

Hyperdoped black silicon for high-efficiency photovoltaics

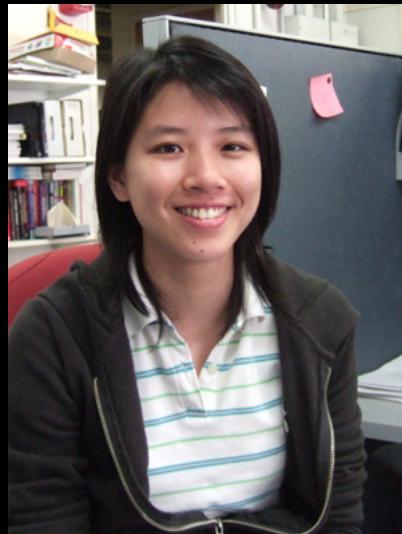


Benjamin Franta
Ateneo Physics Seminar
July 8, 2014





Renee Sher



Yu-Ting Lin



Kasey Philips



Ben Franta



eric_mazur

and also....

Hemi Gandhi
Alexander Raymond
Marc Winkler
Eric Diebold
Haifei Albert Zhang
Dr. Brian Tull
Dr. Jim Carey (SiOnyx)
Prof. Tsing-Hua Her (UNC Charlotte)
Dr. Shrenik Deliwala
Dr. Richard Finlay
Dr. Michael Sheehy
Dr. Claudia Wu
Dr. Rebecca Younkin
Prof. Catherine Crouch (Swarthmore)
Prof. Mengyan Shen (Lowell U)
Prof. Li Zhao (Fudan U)

Prof. Alan Aspuru-Guzik
Prof. Michael Aziz
Prof. Michael Brenner
Prof. Cynthia Friend
Prof. Howard Stone

Dr. Martin Pralle (SiOnyx)
and everyone else at SiOnyx...

Prof. Tonio Buonassisi (MIT)
Prof. Silvija Gradecak (MIT)
Prof. Jeff Grossman (MIT)
Dr. Bonna Newman (MIT)
Joe Sullivan (MIT)
Matthew Smith (MIT)

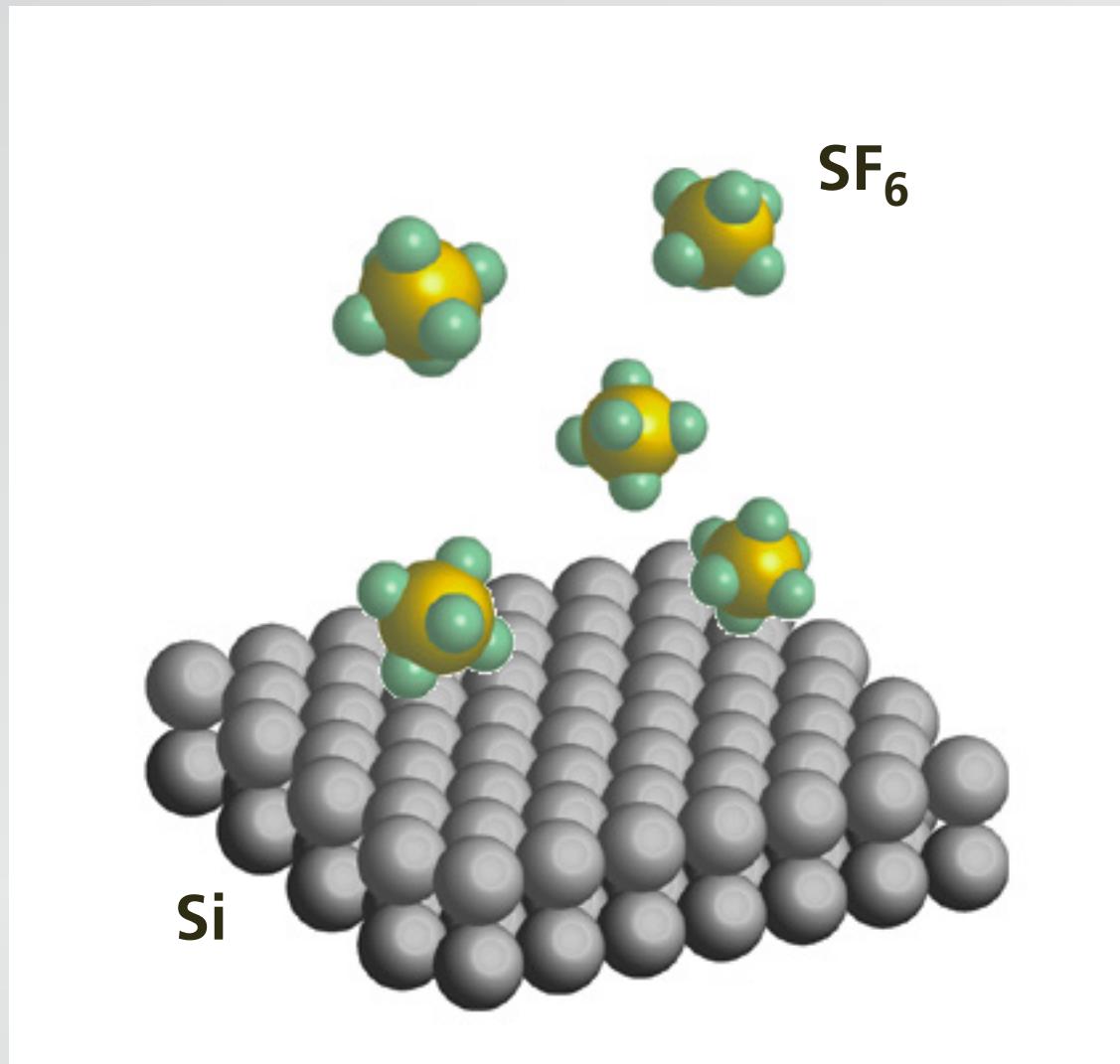
Prof. Augustinus Asenbaum (Vienna)

