

# FEL-pumped Silicon lasers based on hydrogen-like impurity centers

Heinz-Wilhelm Hübers

German Aerospace Center (DLR), Institute of Optical Sensor Systems, Berlin  
and

Humboldt-Universität zu Berlin, Department of Physics

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Knowledge for Tomorrow

# Co-workers

S. Pavlov

*German Aerospace Center, Berlin, Germany*

N. Deßmann, A. Pohl

*Humboldt-Universität zu Berlin, Germany*

N. Abrosimov, H. Riemann

*Institute of Crystal Growth, Berlin, Germany*

V. Shastin, K. Kovalevsky, V. Tsyplenkov, R.Kh. Zhukavin

*Institute for Physics of Microstructures, Nizhny Novgorod, Russia*

J.N. Hovenier (+), T.O. Klaassen

*Delft Technical University, The Netherlands*

A.F.G. van der Meer, B. Redlich

*FELIX, Radboud University, Nijmegen, The Netherlands*

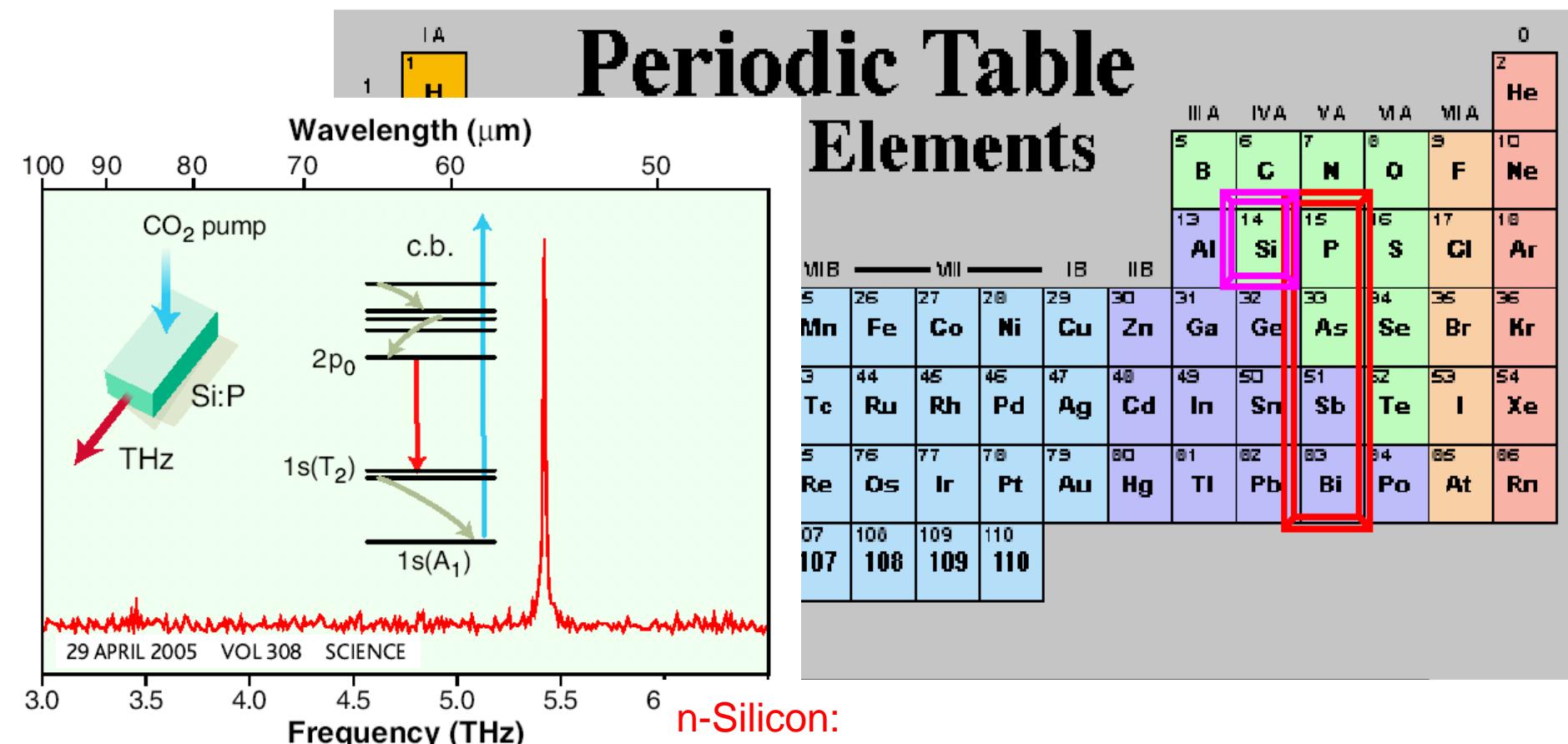


# Outline

- Motivation
- Impurity states in Silicon
- n-type Silicon lasers
  - based on photoionization
  - based on photoexcitation
- THz Silicon Raman laser
- p-type Silicon laser
- Summary



# Basic idea

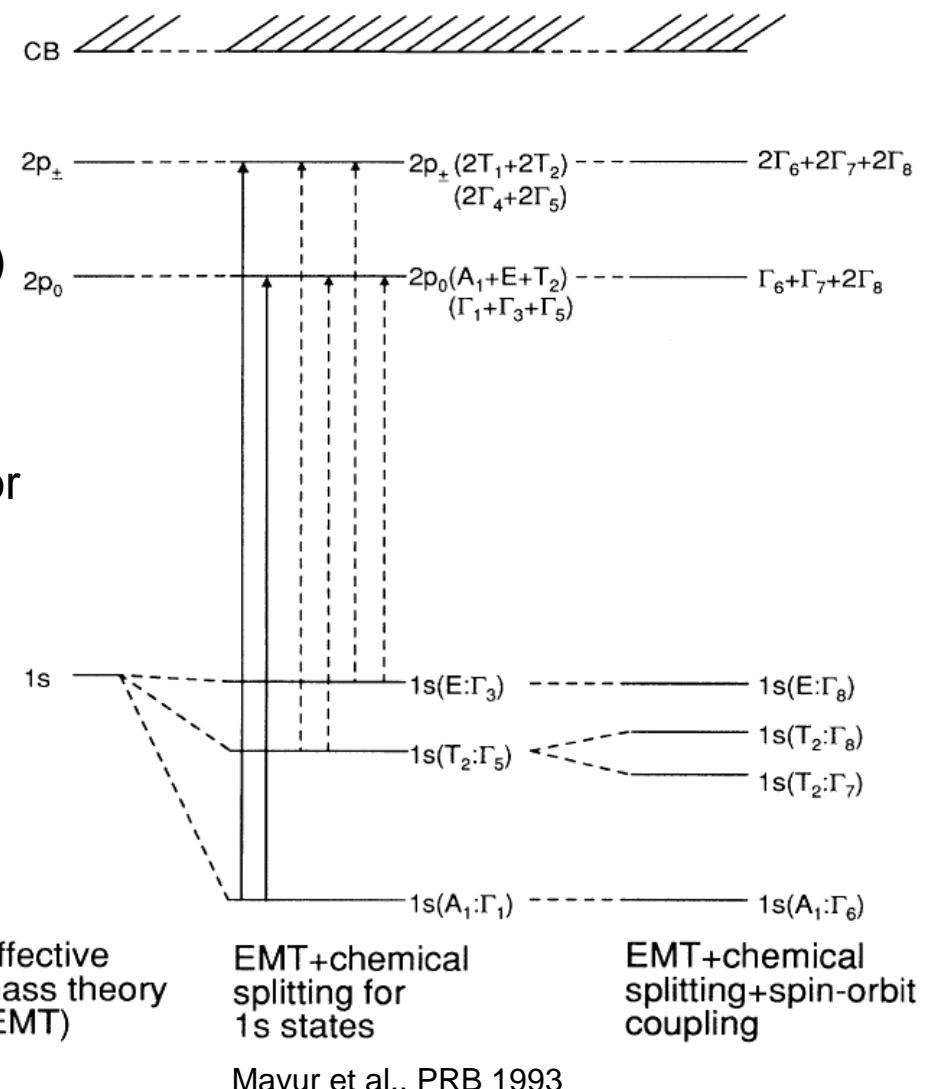


- Hydrogen like energy levels of shallow impurities
- Transitions between states are in the THz range
- Make use of peculiarities of these levels:  
long-lived states, resonant phonon interaction

# Impurity states in n-type Si

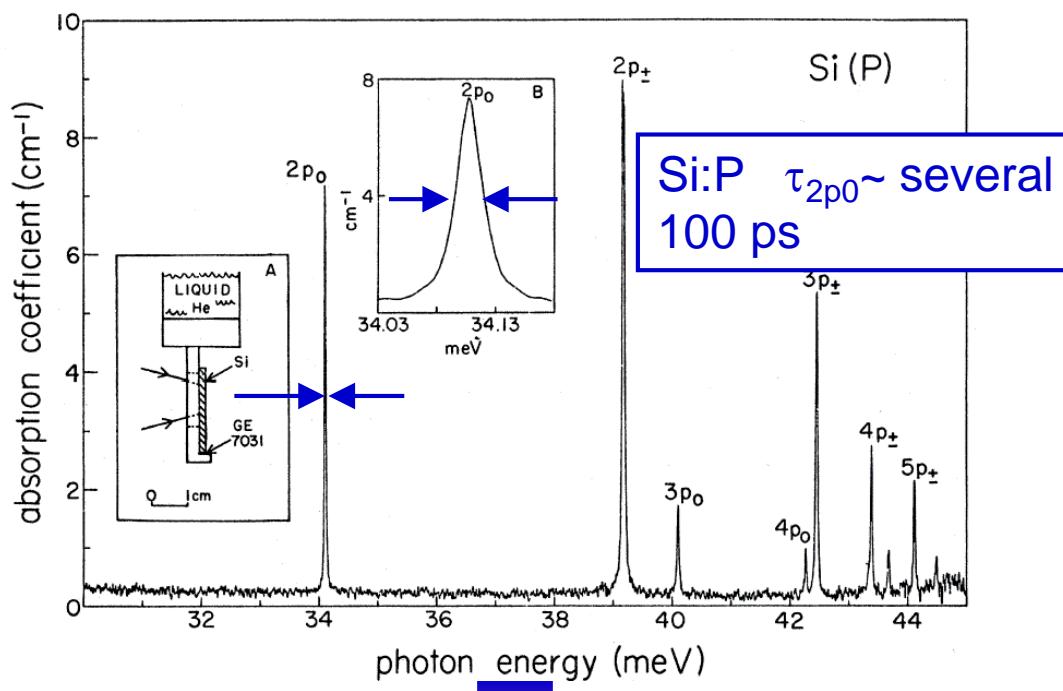
## Doping with group-V donors

- Substitutional Donors (P, As, Sb, Bi)
  - Hydrogen-like spectrum (EMT)
- $$E_n = - R / n_2 \varepsilon^2 (m_D/m)$$
- Chemical splitting depends on donor
  - Spin-orbit coupling



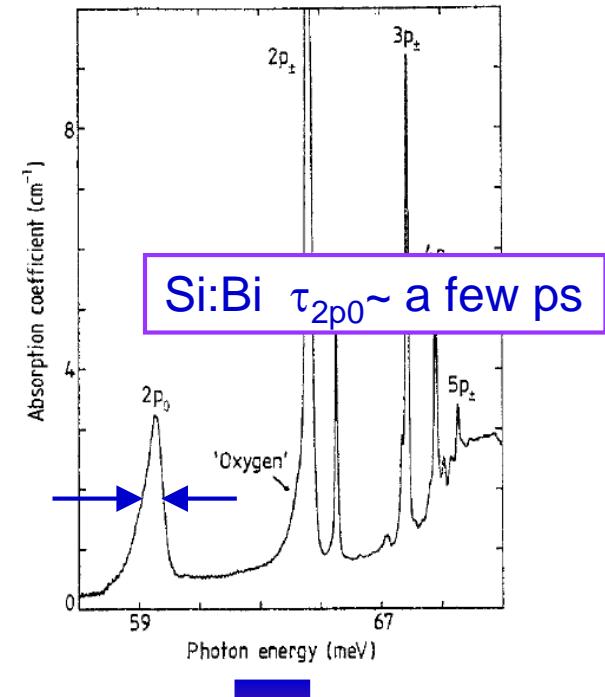
Mayur et al., PRB 1993

# Spectroscopy and life times



long life times of  
impurity states

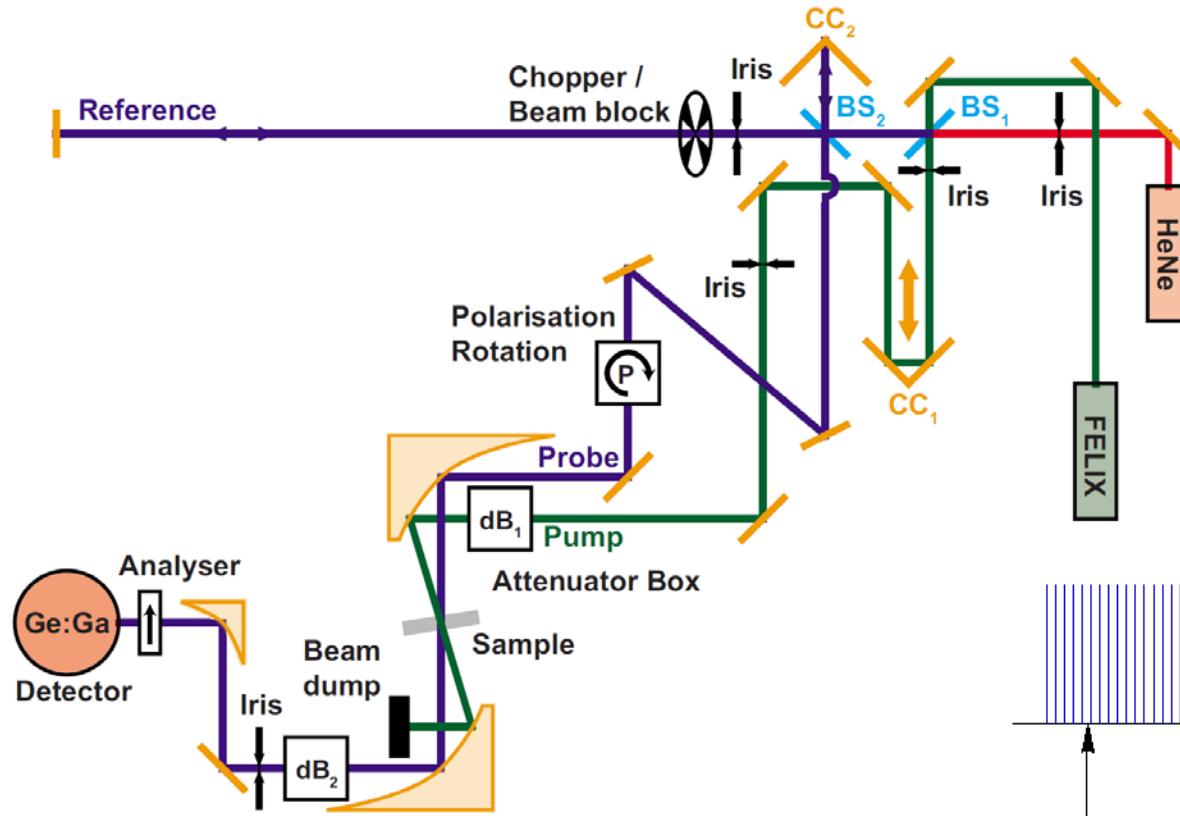
Jagannath et al., PRB **23**, 2082 (1981)



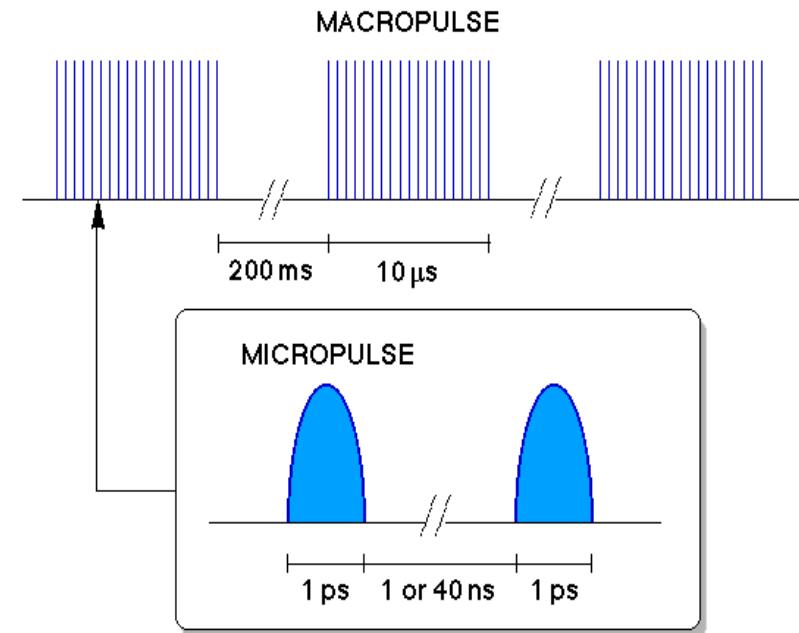
short life times of  
impurity states

Butler et al., PRB **12**, 3200 (1975)

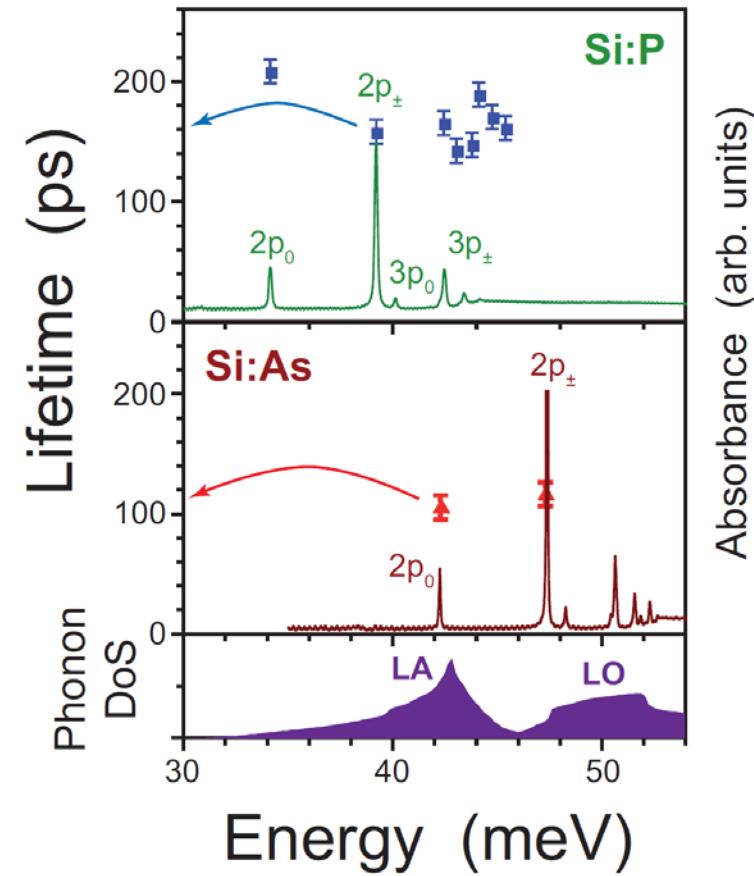
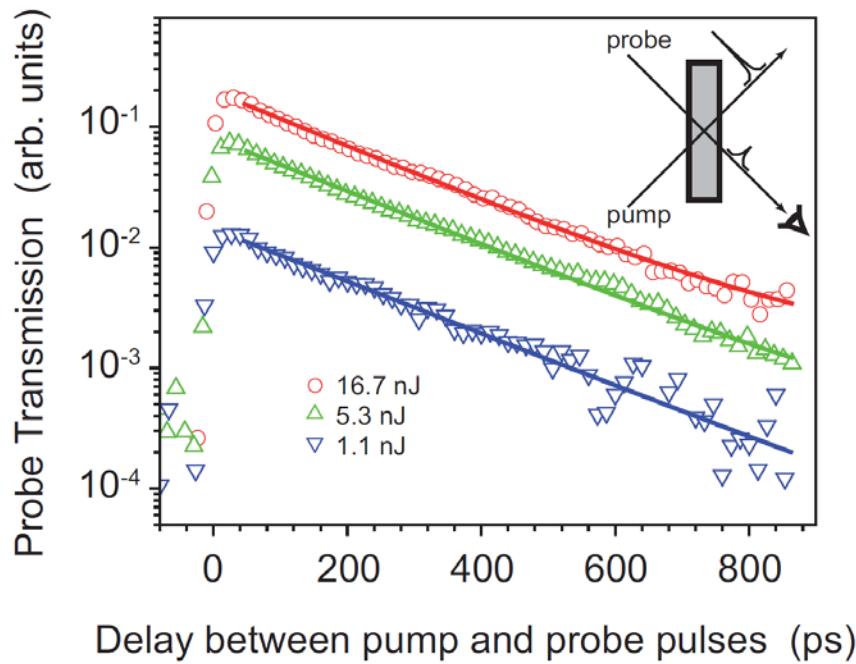
# Pump-probe set-up at FELIX



