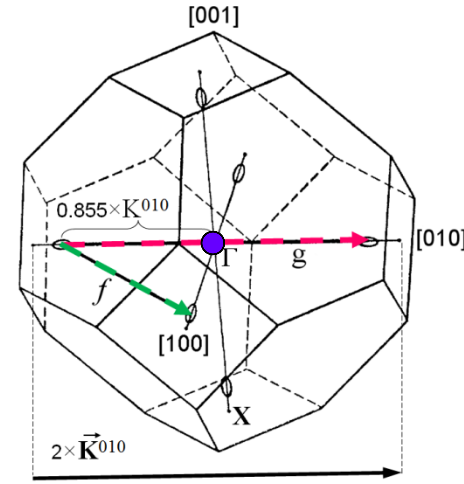
[Handbook on Physical Properties of Ge, Si, GaAs and InP, A.Dargys and J.Kundrotas, Vilnius, Lithuania. 1994..]

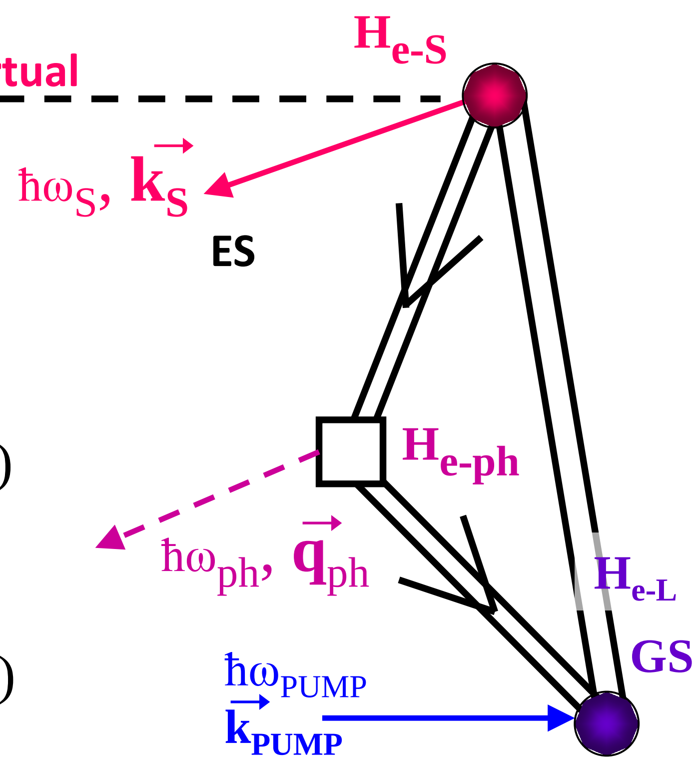
a single inter-valley phonon serves for the intracenter RS

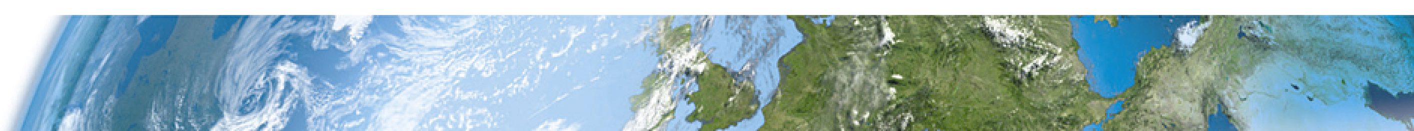
$$\vec{\Delta k} = \vec{K}_{[100]} \pm \vec{q}$$

$$\Delta E = \hbar\omega_q$$







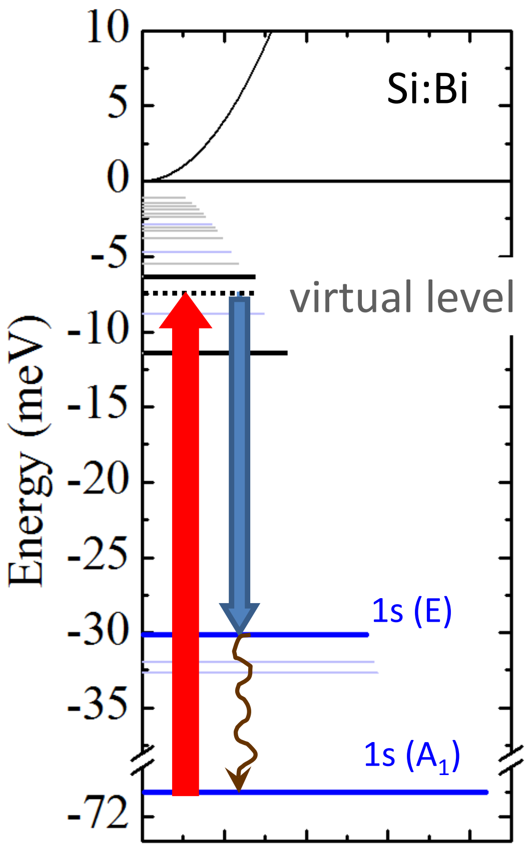


Electronic resonant Raman scattering

$$P(\omega_S) \sim \left| \frac{\langle \mathrm{v}|H_{\text{e-L}}|\mathrm{GS}\rangle \times \langle \mathrm{v}|H_{\text{e-S}}|\mathrm{ES}\rangle \times \langle \mathrm{ES}|H_{\text{e-ph}}|\mathrm{GS}\rangle}{[\hbar\omega_{\mathrm{PUMP}} - \Delta E_{\mathrm{V}}] \times [\hbar\omega_{\mathrm{PUMP}} - \hbar\omega_S - \Delta E_{\mathrm{ES}}]} \right|^2$$