Dr. François Génin (LLNL)
Mark Wall (LLNL)

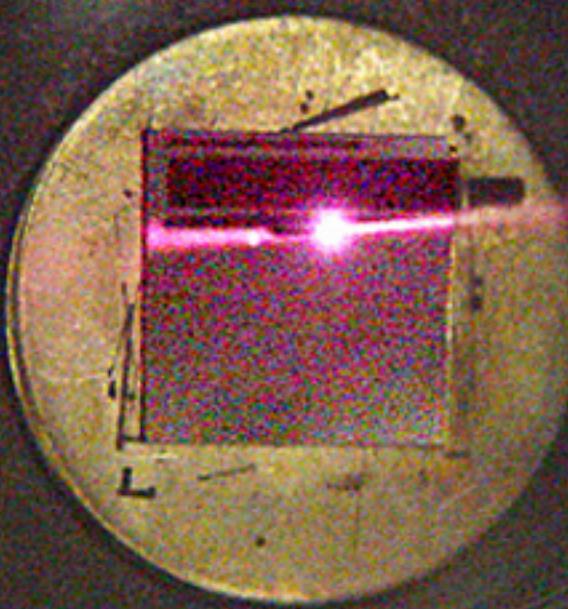
Dr. Richard Farrell (RMD)
Dr. Arieh Karger (RMD)
Dr. Richard Meyers (RMD)

Dr. Pat Maloney (NVSED)

Dr. Jeffrey Warrander (ARDEC)