S. Lynch et al., PRB 82, 245206 (2010)

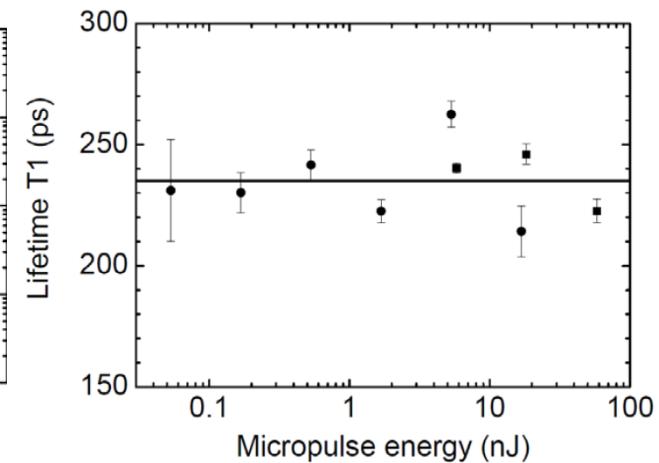
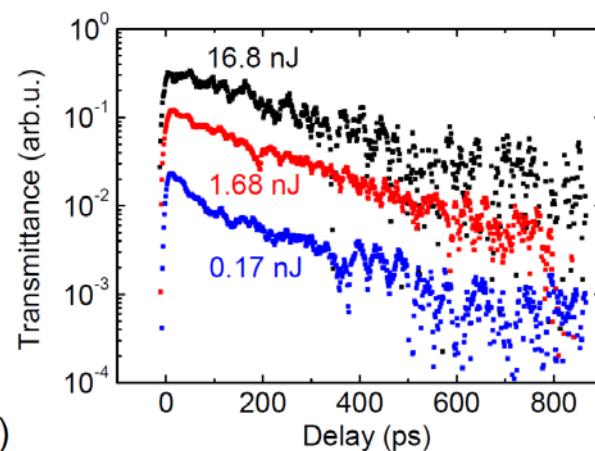
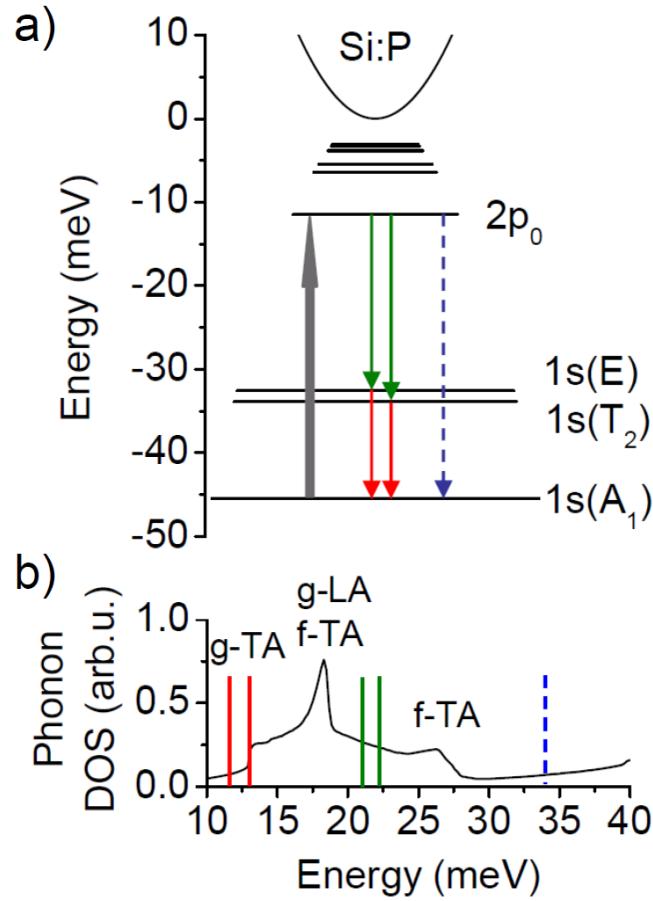


# Lifetime measurements by pump-probe technique



N. Vinh et al., PNAS 105, 10649 (2008)

# Lifetime: natural Si:P vs. isotopically pure $^{28}\text{Si}$



- ${}^{\text{nat}}\text{Si}$ : 92.23%  $^{28}\text{Si}$ , 4.67%  $^{29}\text{Si}$ , 3.1%  $^{30}\text{Si}$
- $^{28}\text{Si}$ : purity > 99.994, doping =  $2 \times 10^{15} \text{ cm}^{-3}$
- $T_1({}^{28}\text{Si}) = 235 \text{ ps} > T_1({}^{\text{nat}}\text{Si}) = 205 \text{ ps}$
- The longer lifetime of  $^{28}\text{Si}$  is due to its higher symmetry and a less pronounced interaction of the  $2p_0, 1s(E)$  and  $1s(T_2)$  states with the g-LA and f-TA phonons.

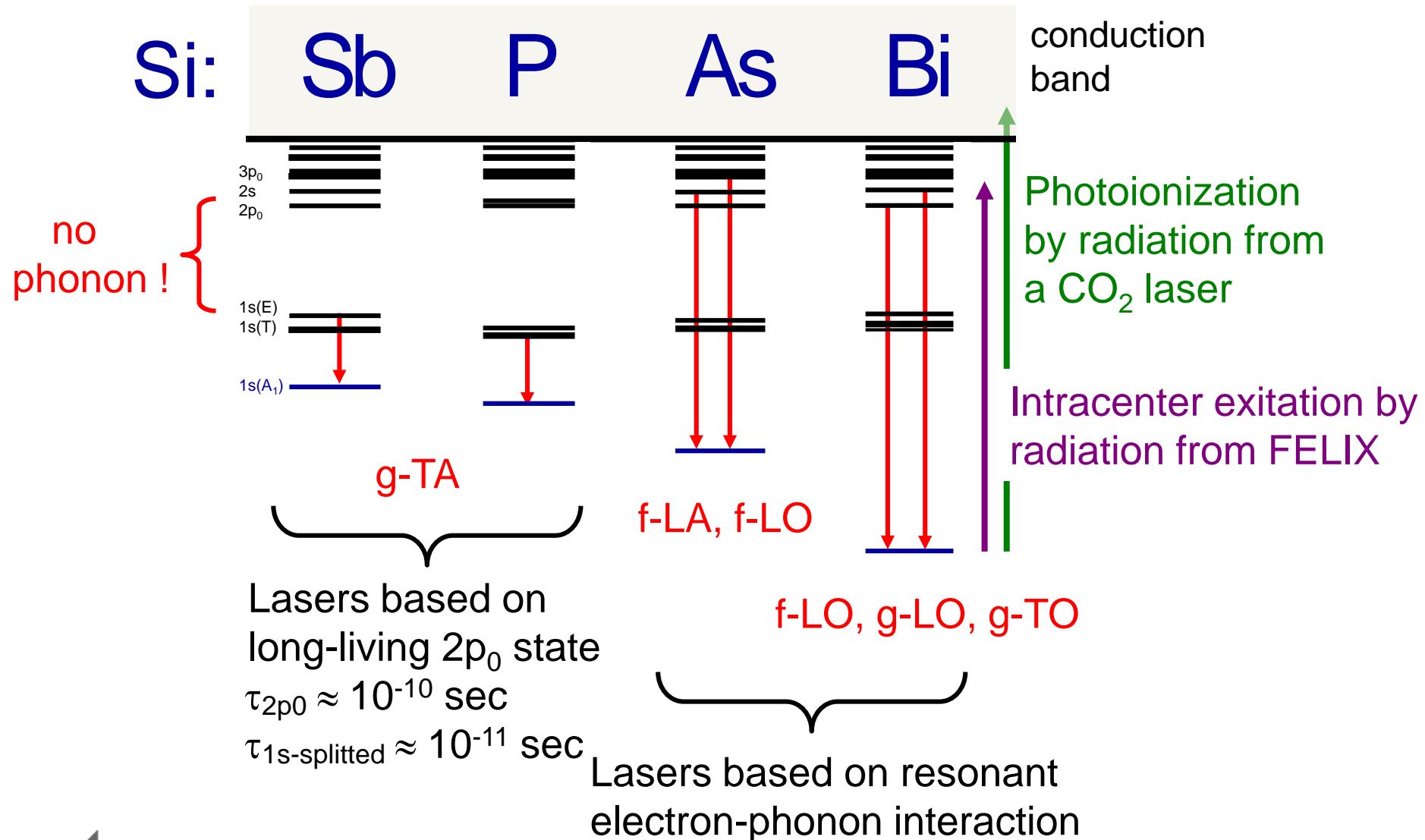
H.-W. Hübers et al., PRB (2013)

# Outline

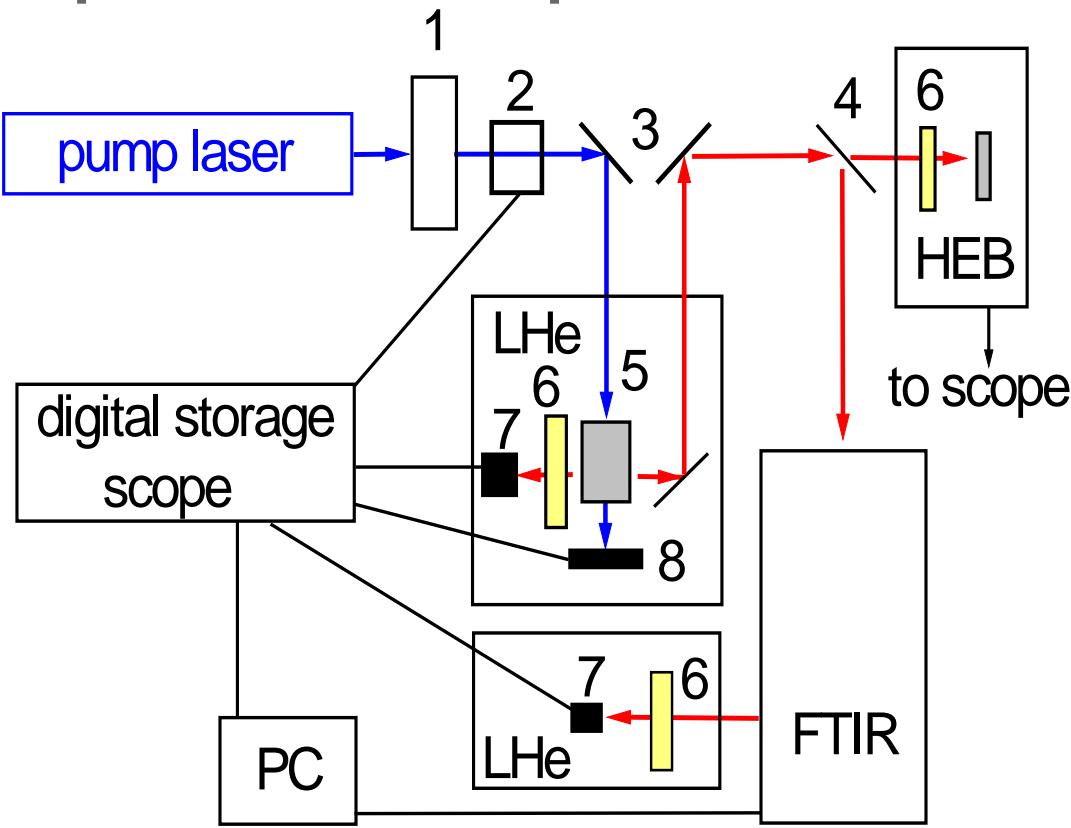
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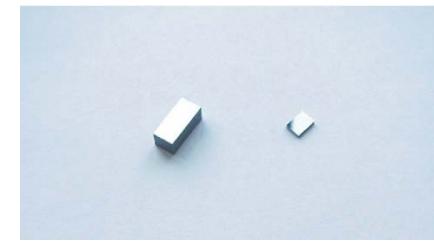
# Interaction with phonons



# Experimental setup

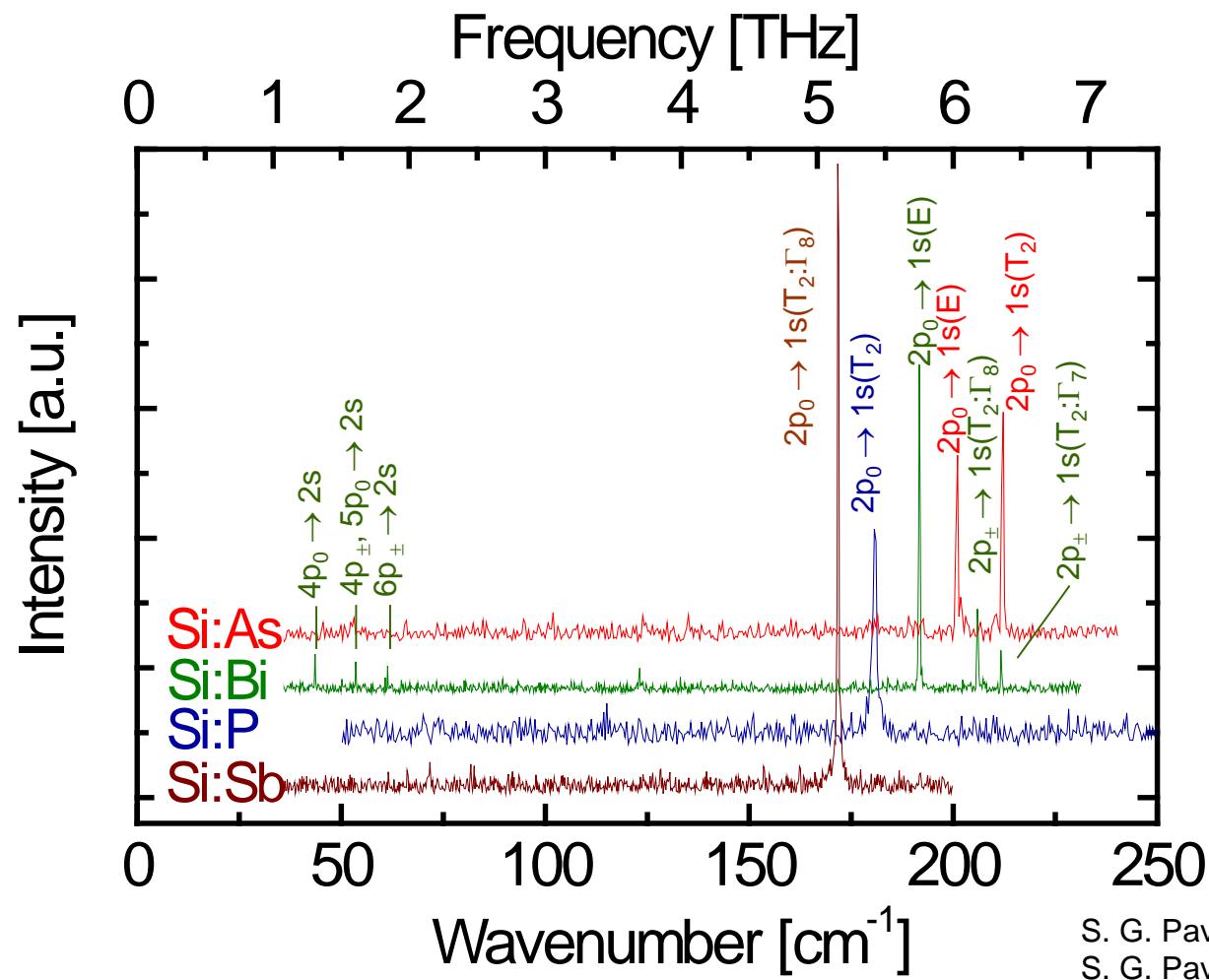


- 1 - pump attenuator
- 2 - photon drag monitor
- 3 - mirrors
- 4 - beam splitter
- 5 - sample
- 6 - FIR filters
- 7 - FIR detector
- 8 - alignment detector



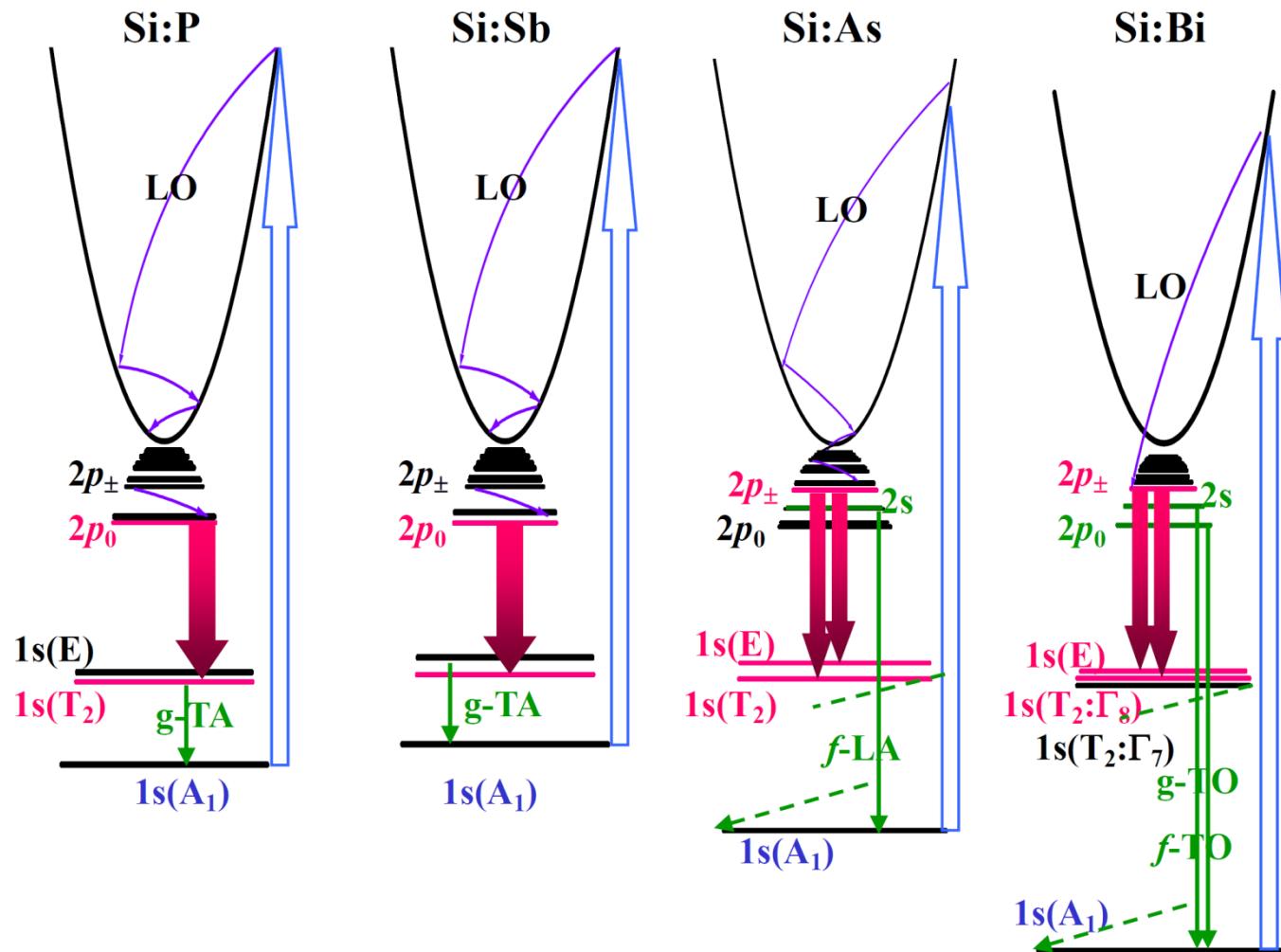
n-Si monocrystals  
doped  $\sim 10^{15} - 10^{16} \text{ cm}^{-3}$ ,  
compensated < 30 %  
length is 5-15 mm  
FZ or CZ grown

# Photoexcitation with a CO<sub>2</sub> laser: spectra



S. G. Pavlov et al., PRL **84**, 5220, 2000  
S. G. Pavlov et al., APL **80**, 4717, 2002  
S. G. Pavlov et al., JAP **92**, 5632, 2002  
H.-W. Hübers et al., APL **84**, 3600, 2004

# Laser schemes under photoexcitation with a CO<sub>2</sub> laser



# Comparison with spectroscopy

**Table 1** Binding energy of some excited states for hydrogen-like donor centers in silicon as derived from an analysis of pump and emission spectra of silicon intracenter lasers under resonant photoexcitation.

