Hyperdoped black silicon for high-efficiency photovoltaics

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Ateneo Physics Seminar
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and also....

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Dr. Richard Meyers (RMD)

Dr. Pat Maloney (NVSED)

Dr. Jeffrey Warrander (ARDEC)
irradiate with 100-fs 10 kJ/m² pulses
“black silicon”
absorptance \( (1 - R_{int} - T_{int}) \)
absorptance \((1 - R_{\text{int}} - T_{\text{int}})\)
absorptance \( (1 - R_{int} - T_{int}) \)
absorptance \( (1 - R_{\text{int}} - T_{\text{int}}) \)
absorptance \((1 - R_{\text{int}} - T_{\text{int}})\)
laser treatment causes:

- surface structuring
- inclusion of dopants
substrate/dopant combinations

dopants:

substrates:

Si
substrate/dopant combinations

dopants:

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substrates:

Si
substrate/dopant combinations

dopants:

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substrates:

Si  Ge  ZnO  InP  GaAs
Ti  Ag  Al  Cu  Pd  Rh  Ta  Pt  TiO$_2$
gap determines optical and electronic properties

- conductor
- semiconductor
- insulator

$E_G (eV)$

Cu, Ge, Si, ZnO
shallow-level dopants control electronic properties

conductor          semiconductor          insulator

CB  VB             CB                      CB
      VB              VB

0  2  4  \( E_g \text{(eV)} \)

Cu  Ge  Si  ZnO
shallow-level dopants control electronic properties

Cu Ge Si ZnO

conductor semiconductor insulator

$E_G$ (eV)

CB
VB

0 2 4

$E_G$ (eV)

Cu Ge Si ZnO
deep-level dopants typically avoided

\[ E_g (eV) \]

- **Conductor**
  - CB
  - VB

- **Semiconductor**
  - CB
  - VB

- **Insulator**
  - CB
  - VB

Cu, Ge, Si, ZnO
femtosecond laser-doping gives rise to intermediate band
intermediate band formation in chalcogen-hyperdoped Si

dopants:

substrates:

Si
properties

2 intermediate band
10 µm

properties
cross-sectional Transmission Electron Microscopy
disordered surface layer
electron diffraction

1 µm

properties
- 300-nm disordered surface layer
- undisturbed crystalline core
- surface layer: polycrystalline Si with 1.6% sulfur
two processes: melting and ablation
relevant time scales

![Graph showing relevant time scales from 0.1 fs to 10 ns and 0.1 μs to 10 ns]
relevant time scales

carrier excitation
relevant time scales

- Carrier excitation
- Thermalization

- Cold
- Hot

- Time scales: fs, ps, ns, µs
relevant time scales

- Carrier excitation
- Cold
- Hot
- Thermalization
- Ablation

Time scales:
- fs (femtoseconds)
- ps (picoseconds)
- ns (nanoseconds)
- µs (microseconds)
relevant time scales

- Carrier excitation
- Thermalization (cold, hot)
- Ablation
- Thermal diffusion
relevant time scales

- Carrier excitation
- Thermalization
- Ablation
- Thermal diffusion
- Resolidification

Time scales:

- fs (femtoseconds)
- ps (picoseconds)
- ns (nanoseconds)
- µs (microseconds)
relevant time scales

relevant time scales

- carrier excitation
- thermalization
- ablation
- thermal diffusion
- resolidification

Room temperature lattice | Molten surface layer | Cooling material

- cold
- hot

0.1 fs | 1 ps | 10 ns | 1 μs
relevant time scales

different thresholds:

melting: 1.5 kJ/m²

ablation: 3.1 kJ/m²
decouple ablation from melting
decouple ablation from melting

doped
decouple ablation from melting

doped
decouple ablation from melting

doped

1 properties
decouple ablation from melting

undoped

doped

1 properties
decouple ablation from melting
decouple ablation from melting

epoxy

laser affected region

substrate

100 nm
decouple ablation from melting
decouple ablation from melting
decouple ablation from melting

doped region

undoped region

10 nm
secondary ion mass spectrometry

![Graph showing concentration vs. depth (nm)]
Things to keep in mind

• near unit absorption extending into IR
• surface structure due to ablation
• hyperdoping due to rapid melting and resolidification
• can decouple both processes
femtosecond laser-doping gives rise to intermediate band

\[
\begin{array}{c}
\text{conductor} \\
\begin{array}{c}
\text{CB} \\
\text{VB}
\end{array}
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\begin{array}{c}
\text{semiconductor} \\
\begin{array}{c}
\text{CB} \\
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\text{VB}
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\begin{array}{c}
\text{insulator} \\
\begin{array}{c}
\text{CB}
\end{array}
\end{array}
\]

\[E_g(\text{eV})\]

0 2 4

Cu Ge Si ZnO
1 part in $10^6$ sulfur introduces donor states in gap

1 part in $10^6$ sulfur introduces donor states in gap

1 part in $10^6$ sulfur introduces donor states in gap

1 part in $10^6$ sulfur introduces donor states in gap.