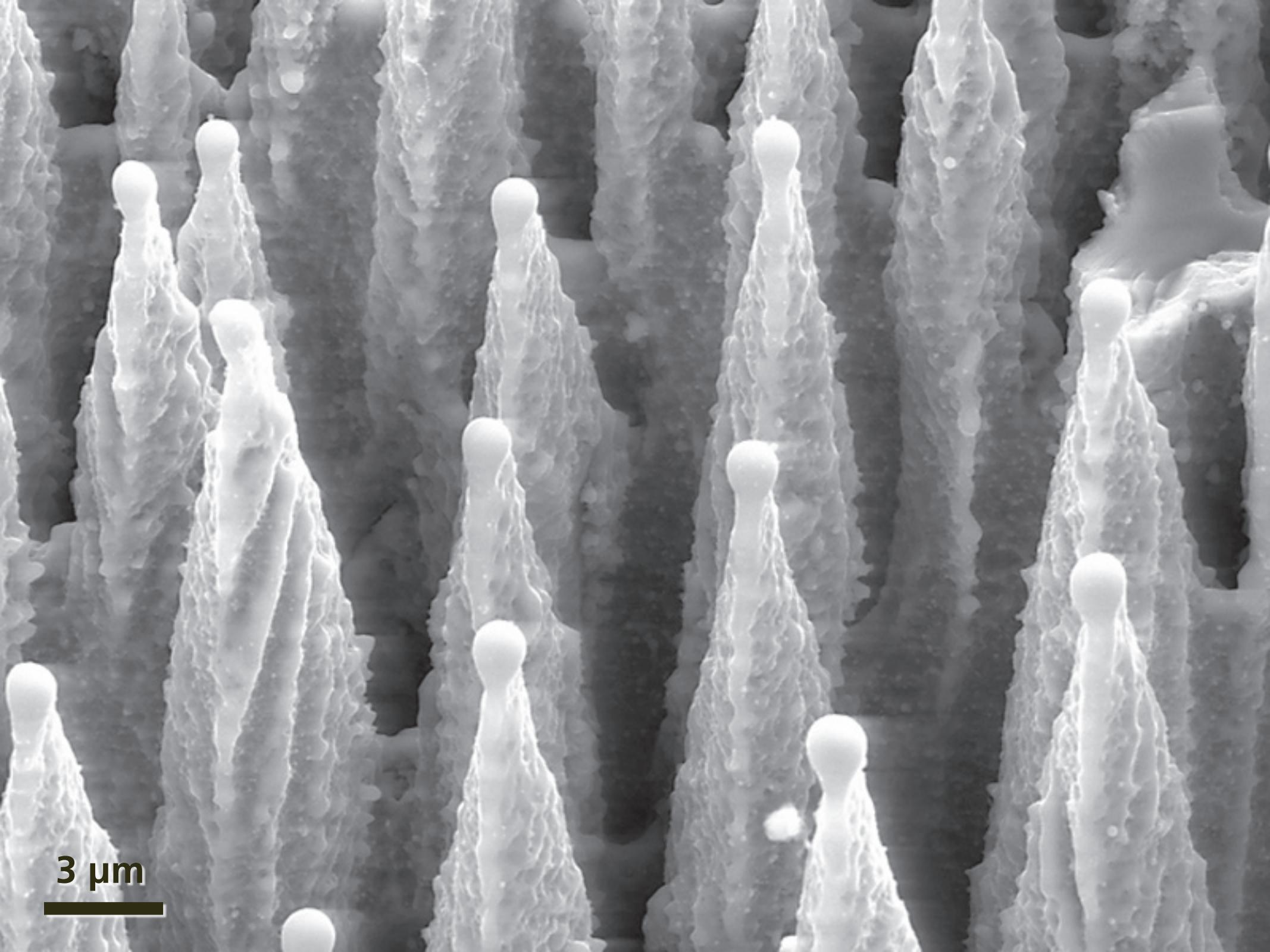


irradiate with 100-fs 10 