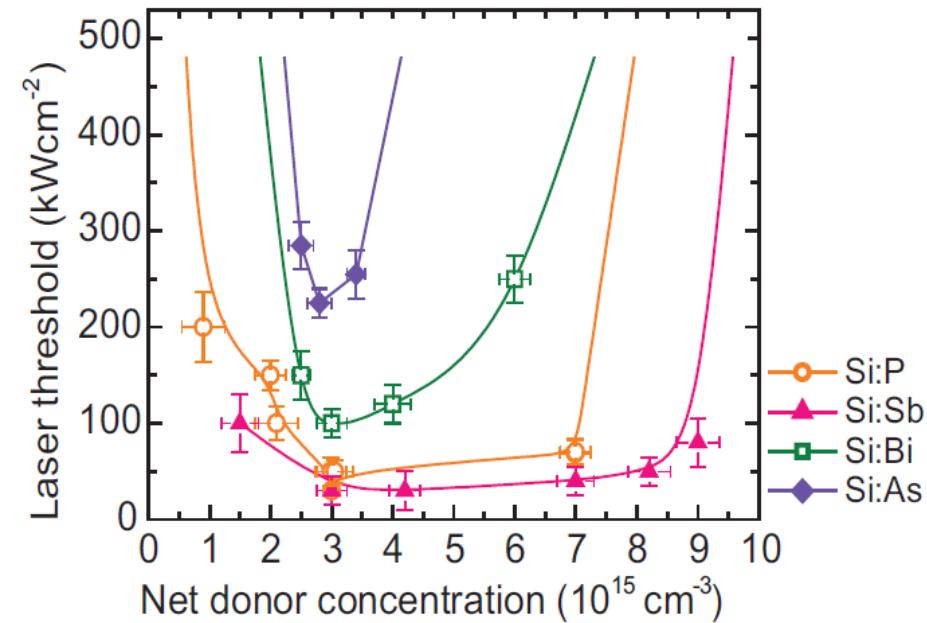
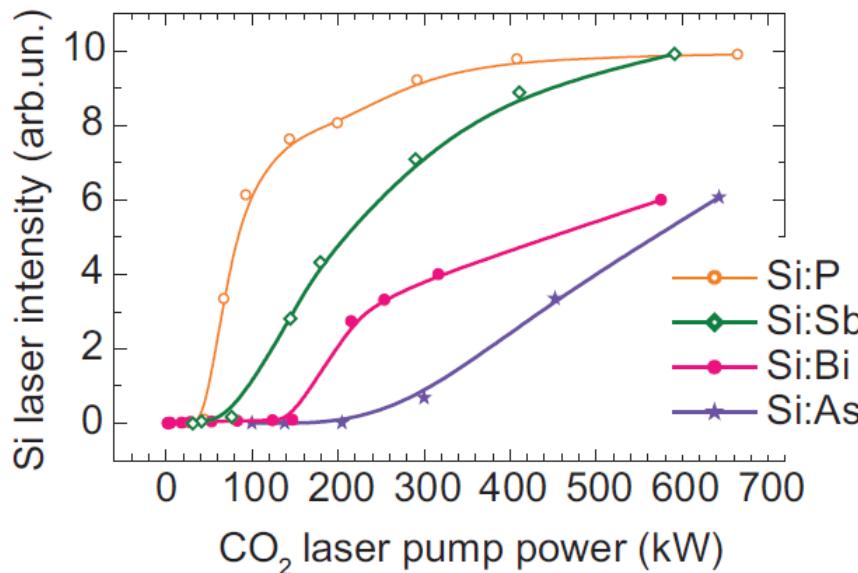
donor	1s( $T_2$ )			1s( $E$ )		
	n-Si lasing	cal.	spectroscopy	n-Si lasing	cal.	spectroscopy
P	$33.91 \pm 0.02$ (4 K)	34.2	$33.89 \pm 0.01$ (45 K)	$32.61 \pm 0.02$ (4 K)	32.7	$32.56 \pm 0.01$ (45 K)
Sb	$32.83 \pm 0.02^a$ (4 K)	32.9	$32.83 \pm 0.01^a$ (30 K) $33.12 \pm 0.01^b$ (30 K)	$30.50 \pm 0.02$ (4 K)	30.5	$30.53 \pm 0.01$ (30 K)
As	$32.73 \pm 0.02$ (4 K)	32.7	$32.68 \pm 0.01$ (60 K)	$31.34 \pm 0.02$ (4 K)	31.3	$31.25 \pm 0.01$ (60 K)
Bi	$31.90 \pm 0.02^a$ (4 K)	31.3	$31.89 \pm 0.01$ (80 K)	$30.17 \pm 0.02$ (4 K)	31.3	$30.47 \pm 0.01$ (80 K)
	$32.63 \pm 0.01^b$ (4 K)		$32.89 \pm 0.01^{HD}$ (10 K)			

<sup>a</sup>Doublet 1s( $T_2:\Gamma_8$ ); <sup>b</sup>singlet 1s( $T_2:\Gamma_7$ ). References: n-Si lasing: as deduced from pump and emission spectra of silicon intracenter lasers [24]; spectroscopy: impurity absorption spectroscopy at elevated crystal temperatures [92]; and for highly doped Si:Bi<sup>HD</sup> at  $T \sim 10$  K [93]; cal.: as calculated by EMT with an empiric model Hamiltonian [90]; for Si:Bi: from the standard EMT [38].

- Laser emission frequencies (measured at 4 K) and transition frequencies as measured by FTIR (measured at 30-80 K) agree very well.



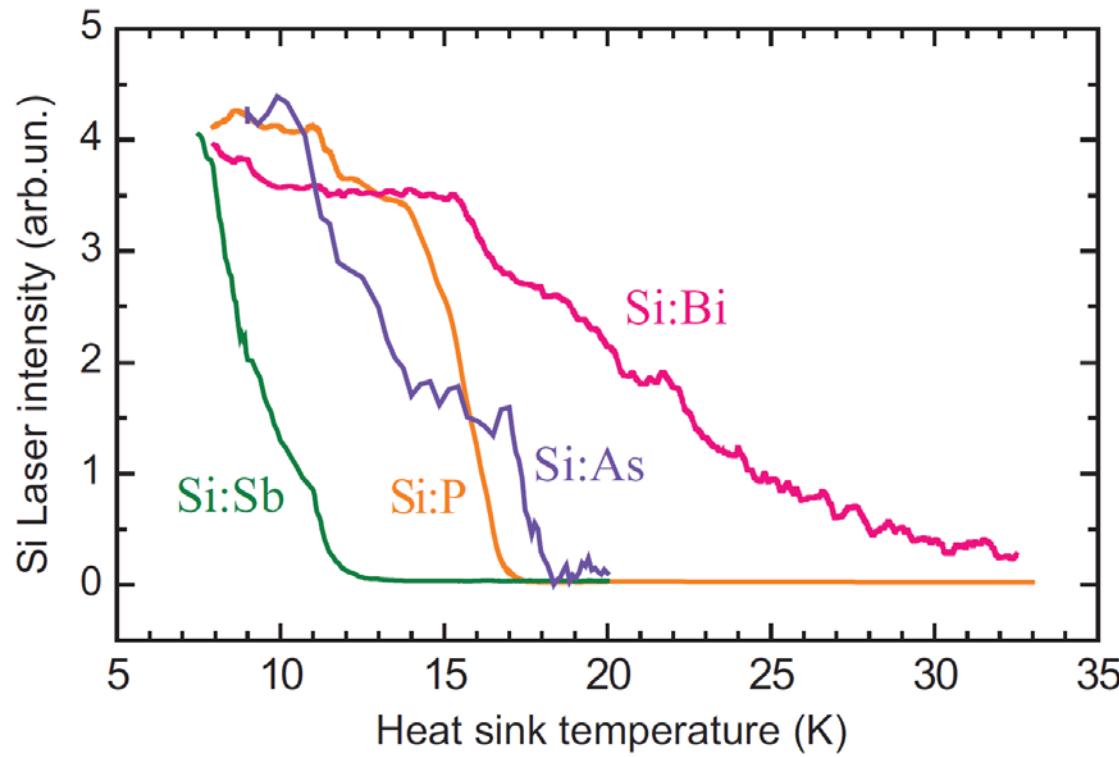
# Laser threshold



- Si samples at optimum doping concentration ( $3 \times 10^{15} \text{ cm}^{-3}$ )
- Pumping at 10.59 μm,  $100 \text{ kW/cm}^2 = 5 \times 10^{24} \text{ photons cm}^{-2}\text{s}^{-1}$
- Si:P, Si:Sb:  $2p_0 \rightarrow 1s(T_2)$ , pumping at 10.59 μm, 400 kW/cm<sup>2</sup>
- Si:As, Si:Bi:  $2p_{\pm} \rightarrow 1s(E)$ ,  $1s(T_2)$ , pumping at 9.6 μm, 80 kW/cm<sup>2</sup>
- Si:As, Si:Bi:  $2p_{\pm} \rightarrow 1s(E)$ ,  $1s(T_2)$

- Si:P, Si:Sb:  $2p_0 \rightarrow 1s(T_2)$ , pumping at 10.59 μm, 400 kW/cm<sup>2</sup>
- Si:As, Si:Bi:  $2p_{\pm} \rightarrow 1s(E)$ ,  $1s(T_2)$ , pumping at 9.6 μm, 80 kW/cm<sup>2</sup>
- Upper limit: impurity broadening, lifetime decreases
- Lower limit: too few donors, less gain

# Temperature



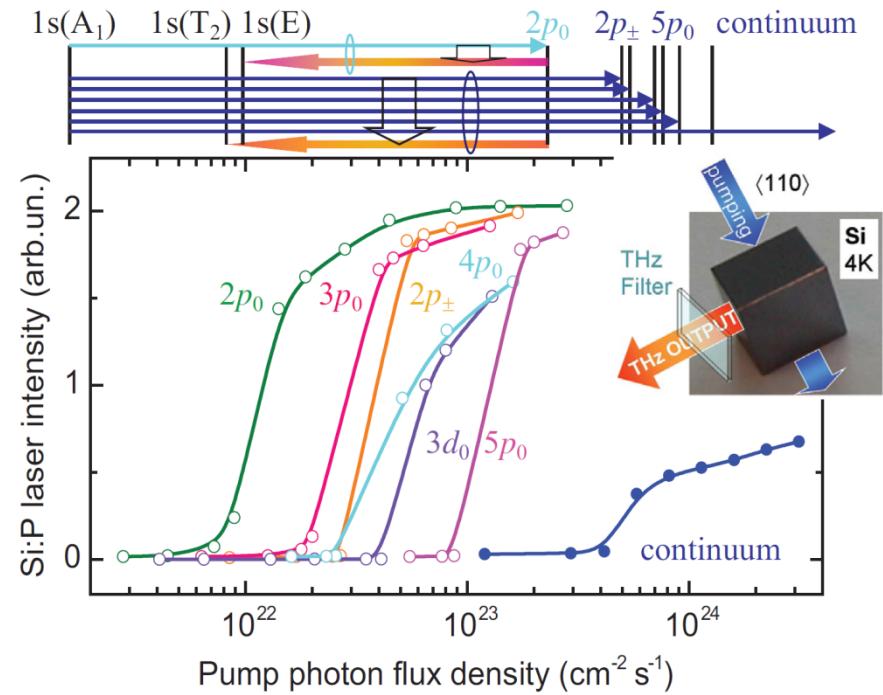
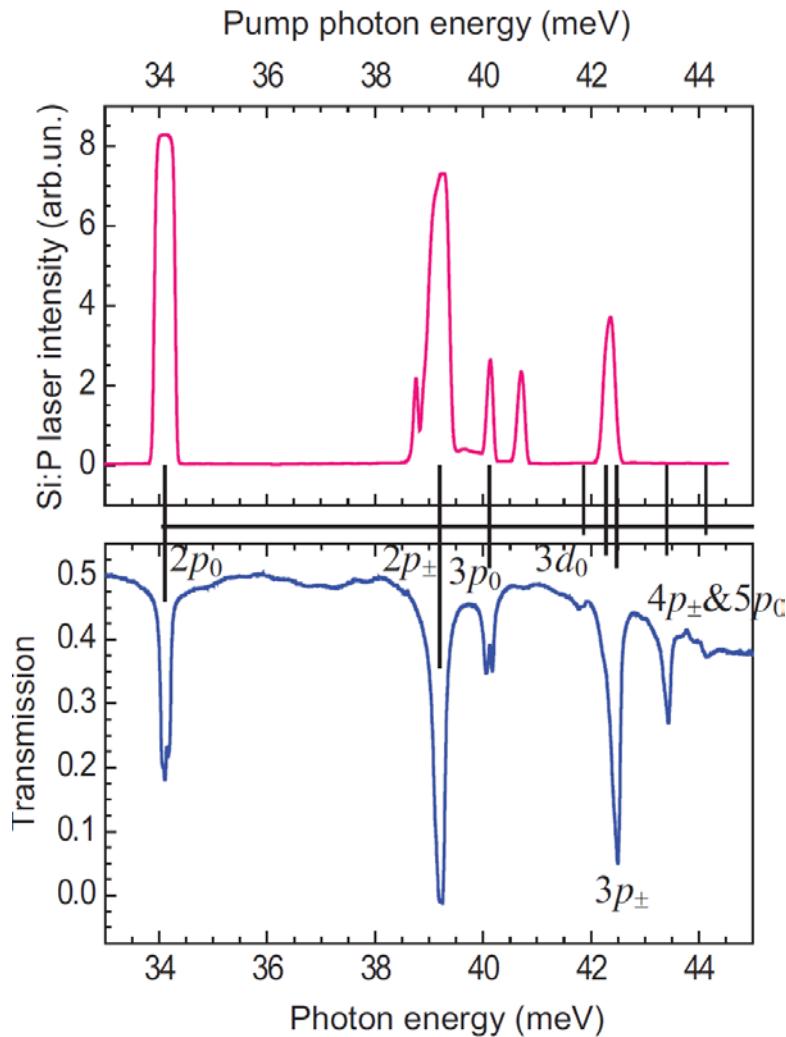
- The larger the gap between the  $1s(A)$  ground state and the  $1s(E)$ ,  $1s(T_2)$  split-off states the higher the operation temperature of the laser (less thermal population of the lower laser level).