1 part in $10^6$ sulfur introduces donor states in gap

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1 part in $10^6$ sulfur introduces donor states in gap

1 part in $10^6$ sulfur introduces donor states in gap

at high concentration states broaden into band

1 properties  
2 intermediate band
isolate surface layer for Hall measurements

1 properties

2 intermediate band
isolate surface layer for Hall measurements

1 properties
2 intermediate band
isolate surface layer for Hall measurements

1 properties
2 intermediate band

- laser doped region
- buried oxide
- silicon substrate
isolate surface layer for Hall measurements

1 properties
2 intermediate band

buried oxide

silicon substrate
isolate surface layer for Hall measurements
1 properties

2 intermediate band
impurity (donor) band centered at 310 meV
Insulator-to-Metal Transition in Selenium-Hyperdoped Silicon: Observation and Origin

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Hyperdoping has emerged as a promising method for designing semiconductors with unique optical and electronic properties, although such properties currently lack a clear microscopic explanation. Combining computational and experimental evidence, we probe the origin of sub-band-gap optical absorption and metallicity in Se-hyperdoped Si. We show that sub-band-gap absorption arises from direct defect-to-conduction-band transitions rather than free carrier absorption. Density functional theory predicts the Se-induced insulator-to-metal transition arises from merging of defect and conduction bands, at a critical concentration, demonstrate that correlation is important to describing the transition accurately, and suggest that it is a classic impurity-driven Mott transition.

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Of all the experimentally measurable physical properties of materials, electronic conductivity exhibits the largest variation, spanning a factor of $10^{3}$ from the best metals to the strongest insulators [1]. Over the last century, the puzzle of why some materials are conductors and others insulators, and the mechanisms underlying the transformation from one to the other, have been carefully scrutinized, yet even after such a vast body of research over such a long period, the subject remains the object of controversy. In 1956, Mott introduced a model for the insulator-to-metal transition (IMT) in doped semiconductors, in which long-ranged electron correlations are the driving force [2]; this model creates a new materials playground to explore defect-mediated IMTs in semiconductors. In this Letter, we identify a defect-induced IMT in silicon hyperdoped with selenium and explore the role of correlation therein.

Hyperdoping (doping beyond the solubility limit) creates new materials playground to explore defect-mediated IMTs in semiconductors. In this Letter, we identify a defect-induced IMT in silicon hyperdoped with selenium and explore the role of correlation therein.

Silicon appears to justify such interest. While isolated S and Se dopants are well-established deep double donors in silicon [3,14], the enhanced optical properties of hyperdoped silicon (in which these chalcogenic impurities are doped silicon at much higher concentrations) are not yet well understood. Further, unlike the prototypical system of phosphorus-doped silicon for which the IMT has been extensively studied and characterized [15,16], there are very few studies of an IMT resulting from deep defects such as chalcogen dopants [17].

We prepared Se-doped silicon (Se:Si) samples using an ion beam energy of 176 keV. The implanted samples were exposed to four laser pulses (fluence regimen 1.7, 1.7, 1.7 and 1.7 J cm$^{-2}$) in sequence. The Se concentration-depth profile was measured by secondary ion mass spectrometry [18]. The Se-rich layer is crystalline, extends approximately 350 nm from the surface, and is electrically isolated from the substrate using cloverleaf-mesa structures. Conductivity was calculated from sheet conductivity using the effective implantation technique.

PACS numbers: 71.30.+h, 61.72.sd, 73.61.Cw, 78.20.Bh
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Department of Mechanical Engineering, Massachusetts Institute of Technology, Cambridge Massachusetts 02139, USA


1 properties

2 intermediate band
Emergence of very broad infrared absorption band by hyperdoping of silicon with chalcogens

Ikurou Umezu, Jeffrey M. Warrender, Supakit Charnvanichborikarn, Atsushi Kohno, James S. Williams, Malek Tabbaa, Dimitris G. Papazoglou, Xi-Cheng Zhang, and Michael J. Aziz

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We report the near through mid-infrared (MIR) optical absorption spectra, over the range 0.05–1.3 eV, of monocrystalline silicon layers hyperdoped with chalcogen atoms synthesized by ion implantation followed by pulsed laser melting. A broad mid-infrared optical absorption band emerges, peaking near 0.5 eV for sulfur and selenium and 0.3 eV for tellurium hyperdoped samples.