kJ/m² pulses



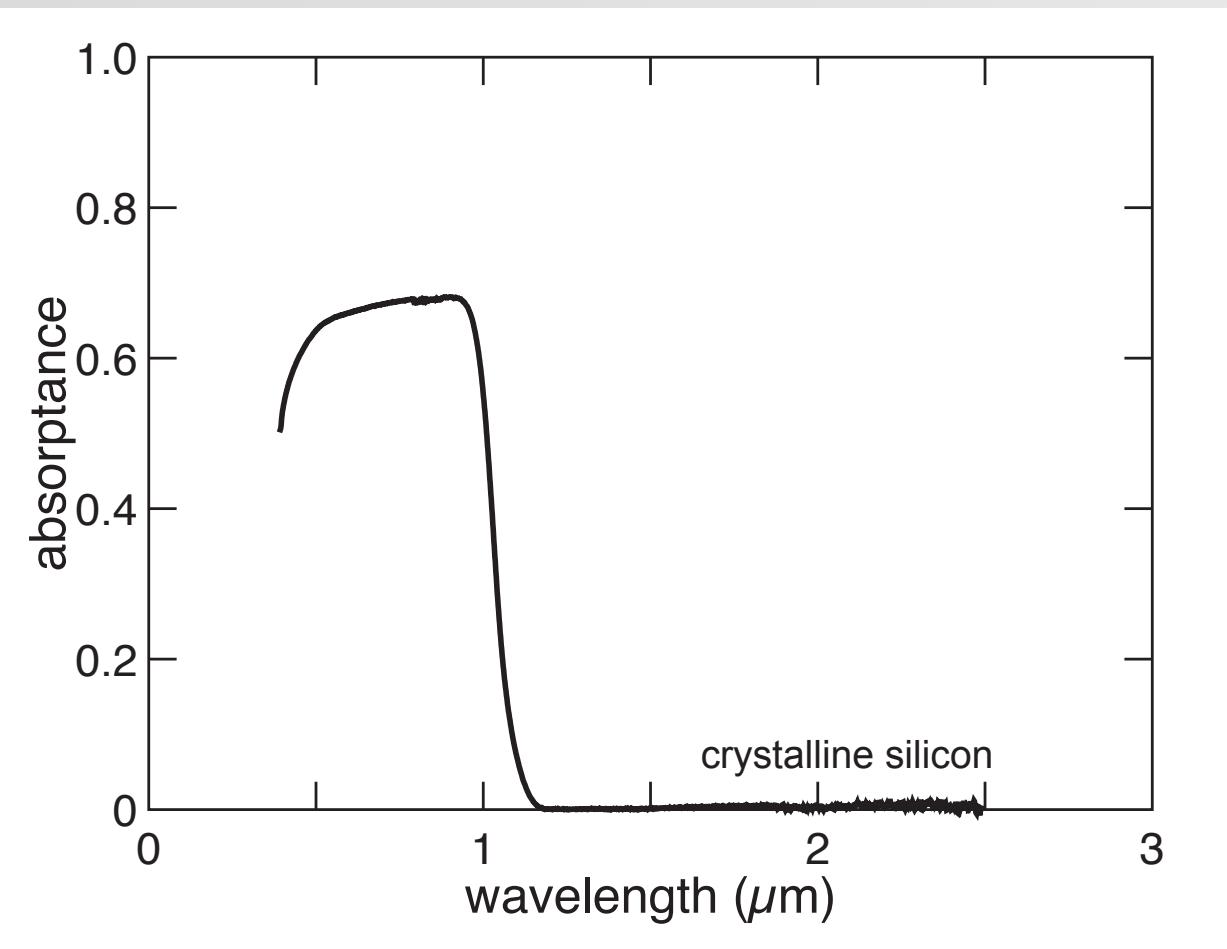


"black silicon"