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# Photoexcitation of Si:P

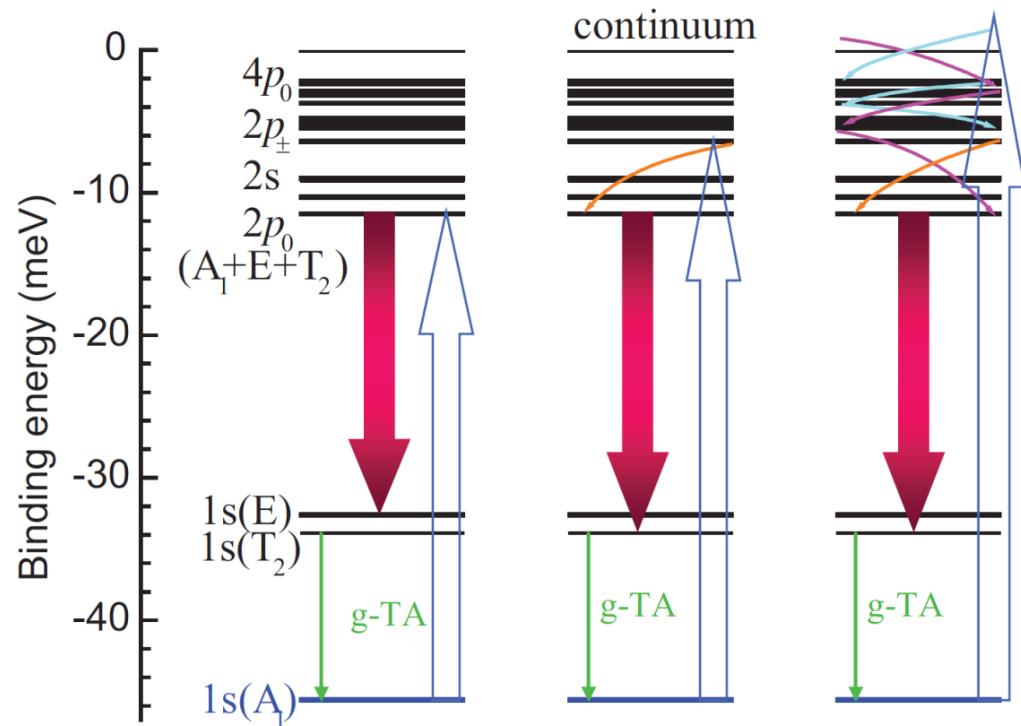


- At low pump power: laser emission only when pumped into a state
- The higher the pumped state the lower the laser threshold, due to increased non-radiative recombination



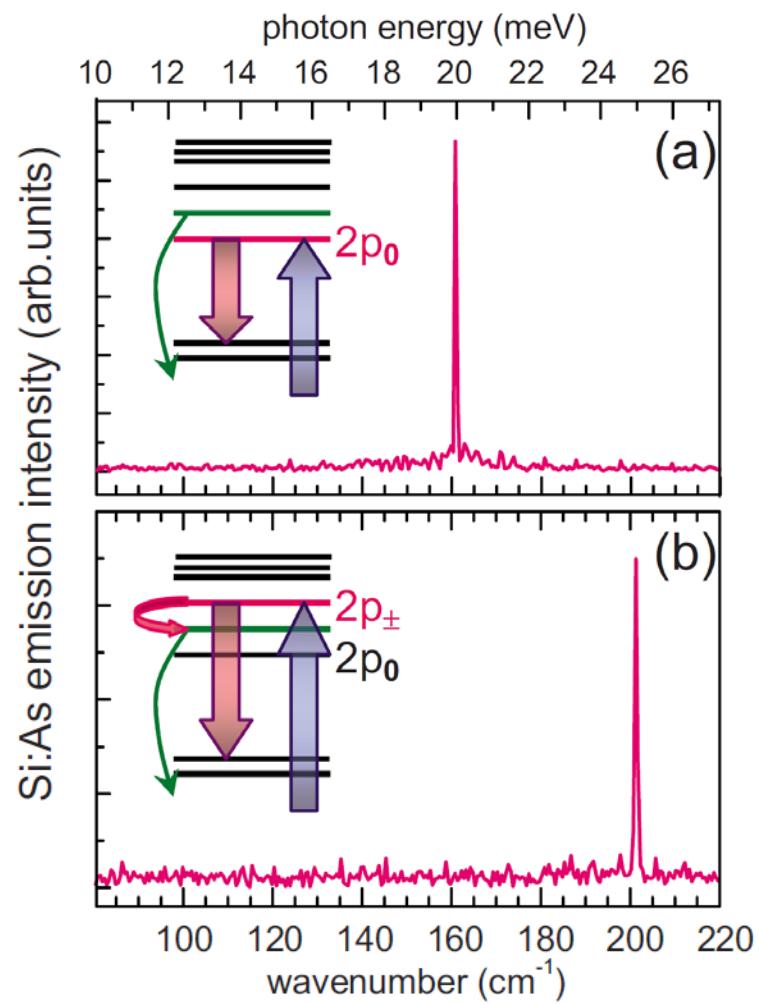
# Photoexcitation of Si:P

- Pumping into  $2p_0$ :  
 $2p_0 \rightarrow 1s(E)$
- Pumping into a state  $> 2p_0$ :  
 $2p_0 \rightarrow 1s(T_2)$
- Pumping into conduction band:  
 $2p_0 \rightarrow 1s(A)$

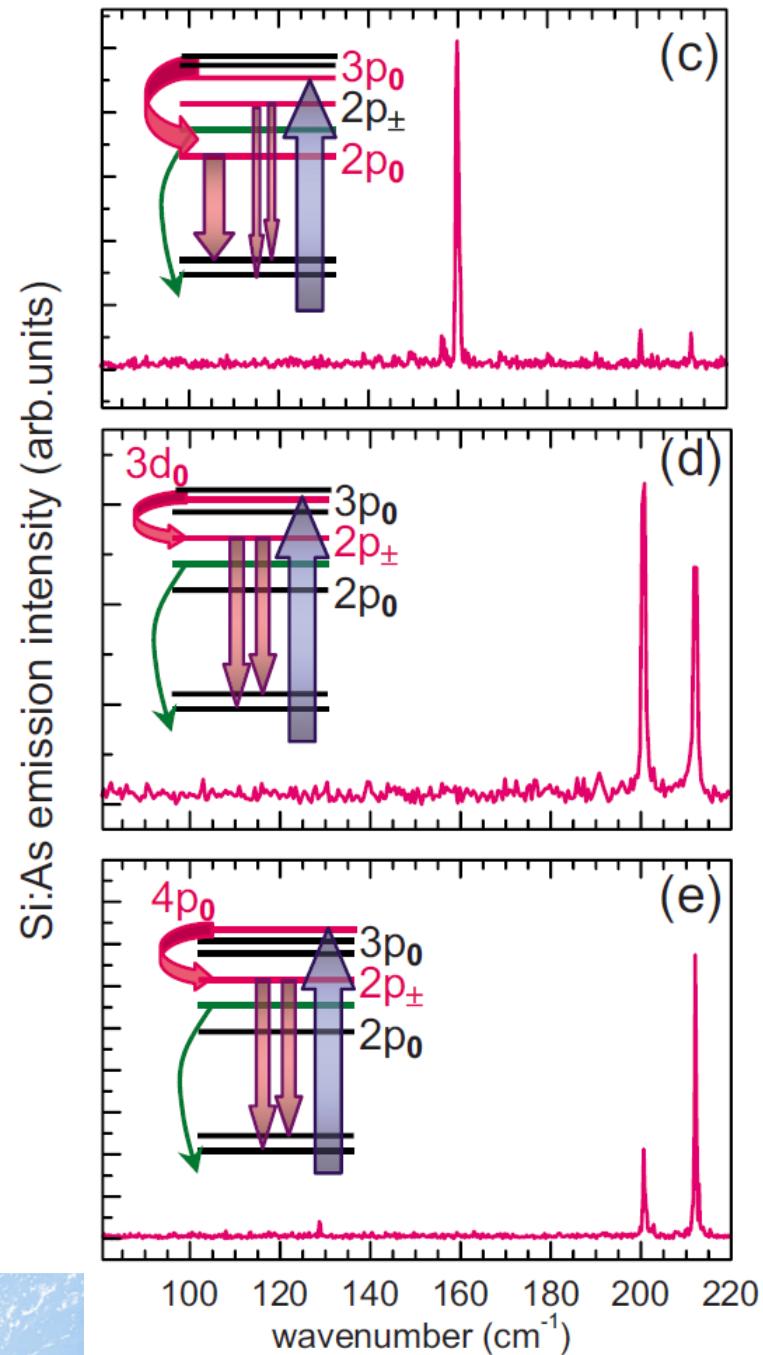


R. Kh. Zhukavin et al., Appl. Phys. B 76, 613 (2003)

# Photoexcitation of Si:As



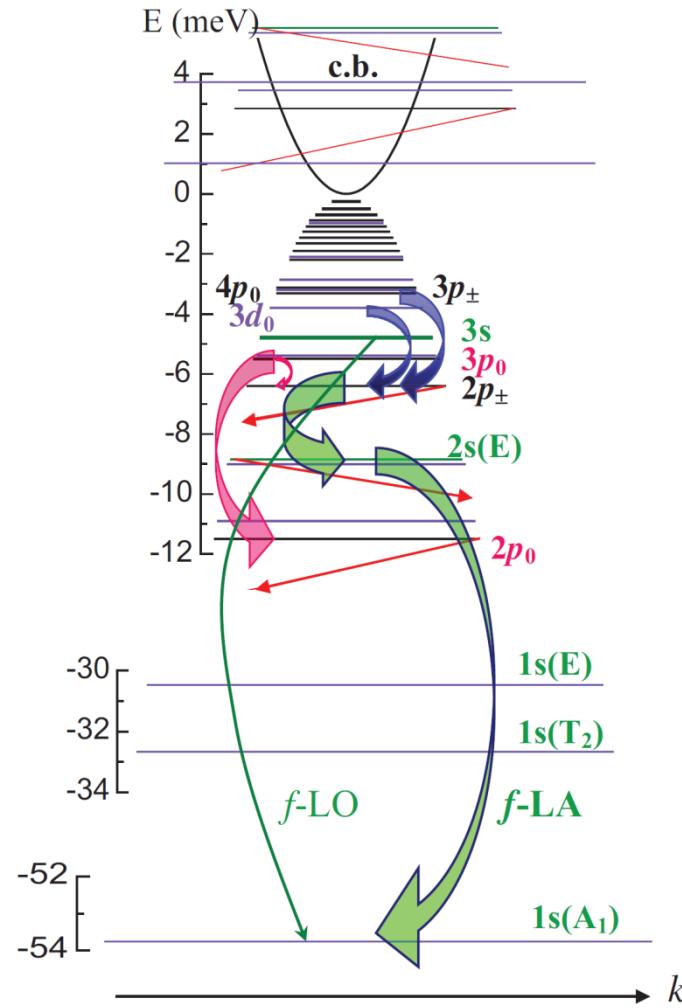
PRB 78, 165201 (2008)



# Photoexcitation of Si:As

# Pumping in

- $2p_0$ :  $2p_0 \rightarrow 1s(E)$
  - $2p_{\pm}$ :  $2p_{\pm} \rightarrow 1s(T_2)$
  - $3p_0$ :  $2p_0 \rightarrow 1s(E)$ ,  
 $2p_{\pm} \rightarrow 1s(T_2)$ ,  
 $2p_{\pm} \rightarrow 1s(E)$
  - $3d_0$ :  $2p_{\pm} \rightarrow 1s(E)$ ,  
 $2p_{\pm} \rightarrow 1s(T_2)$
  - $4p_0$ :  $2p_{\pm} \rightarrow 1s(E)$ ,  
 $2p_{\pm} \rightarrow 1s(T_2)$ ,  
 $3d_0 \rightarrow 1s(E)$ ,  
 $4p_0 \rightarrow 1s(T_2)$



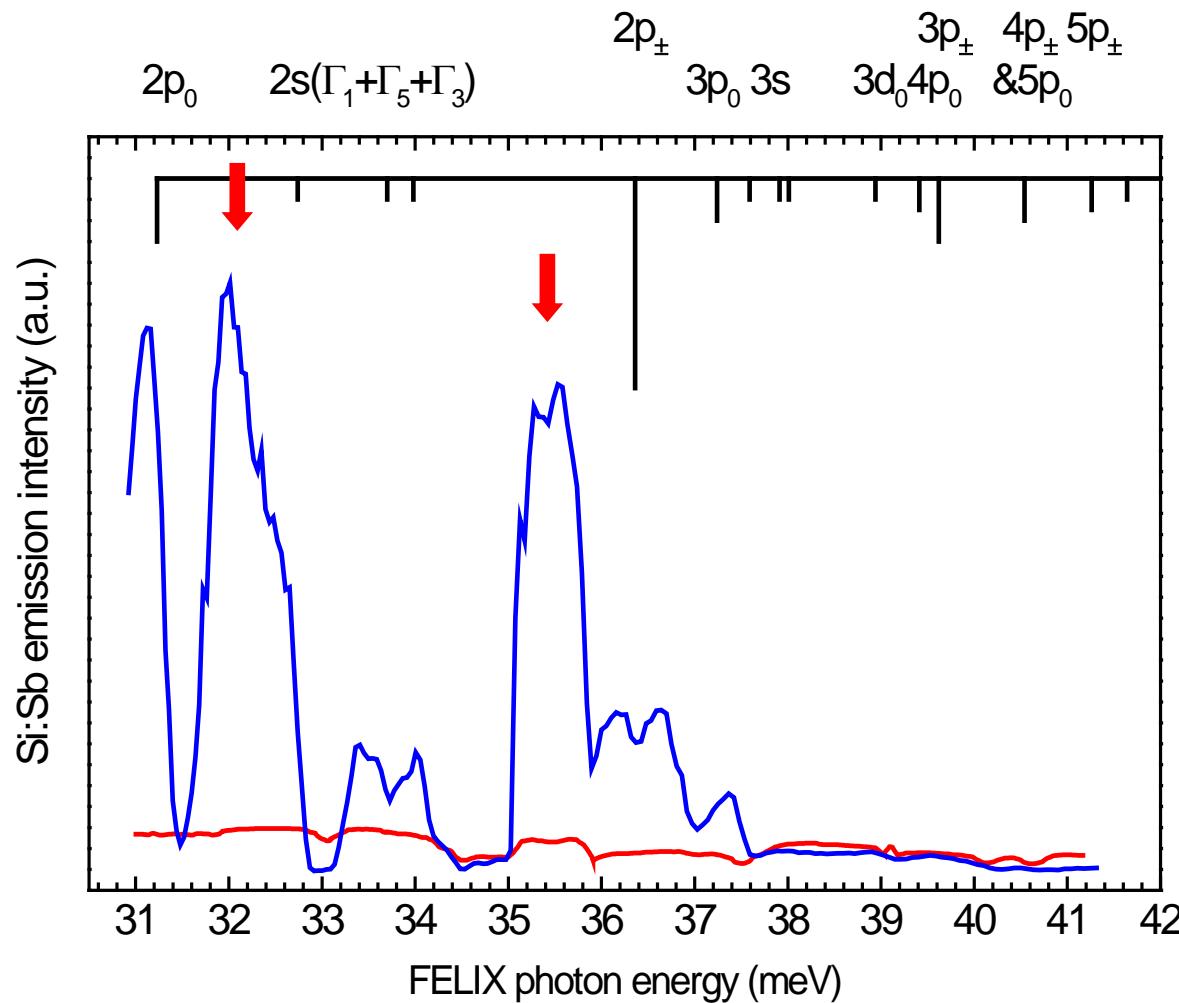
The changes of the laser scheme with changing pump transition indicate that specific relaxation paths exist that are not described by a step-like relaxation from one to the next lower state.

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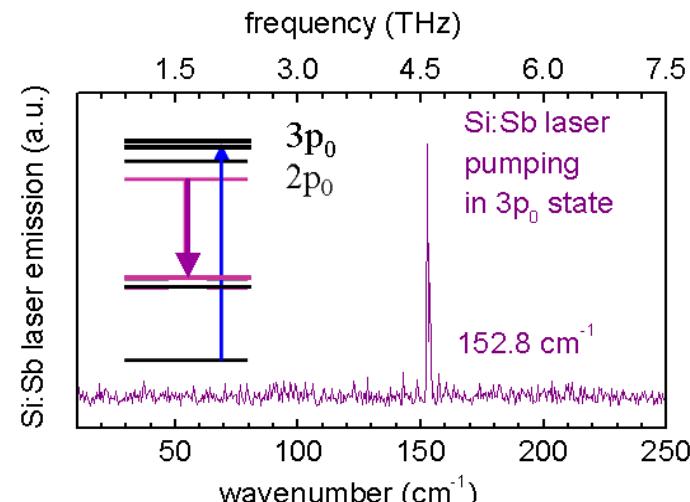
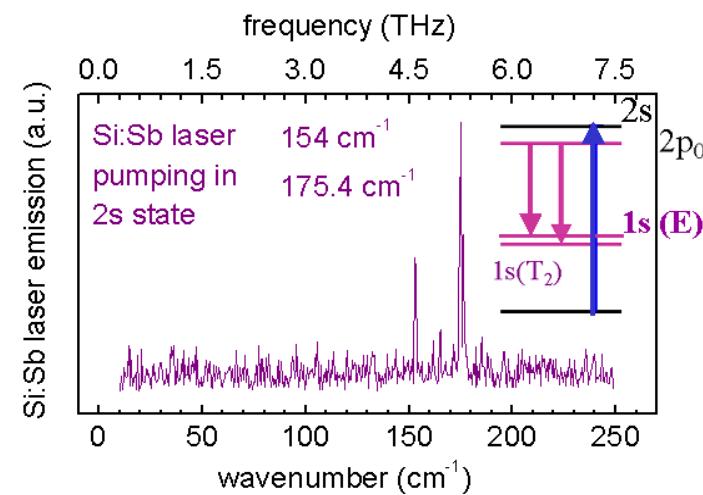
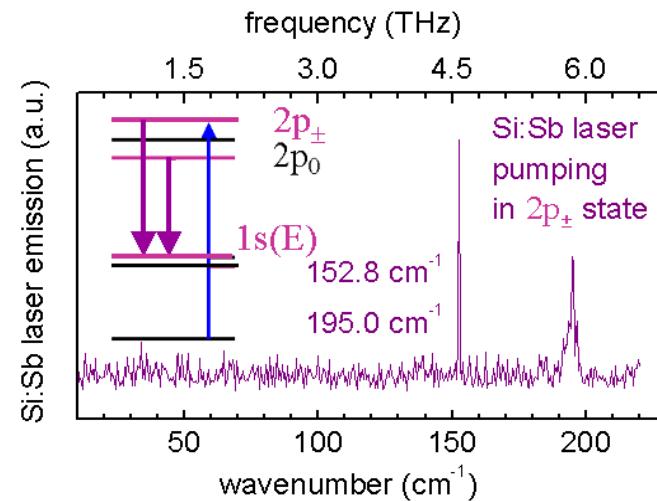
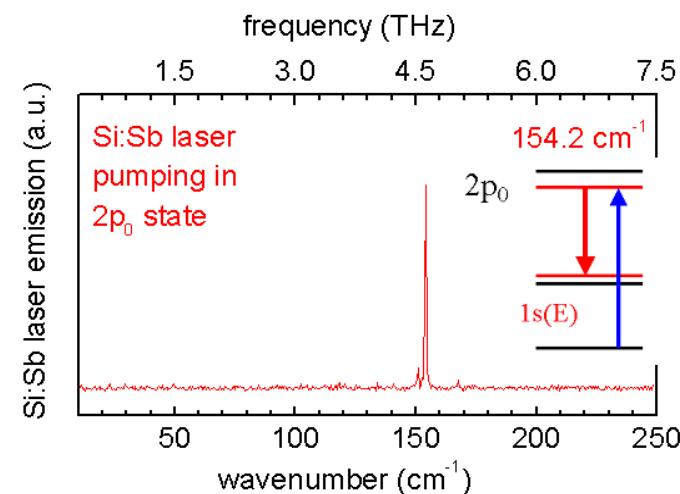


# Photoexcitation of Si:Sb: Non-resonant excitation

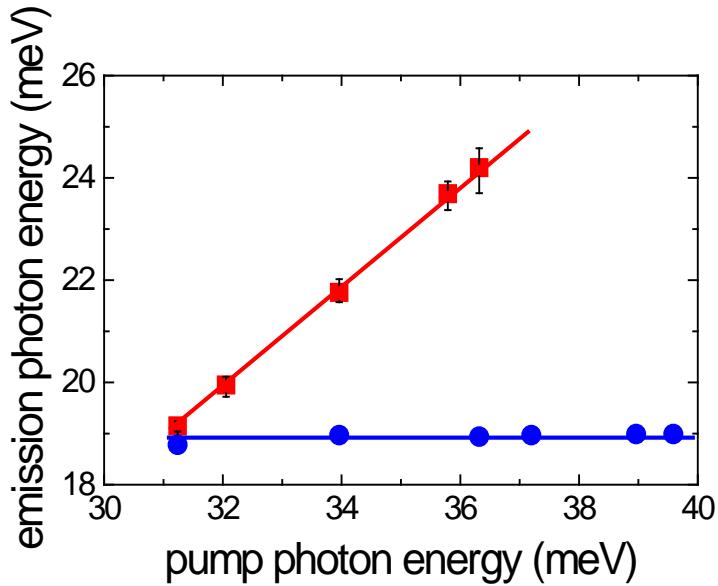


Non-resonant pumping yields laser emission!

