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

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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

n-Silicon:
- 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)
  \[ E_n = - \frac{R}{n^2} \varepsilon^2 \left( \frac{m_D}{m} \right) \]
- Chemical splitting depends on donor
- Spin-orbit coupling

Mayur et al., PRB 1993
Spectroscopy and life times

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

Si:Bi $\tau_{2p0} \sim$ a few ps

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

Si:P $\tau_{2p0} \sim$ several 100 ps

long life times of impurity states

short life times of impurity states
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 = $2 \times 10^{15}$ cm$^{-3}$
- $T_1^{(28}\text{Si}) = 235$ ps > $T_1^{(\text{nat}\text{Si})} = 205$ 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.
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Interaction with phonons

**Si:**

- **Sb**
- **P**
- **As**
- **Bi**

**No phonon!**

- **3p₀**
- **2s**
- **2p₀**

- **1s(E)**
- **1s(T)**
- **1s(A₁)**

**Lasers based on long-living 2p₀ state**

\[ \tau_{2p₀} \approx 10^{-10} \, \text{sec} \]

\[ \tau_{1s\text{-split}} \approx 10^{-11} \, \text{sec} \]

**Conduction band**

**Photoionization by radiation from a CO₂ laser**

**Intracenter excitation by radiation from FELIX**

**Lasers based on resonant electron-phonon interaction**

**f-LO, g-LO, g-TO**

**g-TA**

**f-LA, f-LO**
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 ~ $10^{15} - 10^{16}$ cm$^{-3}$, compensated < 30 % length is 5-15 mm FZ or CZ grown
Photoexcitation with a CO₂ 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$_2$ 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.

<table>
<thead>
<tr>
<th>donor</th>
<th>1s(T&lt;sub&gt;2&lt;/sub&gt;)</th>
<th>1s(E)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>n-Si lasing</td>
<td>cal.</td>
</tr>
<tr>
<td>P</td>
<td>33.91 ± 0.02 (4 K)</td>
<td>34.2</td>
</tr>
<tr>
<td>Sb</td>
<td>32.83 ± 0.02&lt;sup&gt;a&lt;/sup&gt; (4 K)</td>
<td>32.9</td>
</tr>
<tr>
<td></td>
<td>33.12 ± 0.01&lt;sup&gt;b&lt;/sup&gt; (30 K)</td>
<td></td>
</tr>
<tr>
<td>As</td>
<td>32.73 ± 0.02 (4 K)</td>
<td>32.7</td>
</tr>
<tr>
<td>Bi</td>
<td>31.90 ± 0.02&lt;sup&gt;a&lt;/sup&gt; (4 K)</td>
<td>31.3</td>
</tr>
<tr>
<td></td>
<td>32.63 ± 0.01&lt;sup&gt;b&lt;/sup&gt; (4 K)</td>
<td></td>
</tr>
</tbody>
</table>

<sup>a</sup>Doublet 1s(T<sub>2</sub>;Γ<sub>8</sub>); <sup>b</sup>singlet 1s(T<sub>2</sub>;Γ<sub>7</sub>). 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 ∼ 10 K [93]; cal.: as calculated by EMT with an empirical 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.
Si samples at optimum doping concentration ($3 \times 10^{15}$ cm$^{-3}$)
- Pumping at 10.59 µm, 100 kW/cm$^2$ = $5 \times 10^{24}$ photons cm$^{-2}$s$^{-1}$
- Si:P, Si:Sb: $2p_0 \rightarrow 1s(T_2)$
- 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$^2$
- Si:As, Si:Bi: $2p_\pm \rightarrow 1s(E), 1s(T_2)$, pumping at 9.6 µm, 80 kW/cm$^2$
- Upper limit: impurity broadening, lifetime decreases
- Lower limit: too few donors, less gain
• The larger the gap between the 1s(A) ground state and the 1s(E), 1s(T₂) 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: $2p_0 \rightarrow 1s(T_2)$

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 laser pumping in 2p₀ state
- 154.2 cm⁻¹
- 1s(E)

- Si:Sb laser pumping in 2p₀ state
- 2p₀

- Si:Sb laser pumping in 2s state
- 154 cm⁻¹
- 175.4 cm⁻¹
- 1s(T₂)

- Si:Sb laser pumping in 2p₀ state
- 2p₀

- Si:Sb laser pumping in 3p₀ state
- 3p₀
- 152.8 cm⁻¹
Si:Sb Raman laser

- $h\nu_{\text{emis}} = h\nu_{\text{pump}} - (12.10 \pm 0.02) \text{ meV}$
- 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.
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 μs 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

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

- 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: $\Gamma_7^-(1p_{5/2})$, $\Gamma_6^-(1p_{1/2})$
- Lower laser level: $2\Gamma_8^+(1p_{5/2})$

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!

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± state has a significant population and the laser transitions are 2p→1s(E), 1s(T2).
Laser dynamics

Below laser threshold (Si emission x1000)

Above laser threshold
• Gain of Si:P laser: up to 0.5 cm\(^{-1}\)
• Gain of Si:Bi laser: 0.005 – 0.02 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:As

1s(E)

1s(T₂:Γ₈)

1s(A₁)

2p⁺

2p₀

2s

f-LO

f-LA

Donor state energy [meV]

3p₀⁻(A₁+B₂)

2p⁻(2E)

2s

1s(E)

1s(T₂)

2p₀⁻(A₁+B₂)

1s(B₂)

f-LA

g-TA

Si:As emission [a. u.]

#S, F ∥ [100]

#B, F ∥ [130]

Stress [10⁸ Pa]
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

![Graph showing Si:Bi laser output versus magnetic field](image)

- Magnetic field \([ T ]\)
- Wavenumber \([\text{cm}^{-1}]\)

**Si:P laser**

**Si:Bi laser**:
- \(B \parallel [110]\)
- \(B \parallel [111]\)
- \(B \parallel [112]\)

**Magnetic field** \([T]\)

**Wavenumber** \([\text{cm}^{-1}]\)

**APL 89, 021108 (2006)**
Frequency tunability and crystal orientation

$\frac{\partial f}{\partial B} \sim 40-60 \text{ GHz/T}$
(expected from linear Zeeman effect)
Si:P lasers based with different crystal quality

- $^{28}\text{Si}$ lasers operate at the highest heat sink temperature (best heat conductivity)
- Polycrystalline Si works also.
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 Ω 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 cm⁻¹.