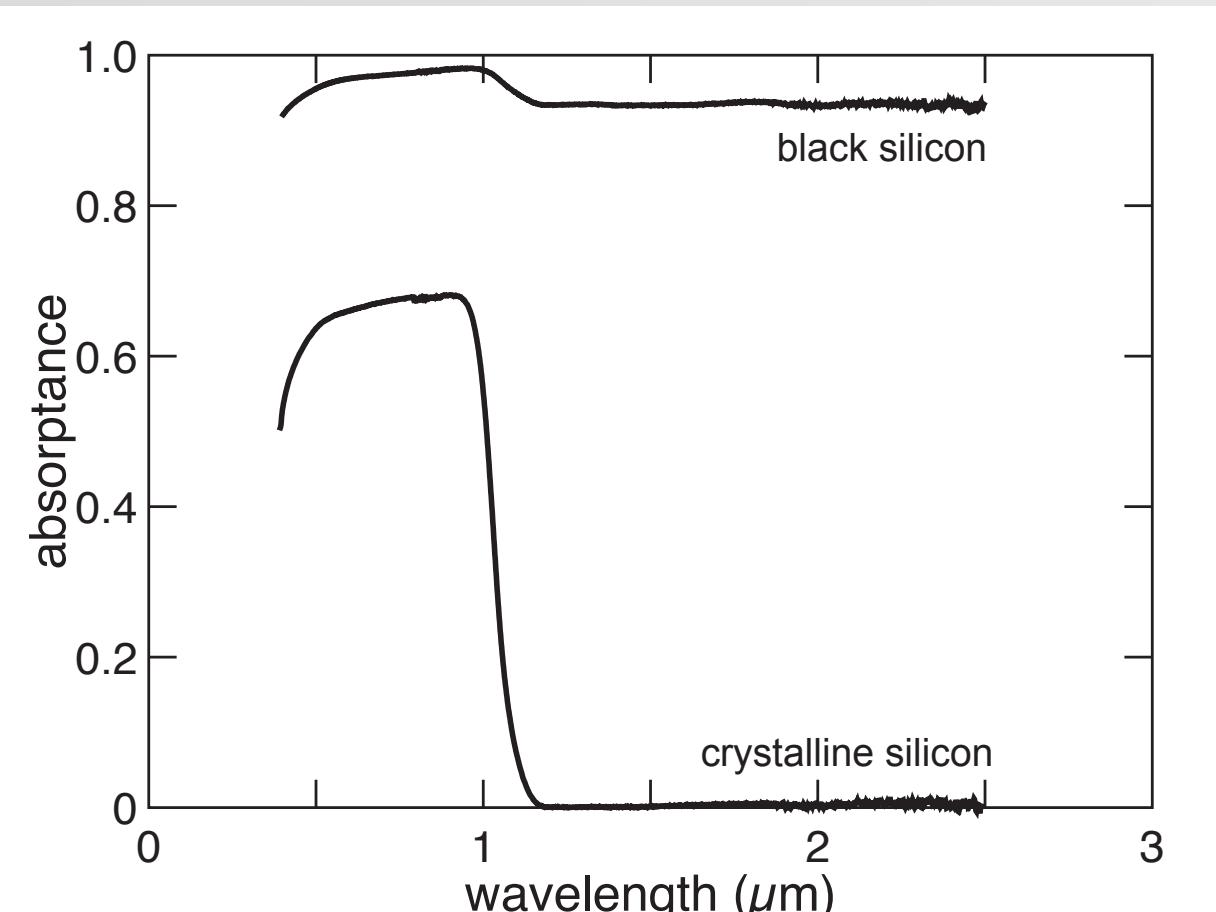


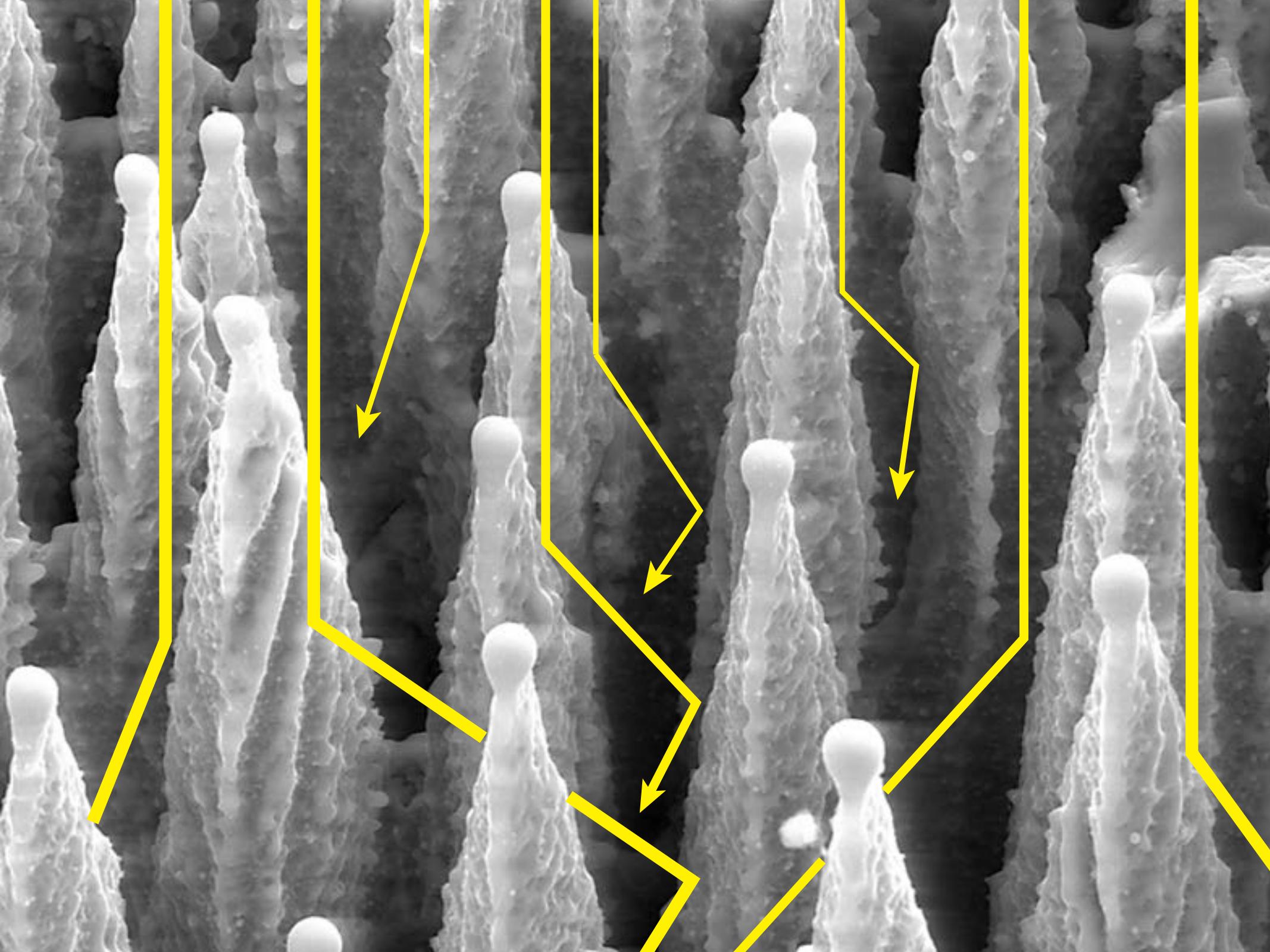
3 μm