# Photoexcitation of Si:Sb

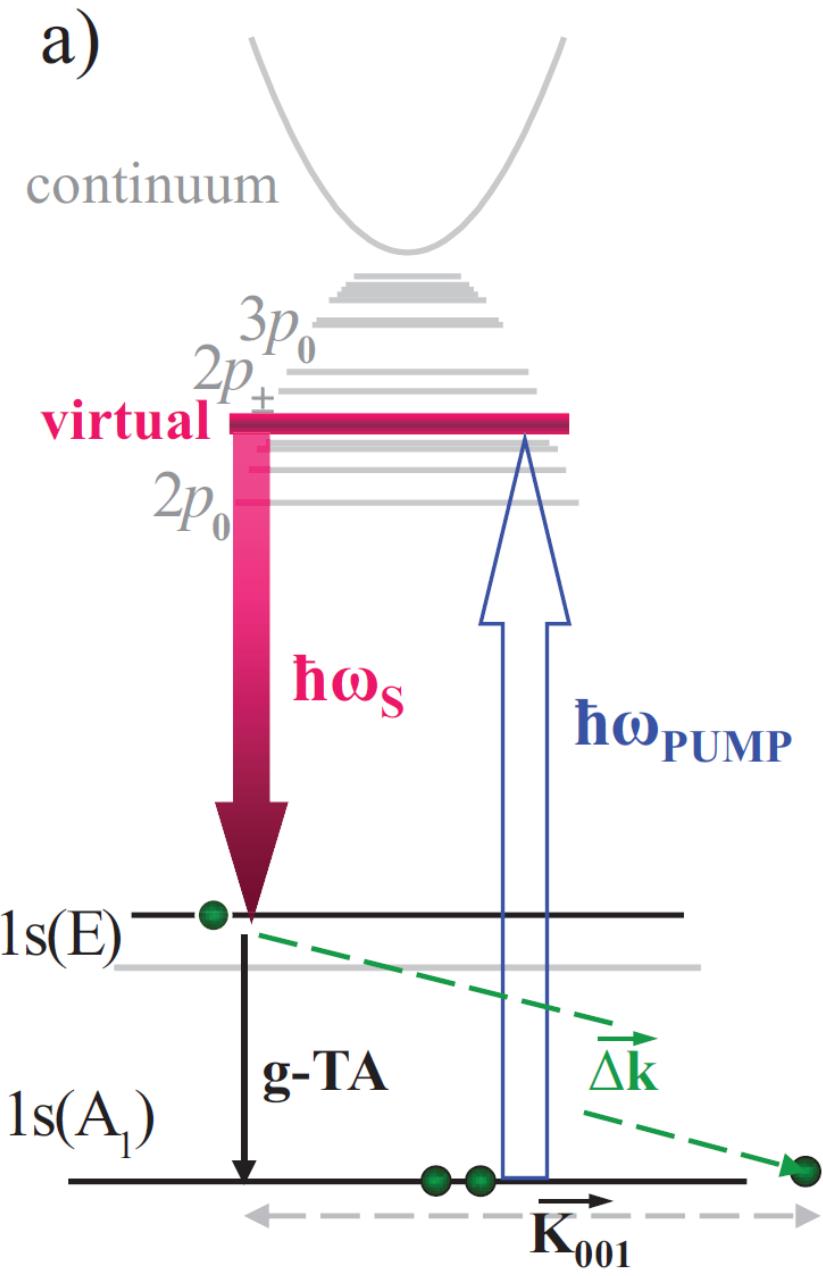


# Si:Sb Raman laser

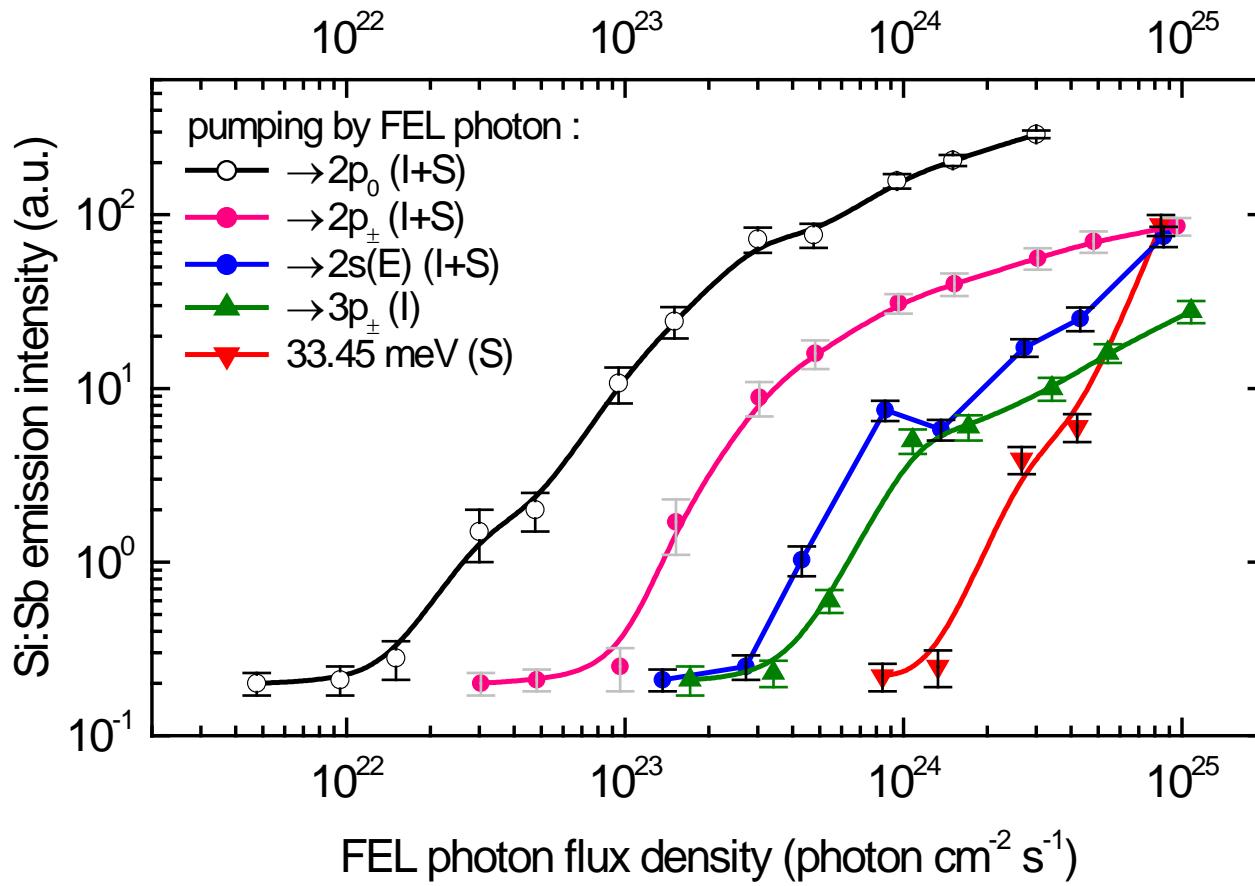


- $\hbar\omega_{\text{emis}} = \hbar\omega_{\text{pump}} - (12.10 \pm 0.02) \text{ meV}$
- g-TA phonon: 11.3 - 12.2 meV

S. Pavlov et al., PRL 96, 037404 (2006)

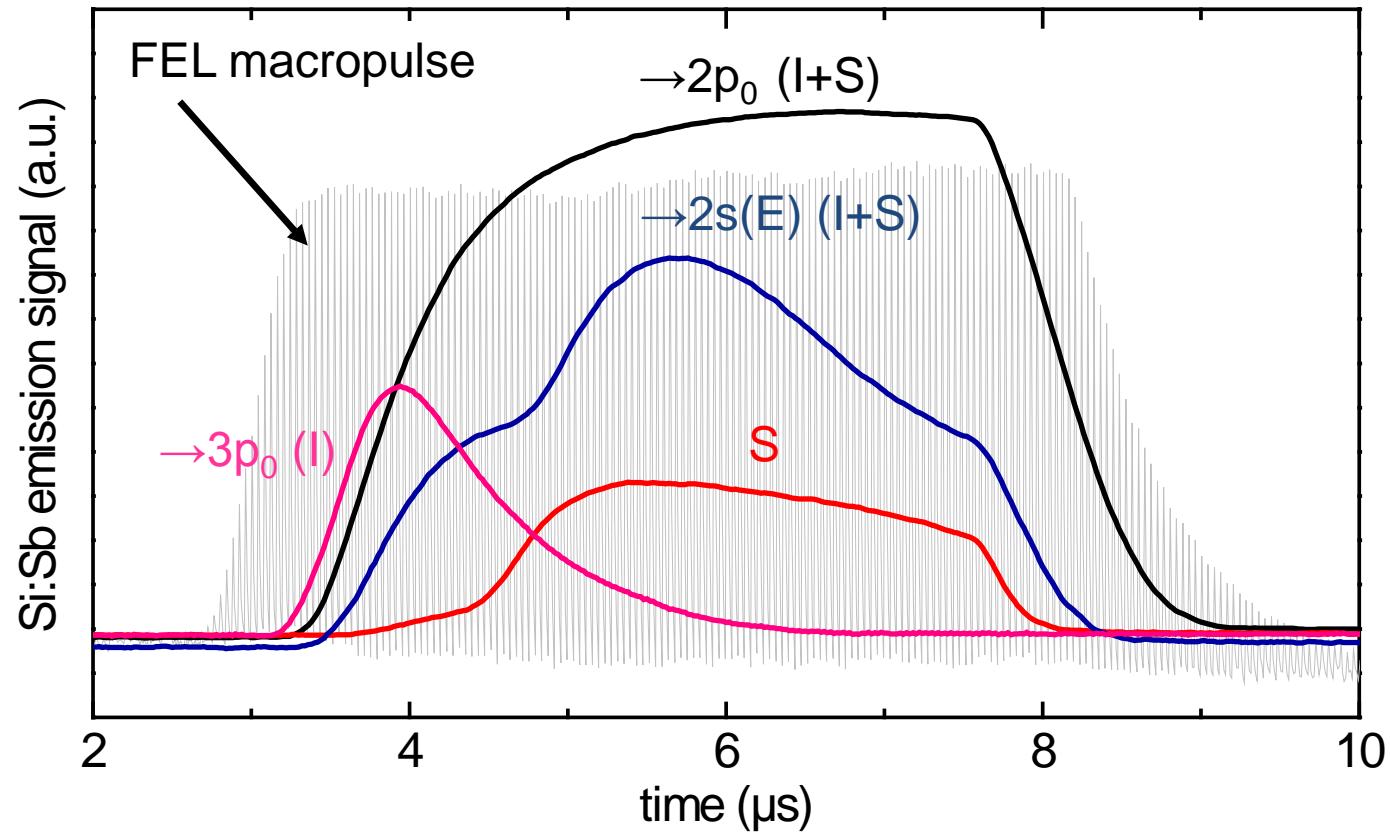


# Laser threshold of Si:Sb Raman laser



- Threshold for Raman laser / Stokes emission is higher than for donor laser.
- Donor laser saturates with increasing pump power, Raman laser does not.

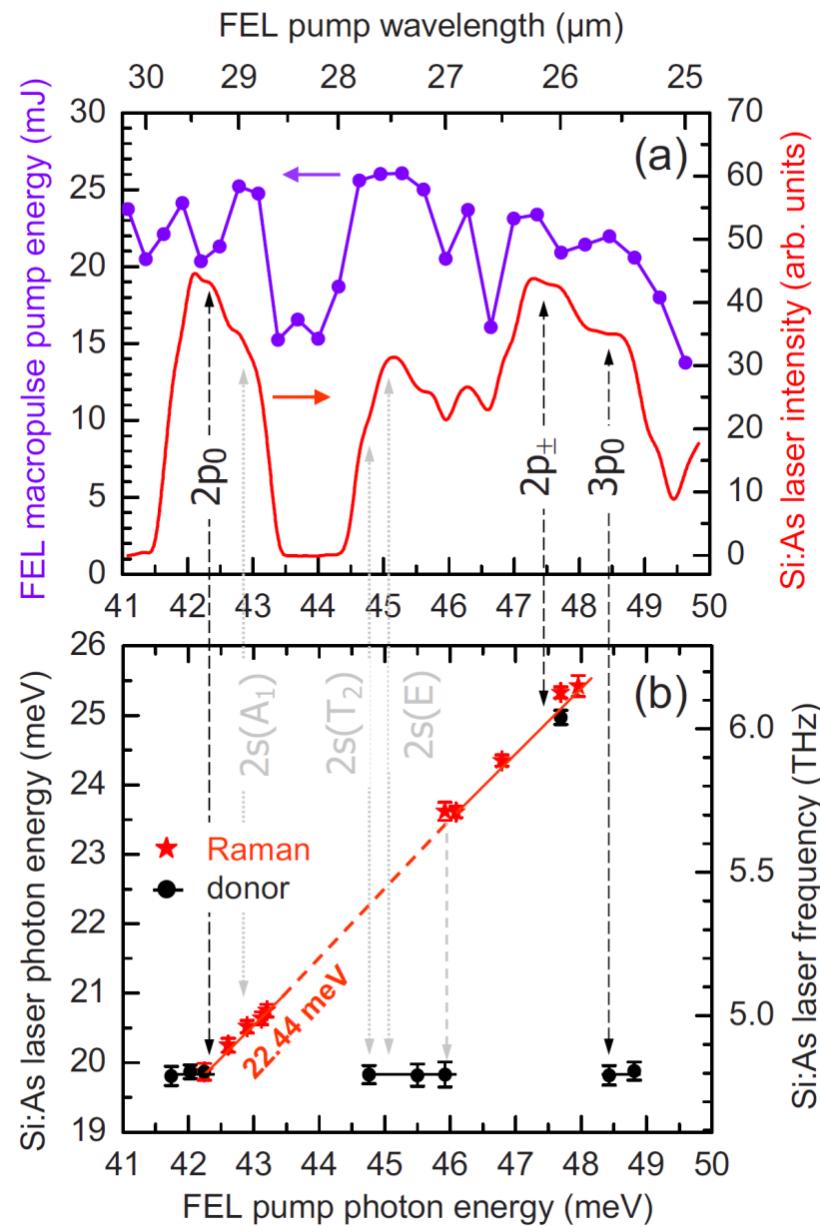
# Emission pulses of the Si:Sb laser



- Raman laser /Stokes emission (S) is delayed with respect to intracenter donor lasing (I).

# Si:As Raman laser

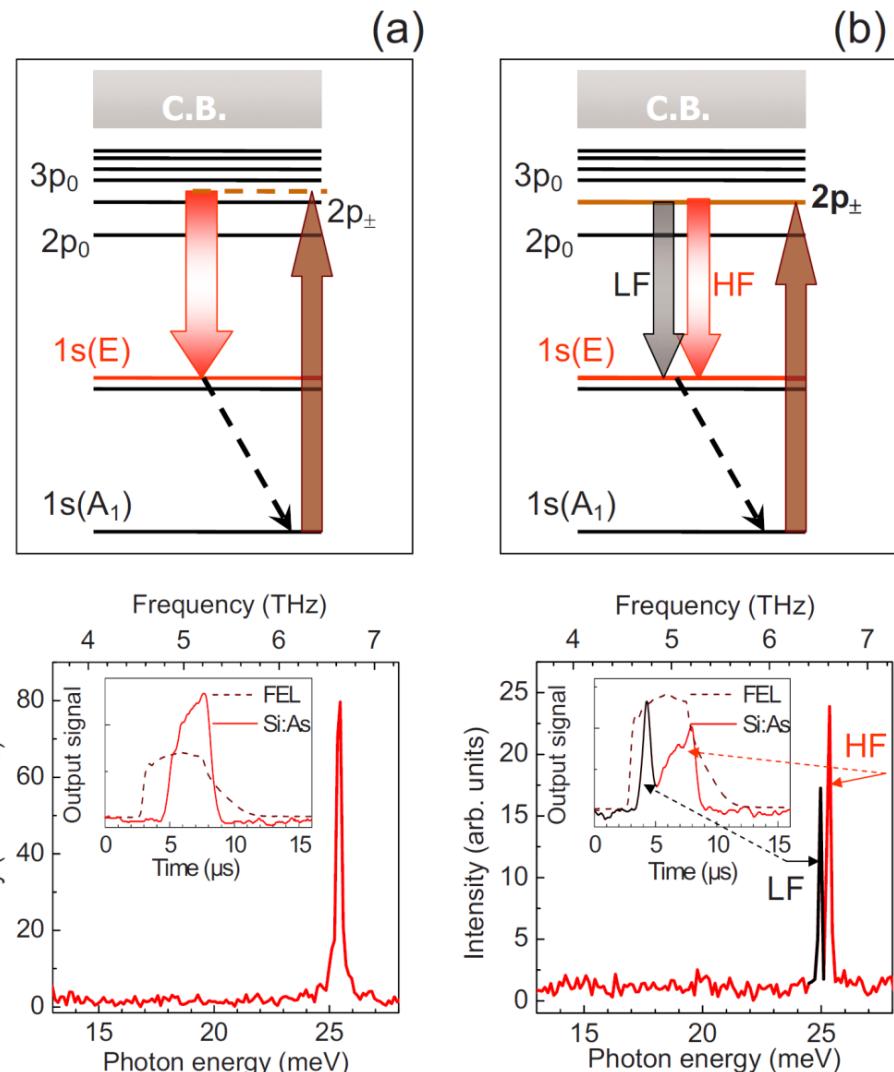
- a) FELIX pump spectrum at maximal power (blue) and laser intensity. The minimum at 44 meV is due to strong absorption of the pump radiation by water vapor in the air.
- b) Emission frequencies of Raman lasing (red) and donor lasing (black). The Stokes shift of the Raman emission is 22.44 meV.



S. Pavlov et al. APL 94, 171112 (2009)

# Dynamics of the Si:As laser

- a) Pumping between  $2p_{\pm}$  and  $3p_0$  states: pure Raman lasing, emission is delayed by about 1.7  $\mu$ s with respect to the pump pulse.
- b) Pumping in vicinity of the  $2p_{\pm}$  state: Combined Raman and donor lasing, donor lasing (black) develops faster (almost no delay with respect to the pump pulse) than Raman lasing (red).