II. EXPERIMENT

Double side polished p type (001) Si wafers, resistivity of 5–25 Ω cm, were ion implanted at room temperature with either 95 keV 32S⁺, 176 keV 100Se⁺, or 245 keV 130Te⁺ to 8×10^{16} ions/cm² and pre-amorphized by a third laser shot at 1.8 J/cm². The dose of 32S⁺ was varied from 3×10^{14} to 1×10^{16} ions/cm² and pre-amorphized when 32S⁺ dose is not greater than 1×10^{15} ions/cm². Pulsed laser melting was performed using a XeCl excimer laser beam (308 nm, 25 ns FWHM, 50 ns total duration). Each sample received three laser shots at 1.7 J/cm² followed by a fourth laser shot at 1.8 J/cm². Time-resolved reflectivity of a laser, 488 nm Ar⁺ ion laser was used to measure the melt duration. The laser fluence was calibrated by comparing the one-dimensional heat conduction for numerical solutions to the sample preparation method. The details of the sample preparation method is the same as reported elsewhere.

1 properties
2 intermediate band
3 32S⁺
4 100Se⁺
5 130Te⁺
Emergence of very broad infrared absorption band by hyperdoping of silicon with chalcogens

Ikurou Umezu, 1 Jeffrey M. Warrender, 2 James S. Williams, 3 Atsushi Kohno, 4 Dimitris G. Papazoglou, 5 Supakit Charnvanichborikarn, 6 Atsushi Kohno, 7 Xi-Cheng Zhang, 8, 9

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I. INTRODUCTION

Silicon-based infrared (IR) photodetectors have received much interest as they have high quantum efficiency and are compatible with silicon complementary metal–oxide–semiconductor (CMOS) technology. 

Silicon photodetectors are one of the most important materials for telecommunications, environmental monitoring, and security applications. Silicon photodetectors have been widely used in the near infrared (NIR) region, or to photon energies above the band gap. However, the IR absorption band is extremely weak even over a broad range of photon energies. 

Silicon has been shown to cause an absorption coefficient over a broad range of photon energies, whereas the previous work employed a somewhat higher fluence for Selenium and Tellurium doped samples. Following laser melting, the absorption strength decreases markedly with the concentration of chalcogen atoms observed by secondary ion mass spectrometry (SIMS). The details of the sample preparation method were reported elsewhere.

II. EXPERIMENT

Double side polished p type (001) Si wafers, resistivity 0.05–1.3 eV, of monocrystalline silicon layers hyperdoped with chalcogen atoms synthesized by ion implantation followed by pulsed laser melting. A broad mid-infrared optical absorption band emerges, peaking near 0.5 eV for sulfur and selenium and 0.3 eV for tellurium hyperdoped samples. Its strength and width increase with impurity concentration. Its strength decreases markedly with pulsed laser mixing.

We report the emergence of a broad MIR absorption band is consistent with the formation of an impurity band from isolated deep donor levels as the concentration of chalcogen atoms in metastable local configurations increases. The hyperdoping-induced changes causing the sub-bandgap absorption have remained unclear. One reason for this is that most measurements of optical absorption have been limited to the near IR (NIR) region, or to photon energies above the band gap. However, for sufficiently high concentrations of titanium ion laser was used to measure the melt duration.

The laser fluence was calibrated by comparing the melt duration for some of the samples were thermally annealed in a flowing argon gas atmosphere for 30 min. We measured the transmittance and reflectance of samples in the NIR region using a grating spectrophotometer. The details of the sample preparation method were reported elsewhere.


properties

intermediate band
Things to keep in mind

- IR absorption rolls off around 8 µm
- Consistent evidence of intermediate band formation
- IB forms at 0.1% at. doping, broadens at higher doping
- IB merges with CB at 0.4% at. yielding metallic behavior
properties
intermediate band
devices
solar spectrum
solar spectrum

![Graph of solar spectrum](image)

- Spectral irradiance (kW/m²/µm)
- Wavelength (µm)

- 1353 W/m²

1. properties
2. intermediate band
3. devices
crystalline silicon: transparent to 23% of solar radiation

![Graph showing spectral irradiance vs wavelength for crystalline silicon. The graph includes a shaded region indicating the c-Si band gap at 1.12 μm.](image-url)
amorphous silicon: transparent to 53% of solar radiation

Properties

Intermediate Band

Devices
1 properties  2 intermediate band  3 devices
photon with gap energy

1 properties
2 intermediate band
3 devices
photon creates electron-hole pair...