absorptance ($1 - R_{int} - T_{int}$)

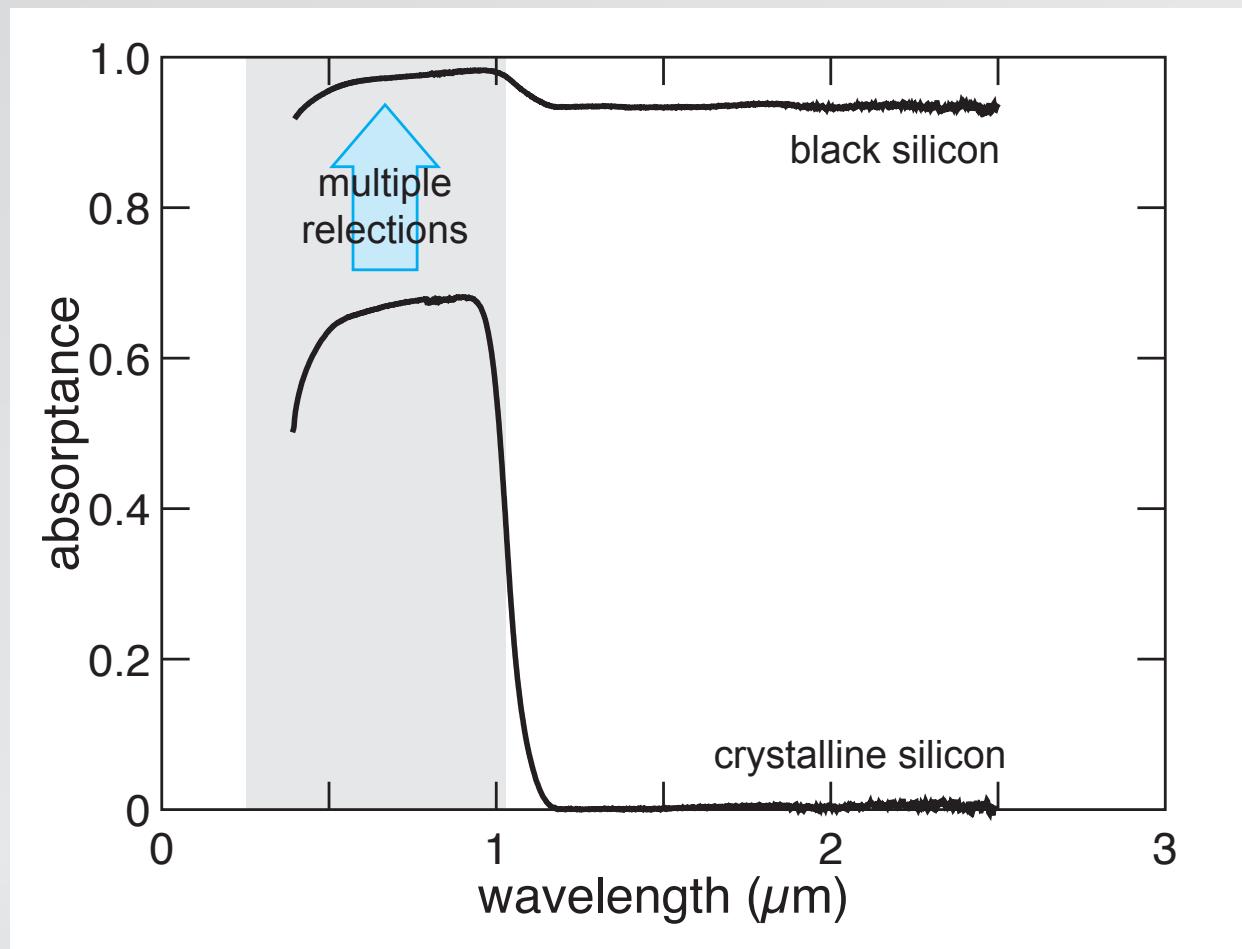


absorptance ($1 - R_{int} - T_{int}$)