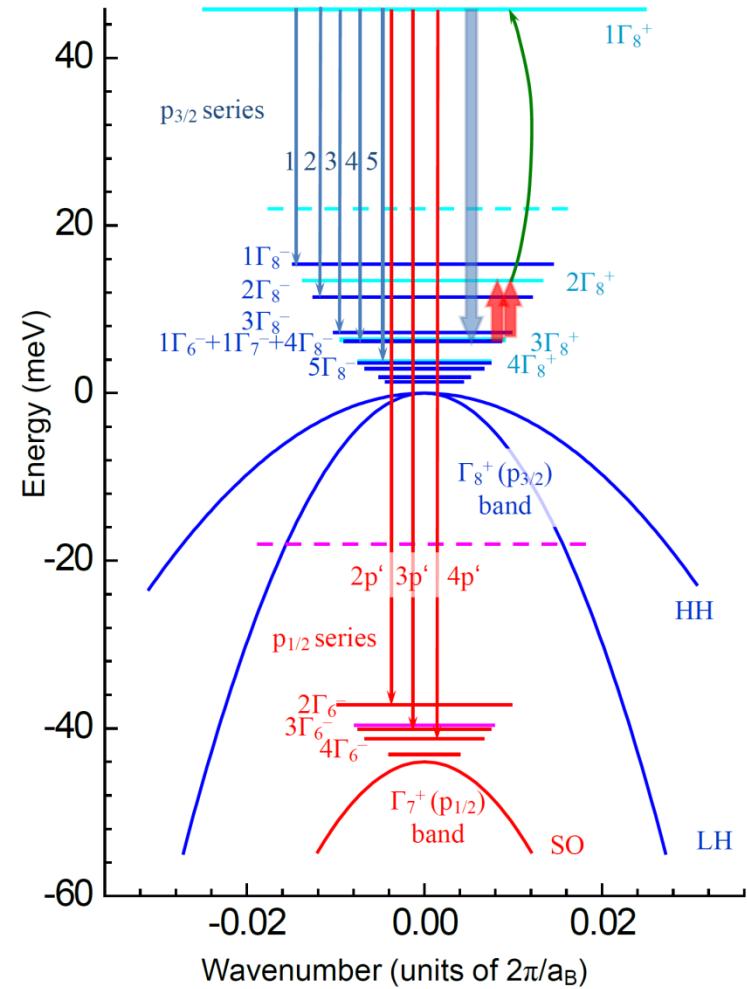
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# Boron-doped Silicon laser

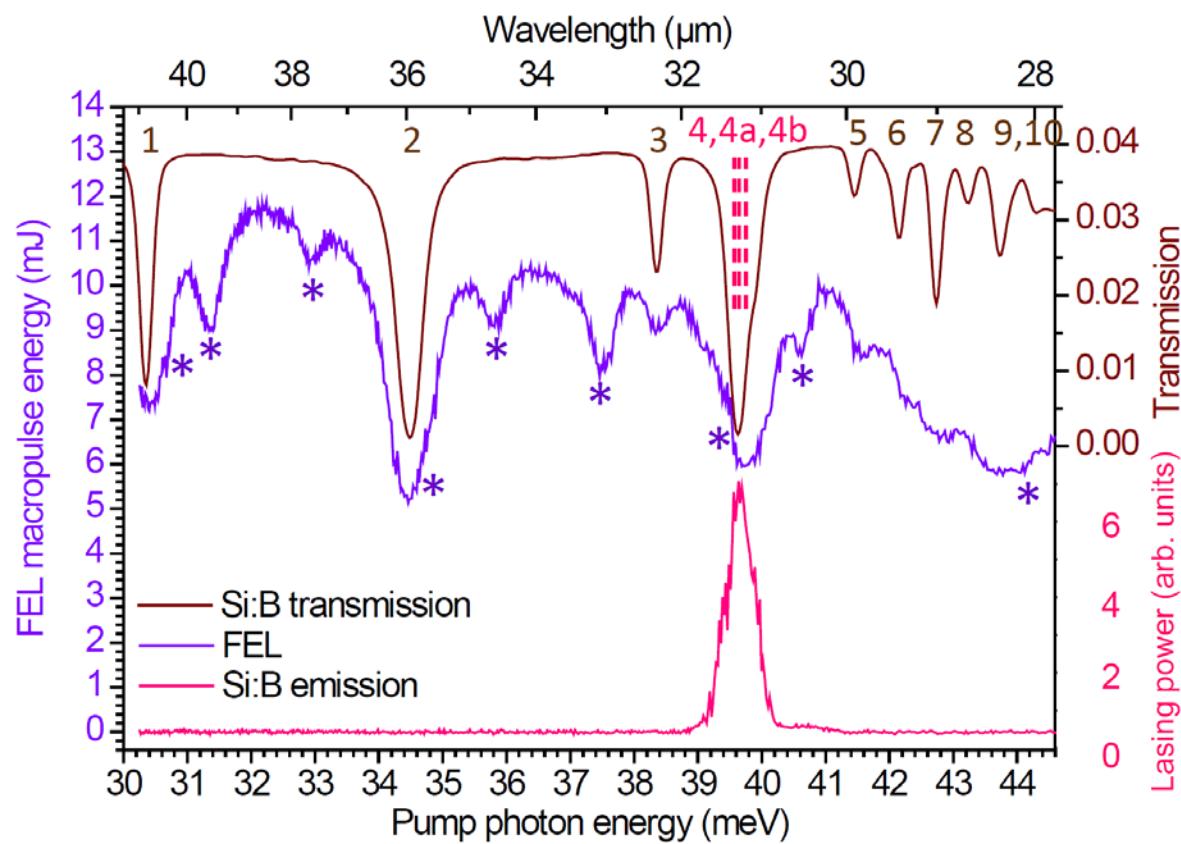
- The  $1\Gamma_7^+$  ground state of the split-off state series ( $\Gamma_7^+$  band) is expected to be between the two horizontal dashed lines.
- Raman spectroscopy:  $1\Gamma_7^+$  ground state is at approx. 22 meV
- IR absorption spectroscopy: state not detected



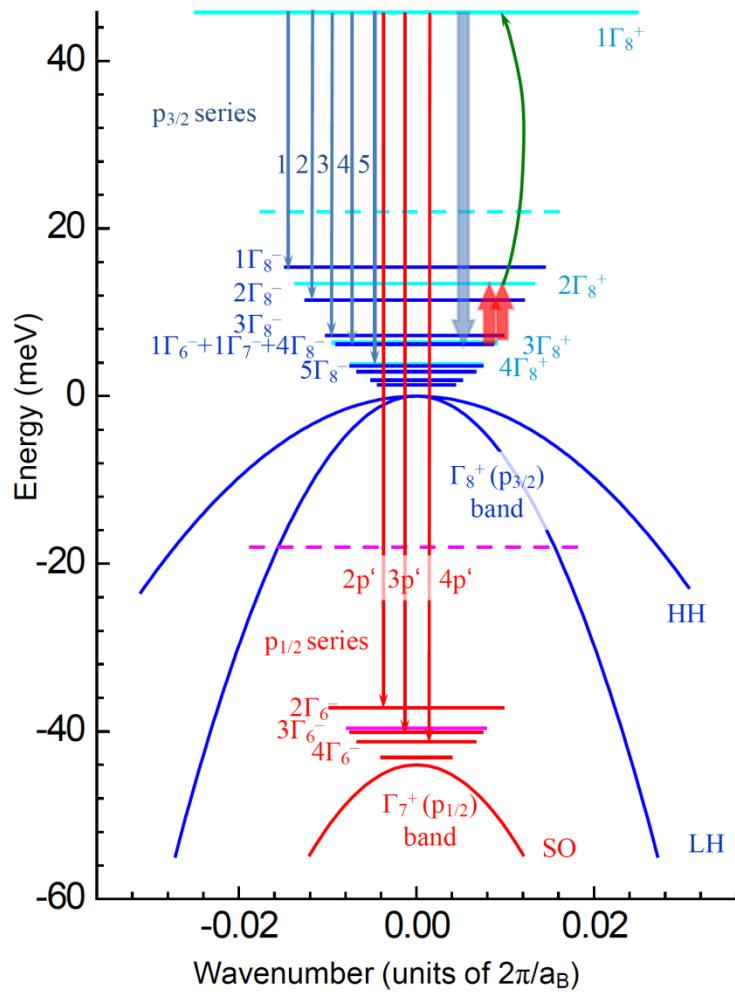
S. Pavlov et al., Phys. Rev. X 4, 021009 (2014)

# Absorption spectrum and laser emission of Si:B

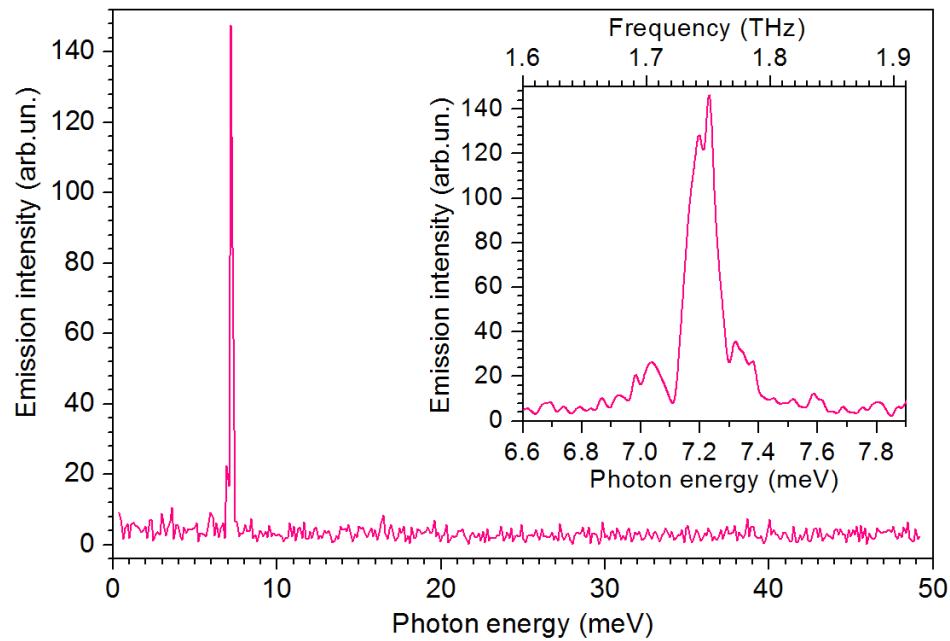
- The absorption spectrum measured with FELIX corresponds to the FTIR spectrum.
- FELIX spectrum is affected by water vapour absorption.
- Only pumping on the boron lines 4, 4a, and 4b results in laser emission.



# Emission frequency and threshold



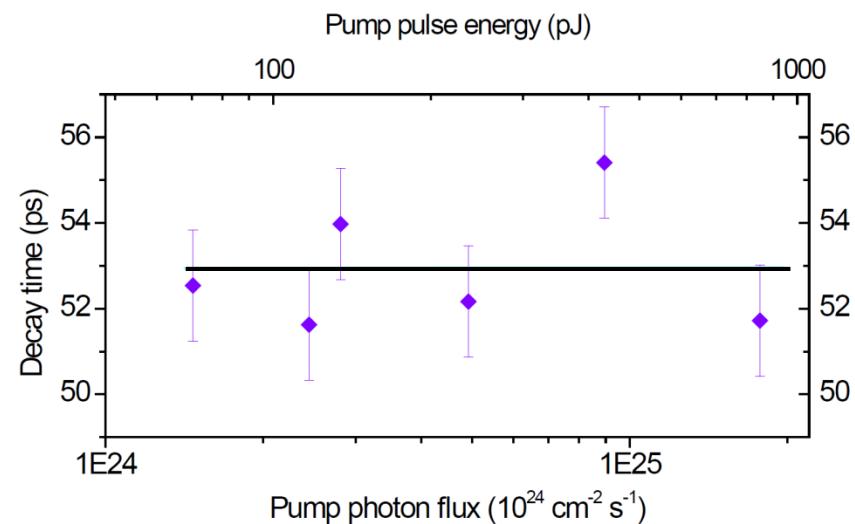
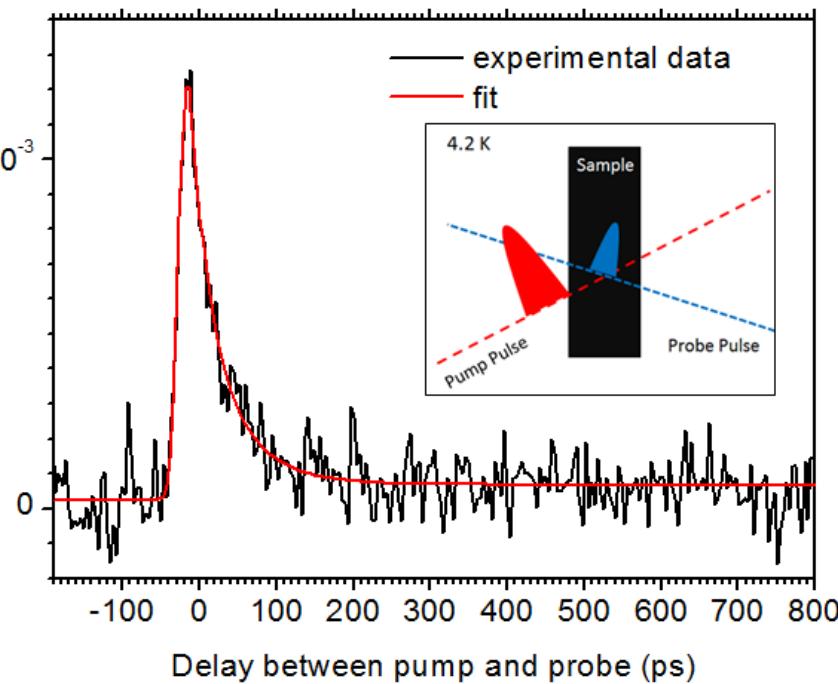
S. Pavlov et al., Phys. Rev. X 4, 021009 (2014)



- Emission spectrum of the Si:B laser when pumped at the boron line 4.
- Two emission lines at 1.740 THz and 1.748 THz are just resolved.
- Upper laser levels: 1Γ<sub>7</sub><sup>-</sup>(1p<sub>5/2</sub>), 1Γ<sub>6</sub><sup>-</sup>(1p<sub>1/2</sub>)
- Lower laser level: 2Γ<sub>8</sub><sup>+</sup>(1p<sub>5/2</sub>)

# Lifetime of the upper laser level

Relative transmission



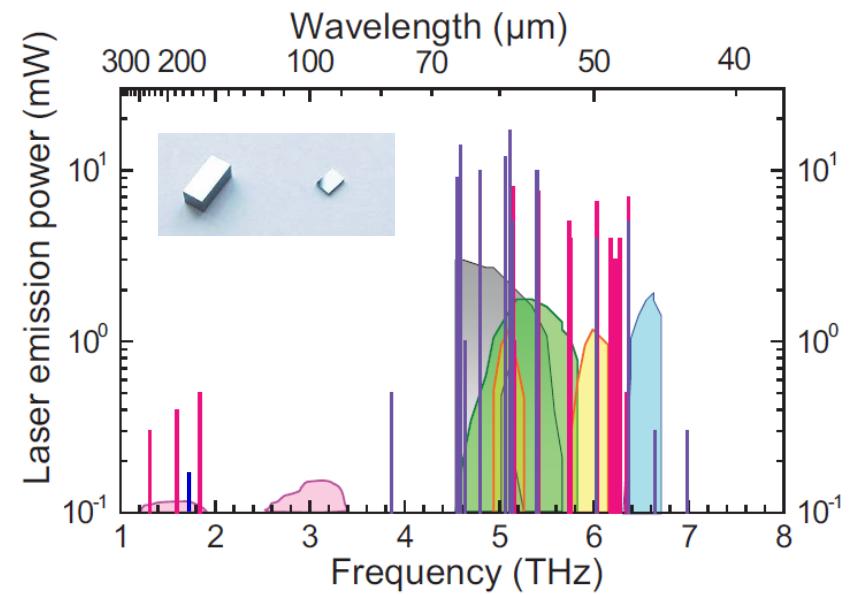
- The short lifetime of the upper laser states (53 ps) limits the laser gain.

# Summary and outlook

- Different types of n-type THz Silicon lasers:
  - Intracenter donor laser
  - Raman laser
- n- and p- type lasers
- Laser mechanism is based on the peculiarities of the electron – phonon interaction in Silicon

Future:

- Apply stress to modify electron-phonon interaction
- Deep donors (move to IR)
- Other host material, e.g. C
- Modelling of life times



**Thank you for your attention!**

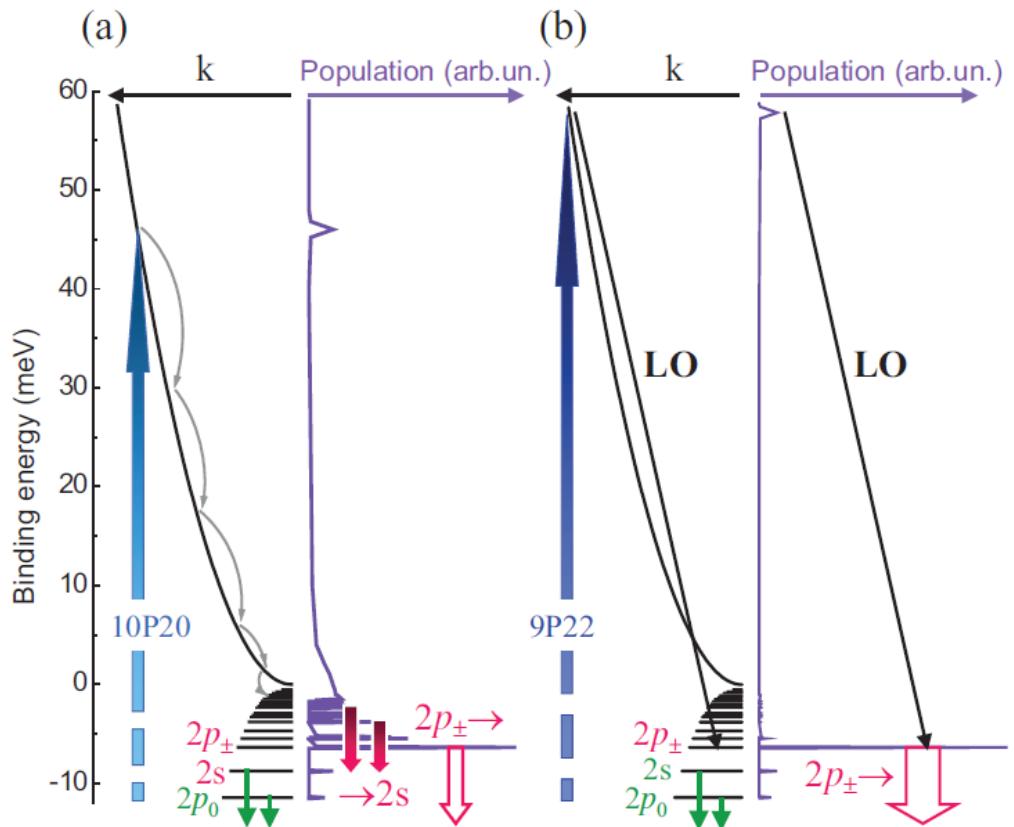
for a review see: S. Pavlov, R. Zhukavin, V. Shastin, H.-W. Hübers, Phys. Status Solidi B 250, 9-36 (2013)

# Backup

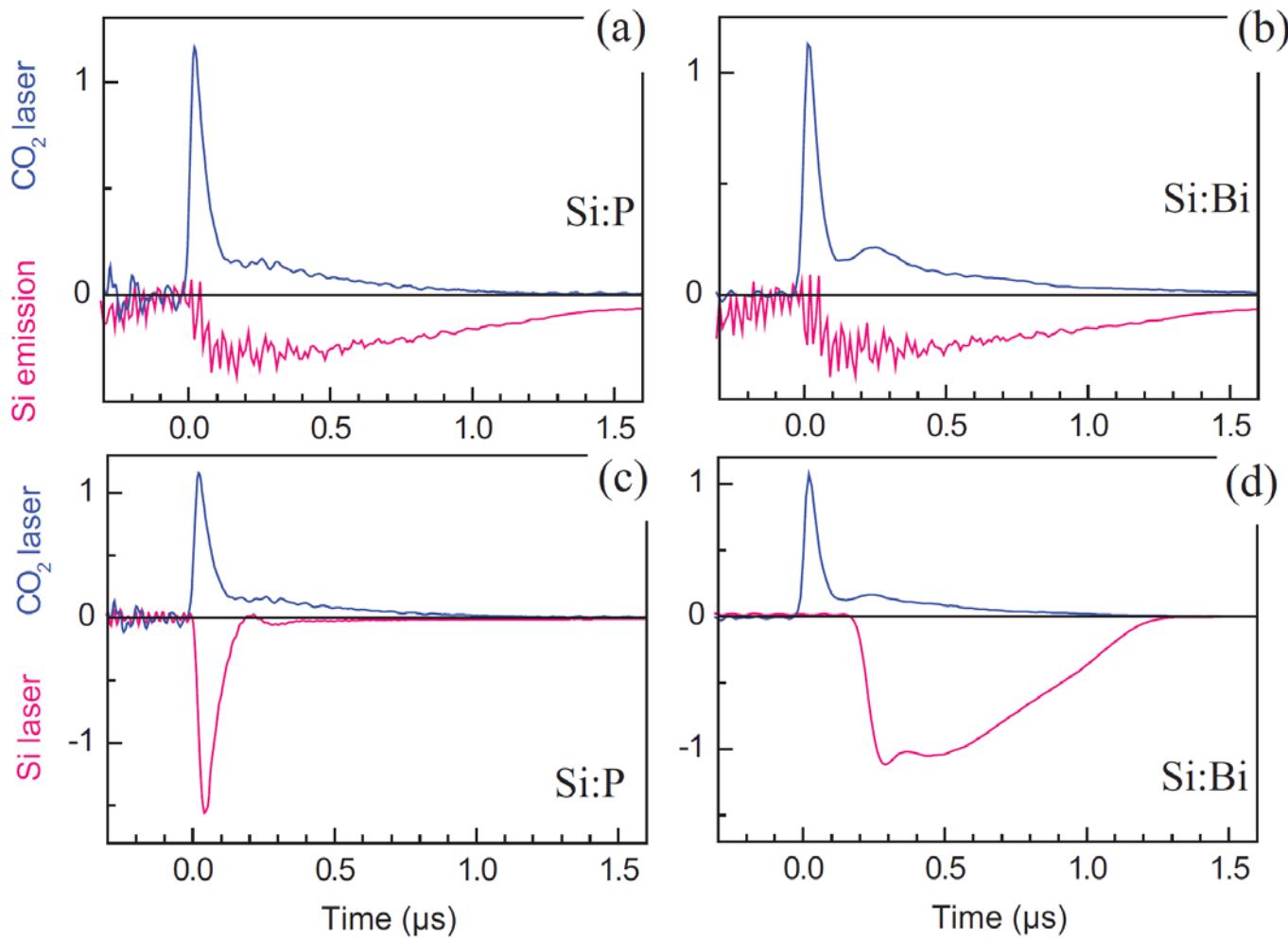


# Si:Bi lasers: long-wavelength (~1.5 THz) emission

- a) Pumping with 10P20 (117 meV)  
 The laser transitions originate from a number of high excited states and terminate in the long-lived 2s state.
- b) Pumping with 9P22 (129 meV):  
 “Shortcut” by the g-LO phonon, only the  $2p_{\pm}$  state has a significant population and the laser transitions are  $2p \rightarrow 1s(E)$ ,  $1s(T_2)$ .