$$\Delta E_{\mathrm{V}} = (E_{\mathrm{GS}} - E_{\mathrm{v}} + ih\Gamma_S)$$

$$\Delta E_{\mathrm{ES}} = (E_{\mathrm{GS}} - E_{\mathrm{ES}})$$



resonance to an impurity

Raman-active transition: outgoing

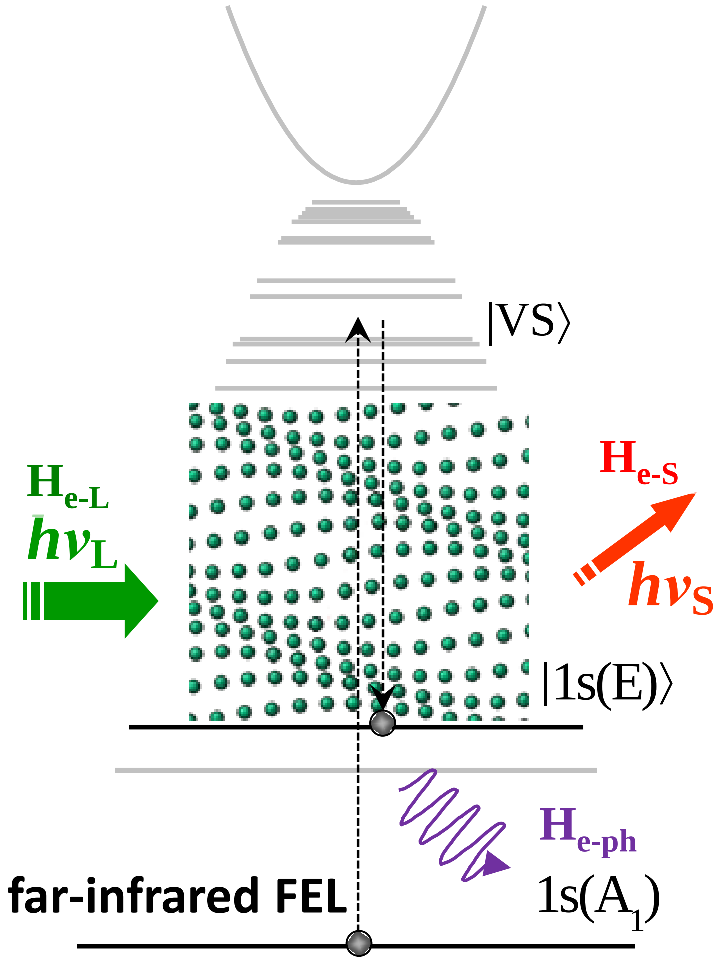
$$\hbar\omega_{\mathrm{PUMP}} - \hbar\omega_S = \Delta E_{\mathrm{ES}}$$

$1s(E) \rightarrow 1s(A_1)$ Raman-active

resonance in pumping: ingoing

$$\hbar\omega_{\mathrm{PUMP}} = E_{\mathrm{GS}} - E_{\mathrm{v}}$$

photon-bound_electron-phonon (free_electron-free) interaction



free carrier absorption $n\lambda^2$

THz Raman scattering: Overcoming the Challenges

> scattering efficiency, **N**λ^{-4}

resonant (outcoming+incoming)

> either free carrier absorption, λ^2

or **excitation avoiding free electrons = intracenter**

FEL

> under optical phonon excitation

= high orders of electron-phonon interaction

intervalley one-phonon intracenter scattering

> technical (filters, collecting optics)

strong lattice absorption in solids = Low Pass filter

DLR

Raman scattering at THz frequencies:

♦ ***enabling 7-17 THz Raman scattering in n-silicon***:

\+ ***resonant*** *to donor electronic states coupled by intervalley phonons*

\+ *large* ***number*** *of scattering centers (volume, up to ~1e14 centers)*

\+ ***free_electron-free*** *(photon-bound_electron-phonon)*
interaction cancels free carrier absorption

♦ ***features of the stimulated THz Raman scattering in n-silicon***

\+ **lasing threshold** exceed 3e23 photons/cm^2/s, the lowest ones are for the Si:Sb and Si:Bi with the particular donor-phonon resonances

\+ the **Stokes shifts** (2.5 – 9 THz) of the Raman lasing corresponds to a Raman-active donor 1s(A_1)-1s(E) transition

\+ the Raman **gain** is estimated to be up to 5.8 cm/MW

\+ the **optical conversion factor** is within 1e-8 – 1e-9

\+ the donor concentration limit < 8e15/cm^3 (1s(A_1)-1s(E) donor transition broadening)

Acknowledgements

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