1 properties
2 intermediate band
3 devices
...whose energy can be extracted

1. properties
2. intermediate band
3. devices
photons with energy smaller than gap...
...do not get absorbed

1 properties  
2 intermediate band  
3 devices
photons with energy larger than the gap...
...create electron-hole pairs with excess energy...

1 properties
2 intermediate band
3 devices
...which is lost rapidly

1 properties
2 intermediate band
3 devices
black silicon has an intermediate band

CB

VB

1 properties
2 intermediate band
3 devices
absorbs same photons as ordinary silicon…

1 properties
2 intermediate band
3 devices
...but extends absorption to longer wavelengths
could theoretically get efficiencies over 50%
should have shallow junction below surface

sulfur-doped layer

p-doped substrate

1 properties            2 intermediate band              3 devices
excellent rectification (after annealing)

![Graph showing current density vs. bias voltage]

1 properties  
2 intermediate band  
3 devices
responsivity

![Graph showing responsivity vs wavelength](image)

- **1** properties
- **2** intermediate band
- **3** devices
responsivity

![Graph showing responsivity vs. wavelength for different materials: Si PIN and InGaAs, Ge. The x-axis represents wavelength in nm, and the y-axis represents responsivity in A/W.](image)

1. properties
2. intermediate band
3. devices
responsivity

![Graph showing responsivity vs wavelength](image)

1. properties
2. intermediate band
3. devices
responsivity

![Graph showing responsivity vs wavelength for different materials: Si:Si, Si PIN, InGaAs, Ge. The graph illustrates the sensitivity of these materials in different wavelength bands.](image)

1. properties
2. intermediate band
3. devices
Responsivity

![Graph showing responsivity versus wavelength for different materials]

1. Properties
2. Intermediate band
3. Devices
Things to keep in mind

- can turn absorption into carrier generation
- very high responsivity in VIS and IR
- intermediate band photovoltaic devices?
Potential benefits for photovoltaics

- surface structure
- absorption in submicrometer layer
- extended IR absorption
- intermediate band
water splitting

water splitting

water splitting

1. properties
2. intermediate band
3. devices
water splitting

(anode)
semiconductor

(cathode)
metal

1 properties
2 intermediate band
3 devices
water splitting

(anode)
semiconductor
metal
CB
VB
e
h

(1) properties
(2) intermediate band
(3) devices
water splitting

1. properties
2. intermediate band
3. devices
water splitting

(anode) semiconductor

(cathode) metal

1 properties
2 intermediate band
3 devices
water splitting

(anode)

semiconductor

(cathode)

metal

CB

VB

e

O2

H+

1 properties

2 intermediate band

3 devices
water splitting

(anode)
semiconductor
(cathode)
metal

CB
VB
O2
H+
e

1 properties
2 intermediate band
3 devices
water splitting

(anode) semiconductor
(cathode) metal

CB
VB
O2
H+
e

1 properties
2 intermediate band
3 devices
water splitting

(anode)
semiconductor
(cathode)
metal

CB
VB
O₂
H⁺
e

1 properties
2 intermediate band
3 devices
water splitting

1. properties
2. intermediate band
3. devices
water splitting

(anode)
semiconductor
(cathode)
metal
CB
VB
O₂
H₂

1 properties
2 intermediate band
3 devices
solar radiation spectrum

- Properties
- Intermediate band
- Devices
solar radiation spectrum

![Diagram of solar radiation spectrum with UV, visible, and infrared regions highlighted.](image)

**Legend:**
- **UV**
- **visible**
- **infrared**

**Graph:**
- **Wavelength (nm):** 0 to 2500
- **Spectral Irradiance (W/m²/nm):** 0 to 2.5
- **TiO₂ Absorptance:** 0 to 1

**Key:**
- **Properties:**
- **Intermediate Band:**
- **Devices:**
solar radiation spectrum

UV visible infrared

0 500 1000 1500 2000 2500

2.0
2.5
1.5
1.0
0.5
0

wavelength (nm)

spectral irradiance (W/m²/nm)

TiO₂ absorptance

1 properties
2 intermediate band
3 devices
1 properties  2 intermediate band  3 devices
Summary

- new doping process
- new class of material
- new types of devices

1 properties  2 intermediate band  3 devices