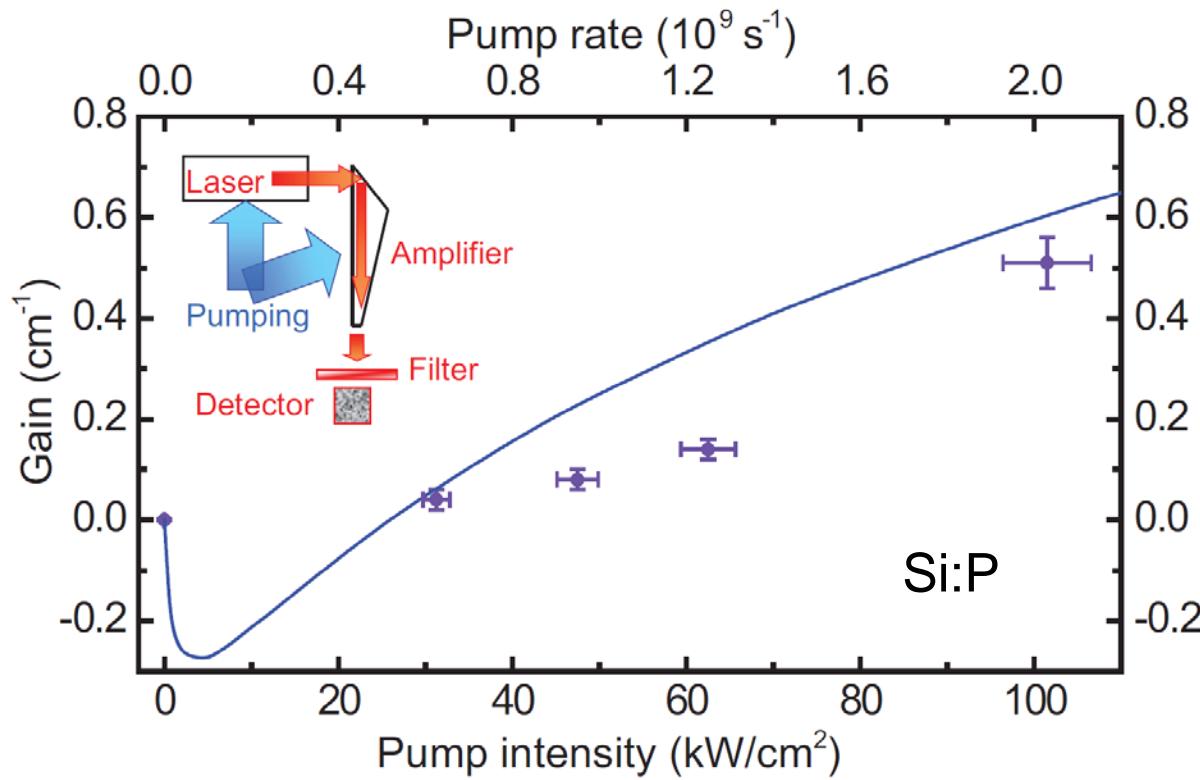
# Laser dynamics



Below laser threshold ( Si emission x1000)

Above laser threshold

# Laser gain

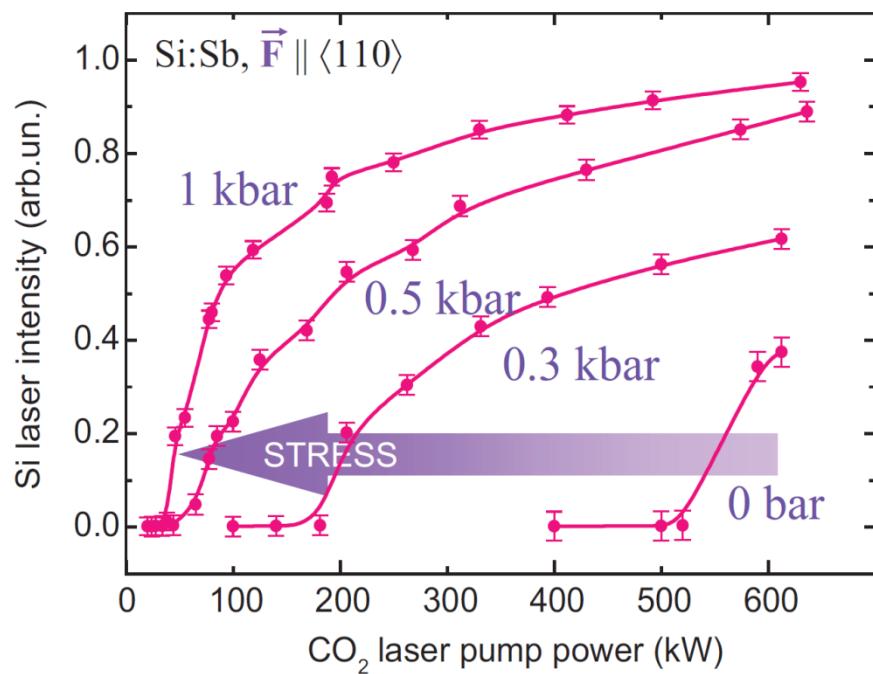


Si:P

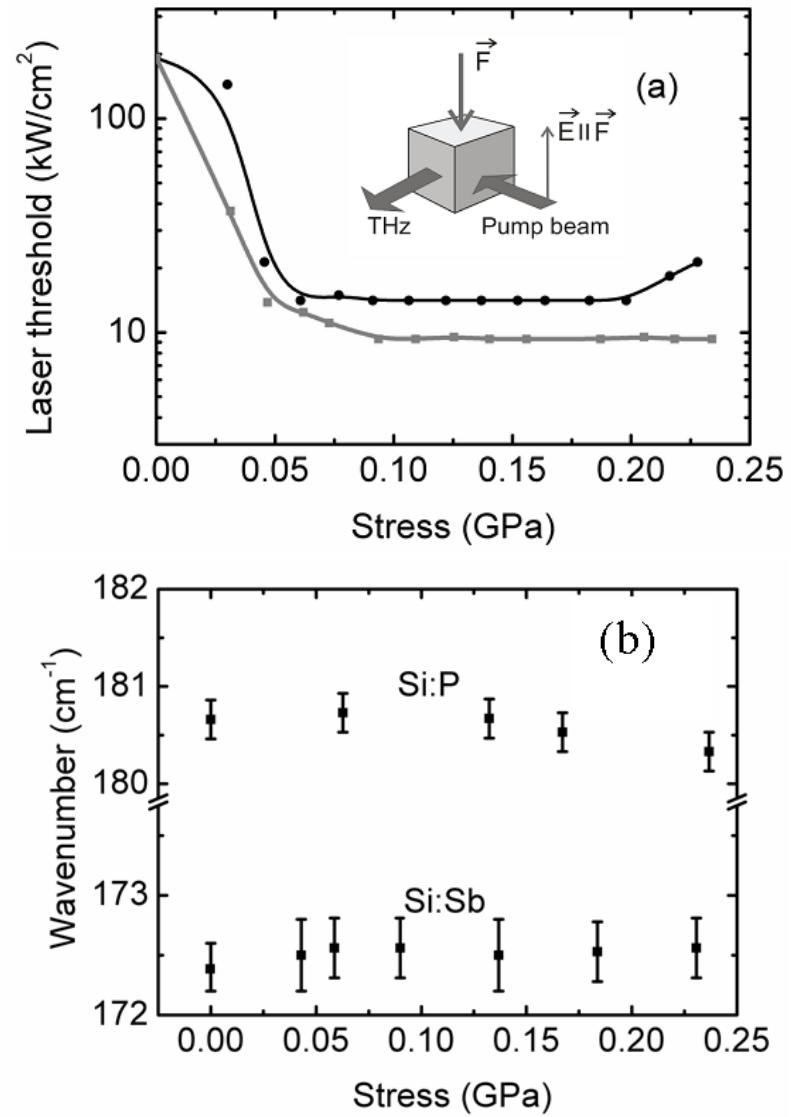
- Gain of Si:P laser: up to  $0.5 \text{ cm}^{-1}$
- Gain of Si:Bi laser:  $0.005 - 0.02 \text{ cm}^{-1}$  (estimated from laser pulse delay, 9P-pumping has higher gain than 10P pumping))

R.Zhukavin et al., JAP 102, 093104 (2007)

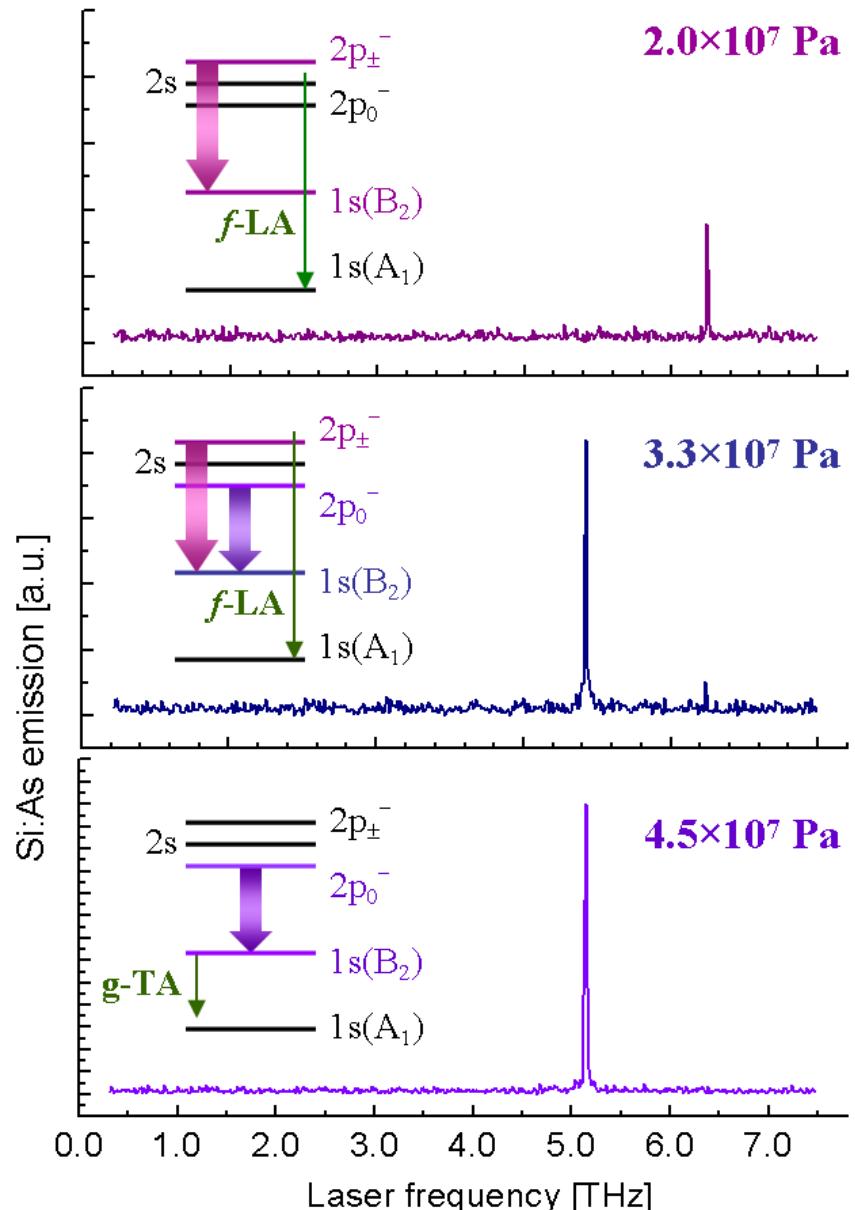
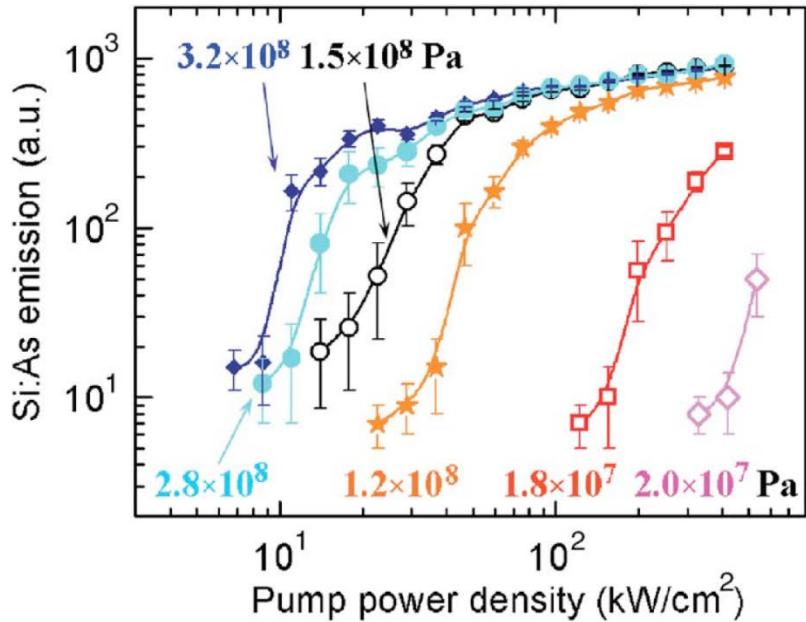
# Compressive force / stress



- Up to 0.1 GPa the laser threshold decreases with stress
- The laser frequency is not affected



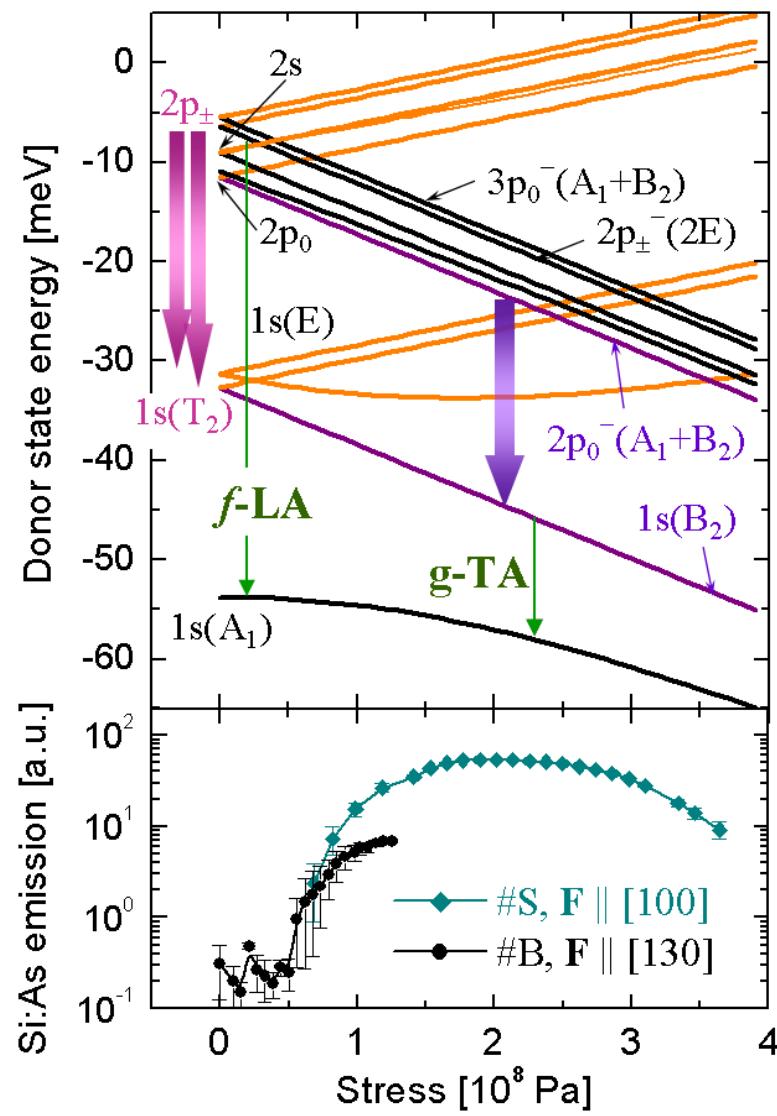
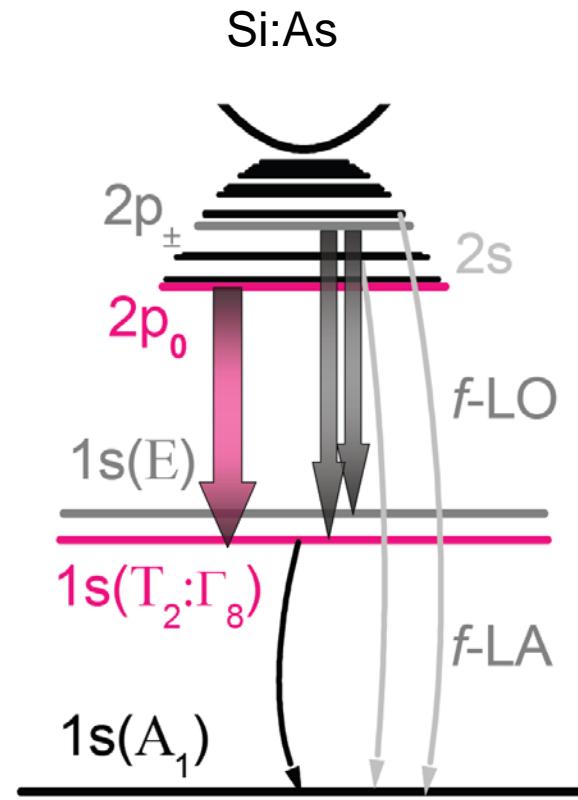
# Stressed Si:As laser



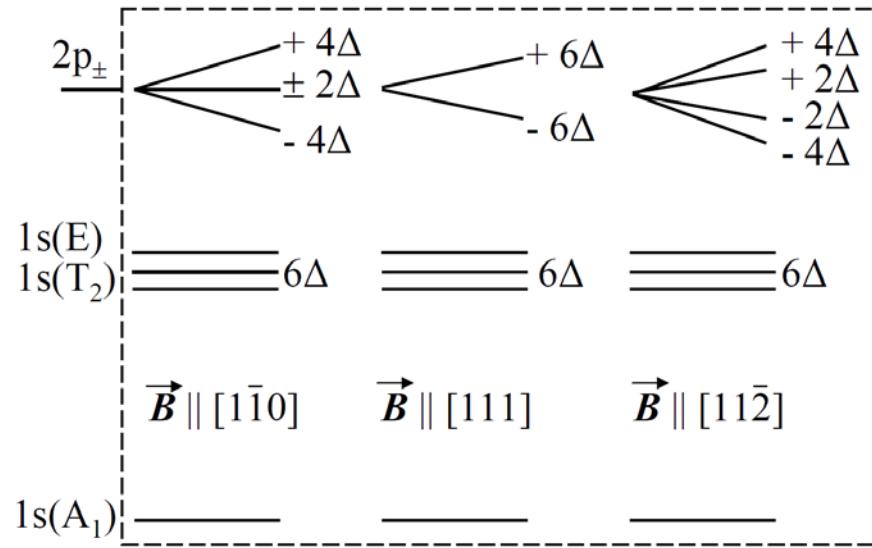
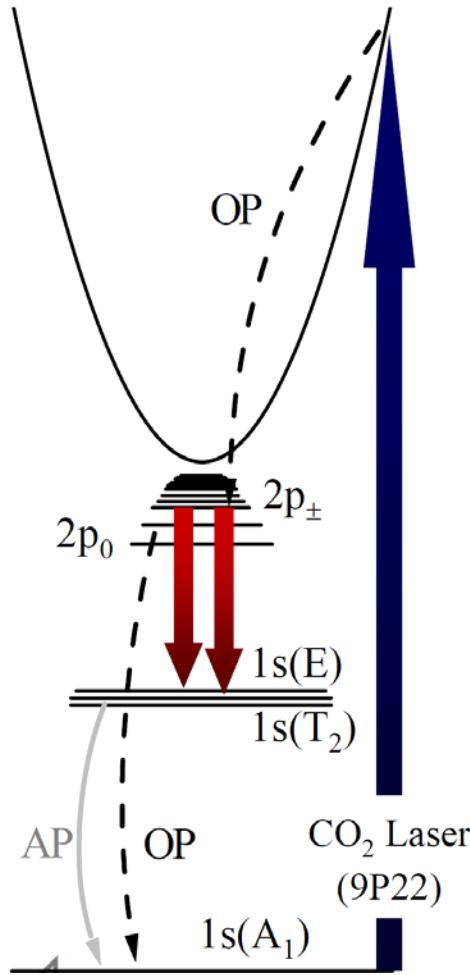
- Lowering of laser threshold by ~100
- Change of emission spectrum
- Si:As laser becomes Si:P-like

APL 94, 171112, (2009)

# Stressed Si:As laser

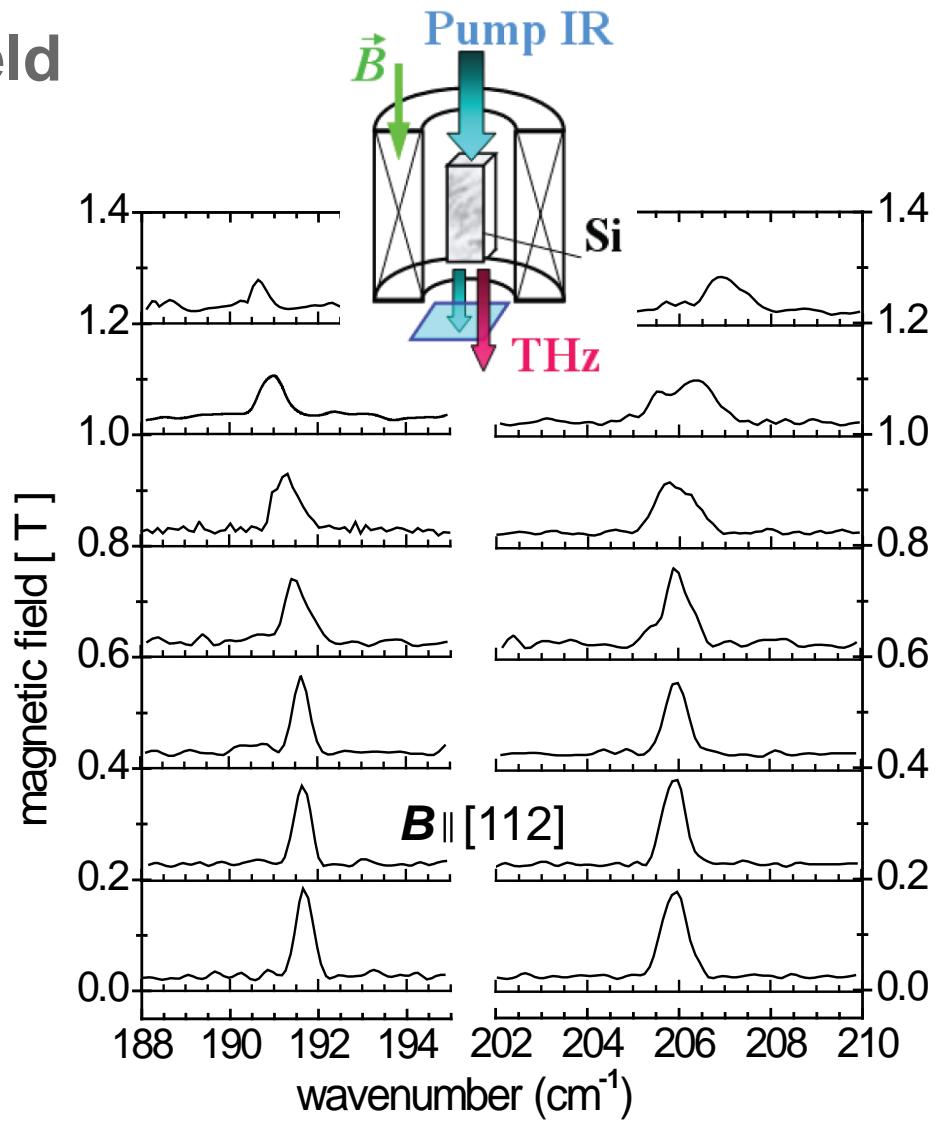
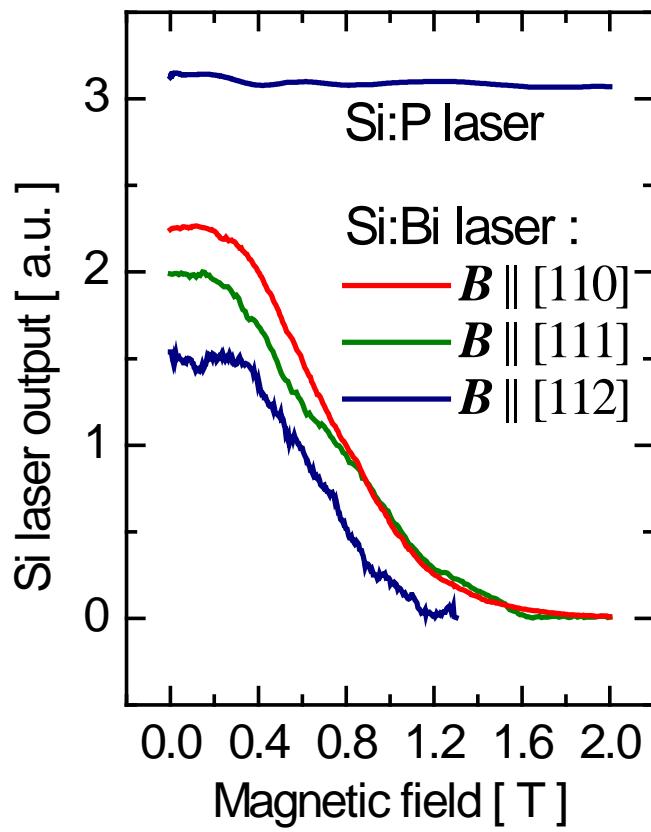


# Si:Bi laser in a magnetic field



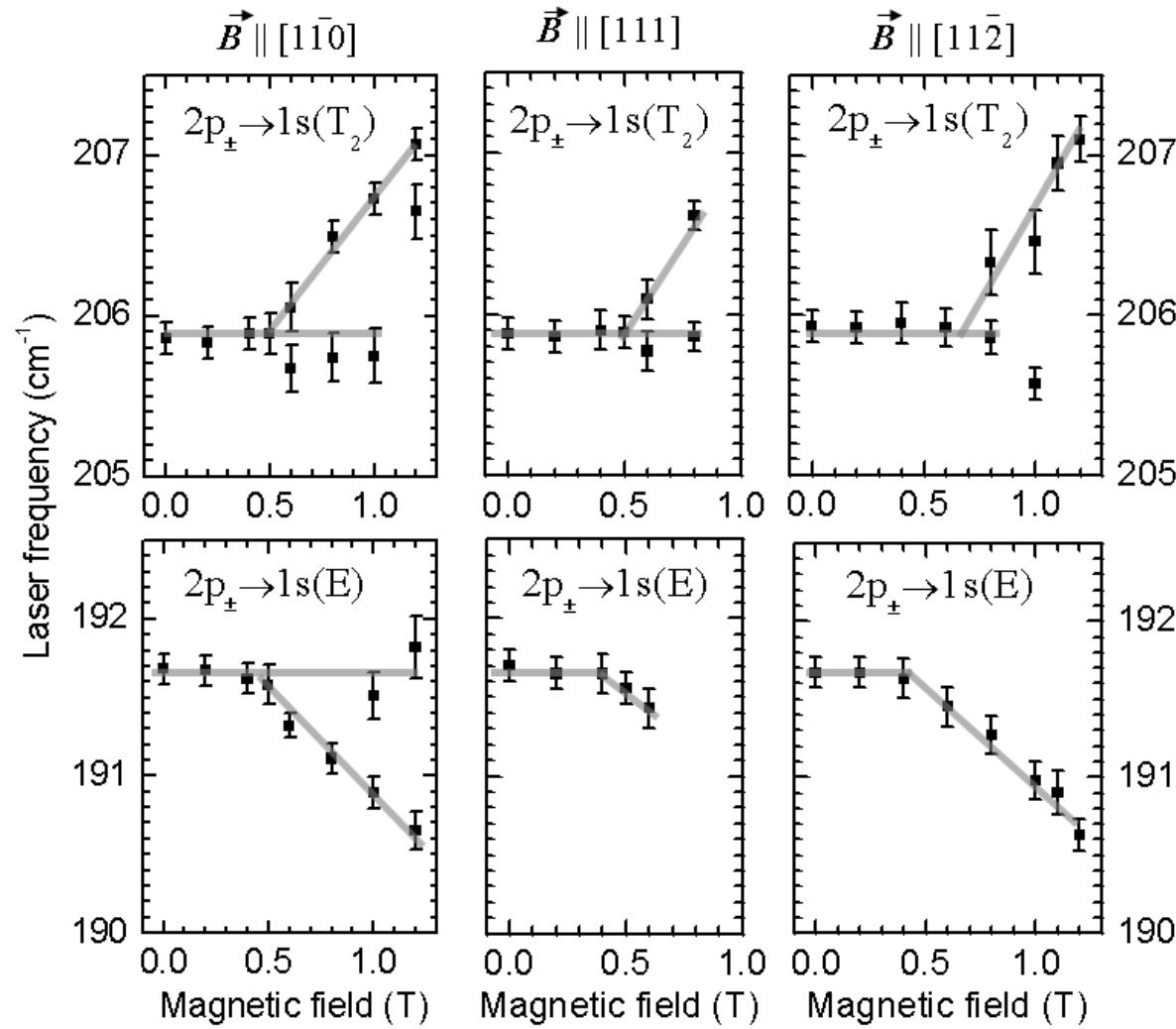
Scheme of the linear Zeeman effect in Si:Bi (not to scale):  
 Splitting of the  $2p_{\pm}$  impurity state; 2, 4, and 6 are the states, which originate from the  $2p_{\pm}$  state (different valleys of the conduction band, different orientations of the magnetic field relative to the crystal axis). Note that all s-type and  $np_0$ -type states do not exhibit splitting.

# Si:Bi laser in a magnetic field



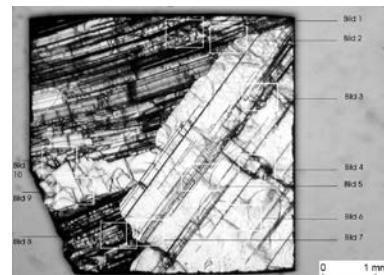
APL 89, 021108 (2006)

# Frequency tunability and crystal orientation



$\partial f / \partial B \sim 40-60 \text{ GHz/T}$   
(expected from linear  
Zeeman effect)

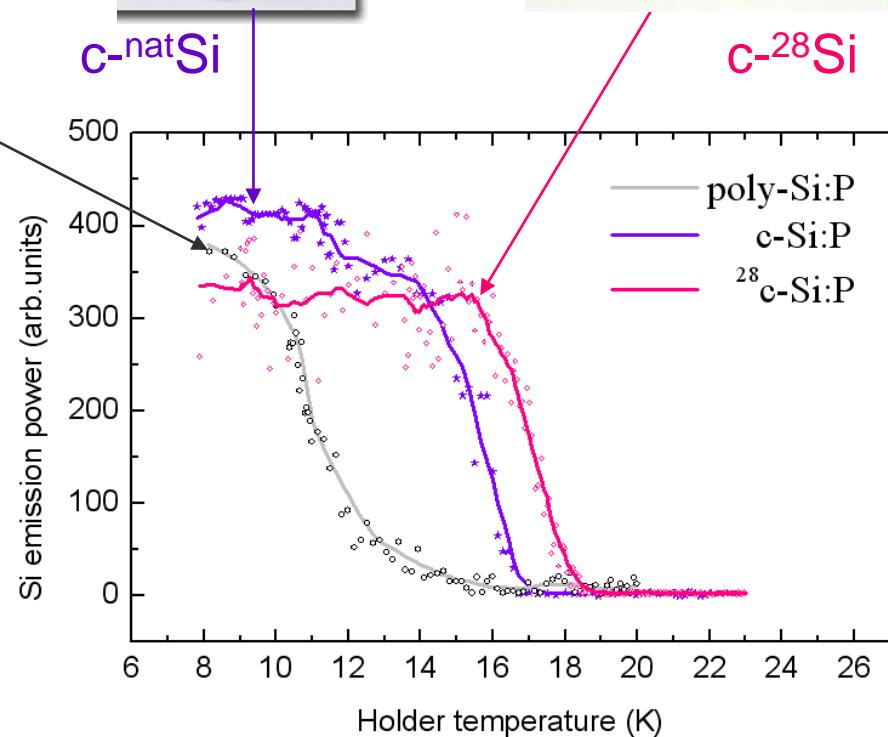
# Si:P lasers based with different crystal quality



mc-Si



- $^{28}\text{Si}$  lasers operate at the highest heat sink temperature(best heat conductivity)
- Polycrystalline Si works also.



unpublished

# Boron-doped silicon

PHYSICAL REVIEW B

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## Spin-orbit splitting of the valence bands in silicon determined by means of high-resolution photoconductive spectroscopy

Zhiyi Yu, Y. X. Huang, and S. C. Shen

*Laboratory for Infrared Physics, Shanghai Institute of Technical Physics, Academia Sinica,  
420 Zhong Shan Bei Yi Road, Shanghai, China*

(Received 8 November 1988)

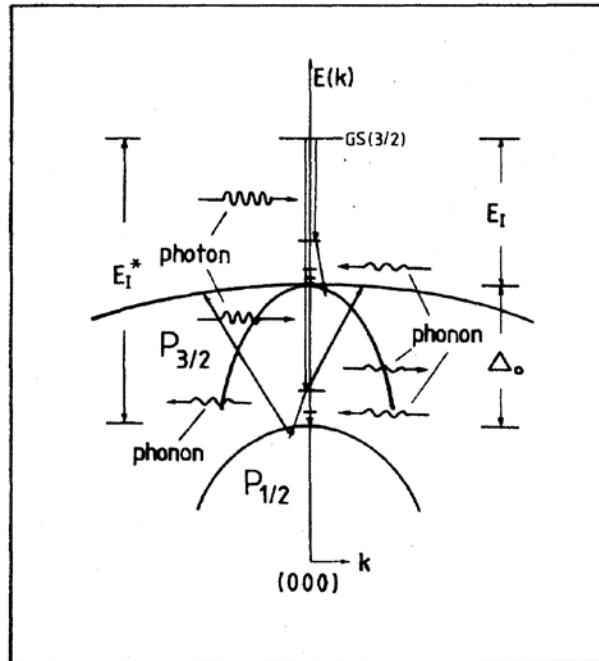


FIG. 1. Schematic diagram showing photoconductive processes of an acceptor in silicon.

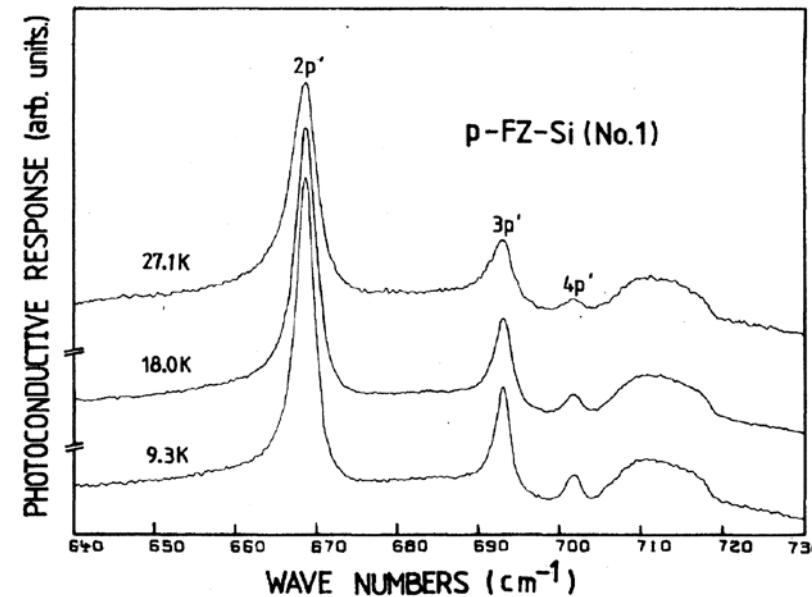


FIG. 2. High-resolution photoconductive spectra of sample No. 1 (room-temperature resistivity is  $1000 \Omega \text{ cm}$ ) at 9.3, 18.0, 27.1 K. (The continuous bands of the curves have exactly the same shape for demonstrating the temperature dependence of the peaks). Instrumental resolution is  $0.25 \text{ cm}^{-1}$ .