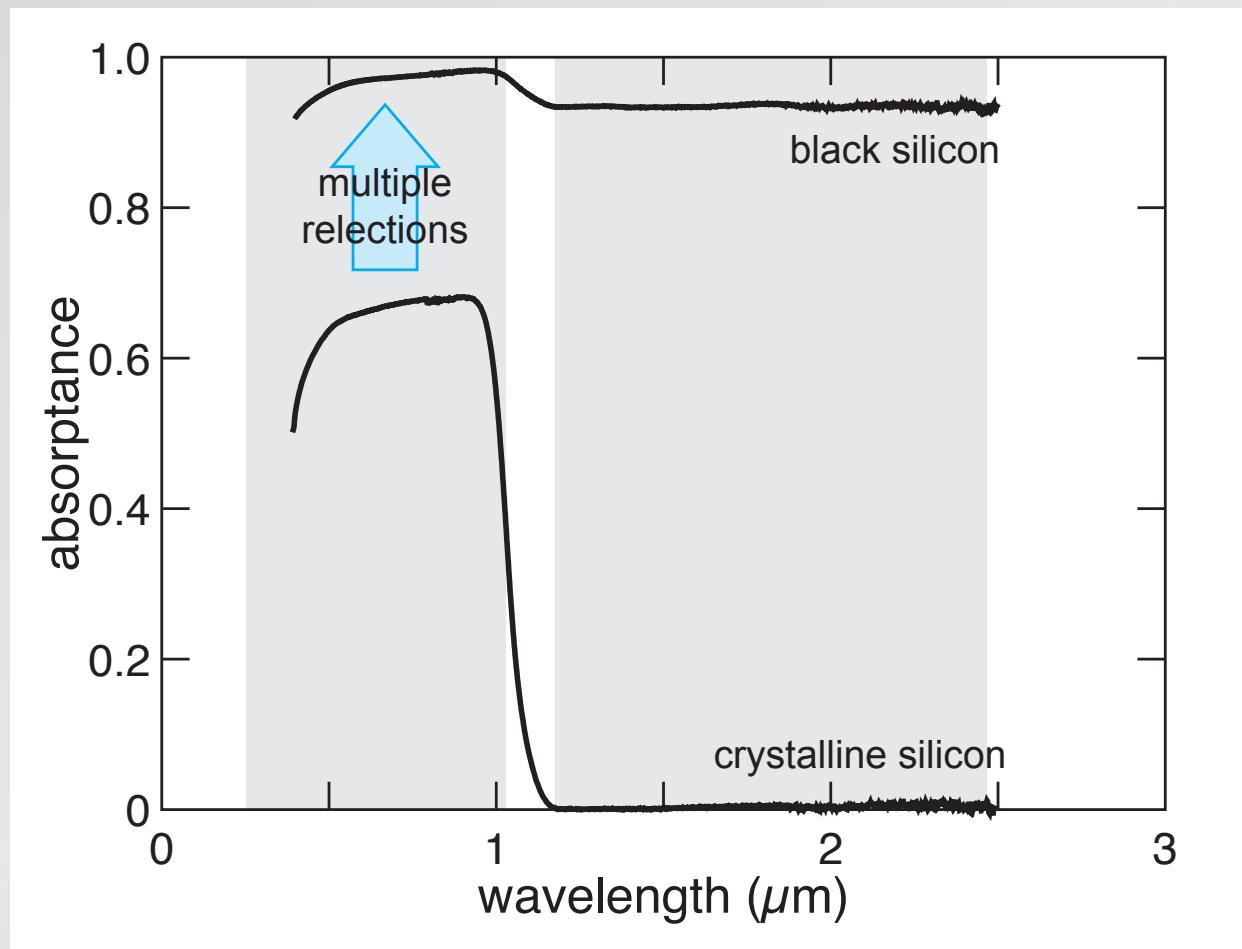




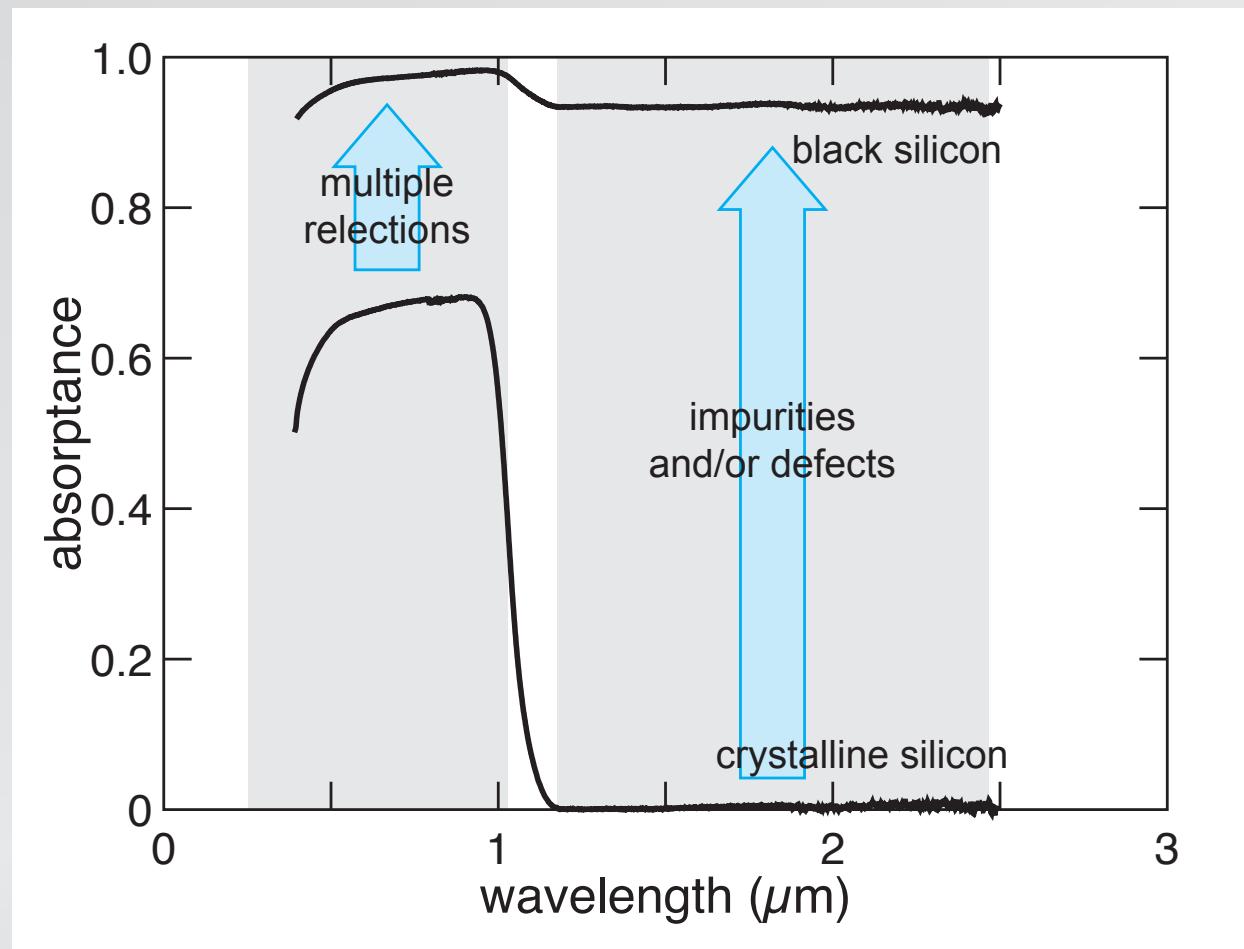
absorptance ($1 - R_{int} - T_{int}$)



absorptance ($1 - R_{int} - T_{int}$)



absorptance ($1 - R_{int} - T_{int}$)



laser treatment causes:

- **surface structuring**
- **inclusion of dopants**

substrate/dopant combinations

dopants:

I															VIII
H	II														
Li	Be														
Na	Mg														
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te
															I
															Xe

substrates:

Si

substrate/dopant combinations

dopants:

I	II	III	IV	V	VI	VII	VIII										
H	Be	B	C	N	O	F	He										
Li	Mg	Al	Si	P	S	Cl	Ne										
Na		Ga	Ge	As	Se	Br	Ar										
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe

substrates:

Si

substrate/dopant combinations

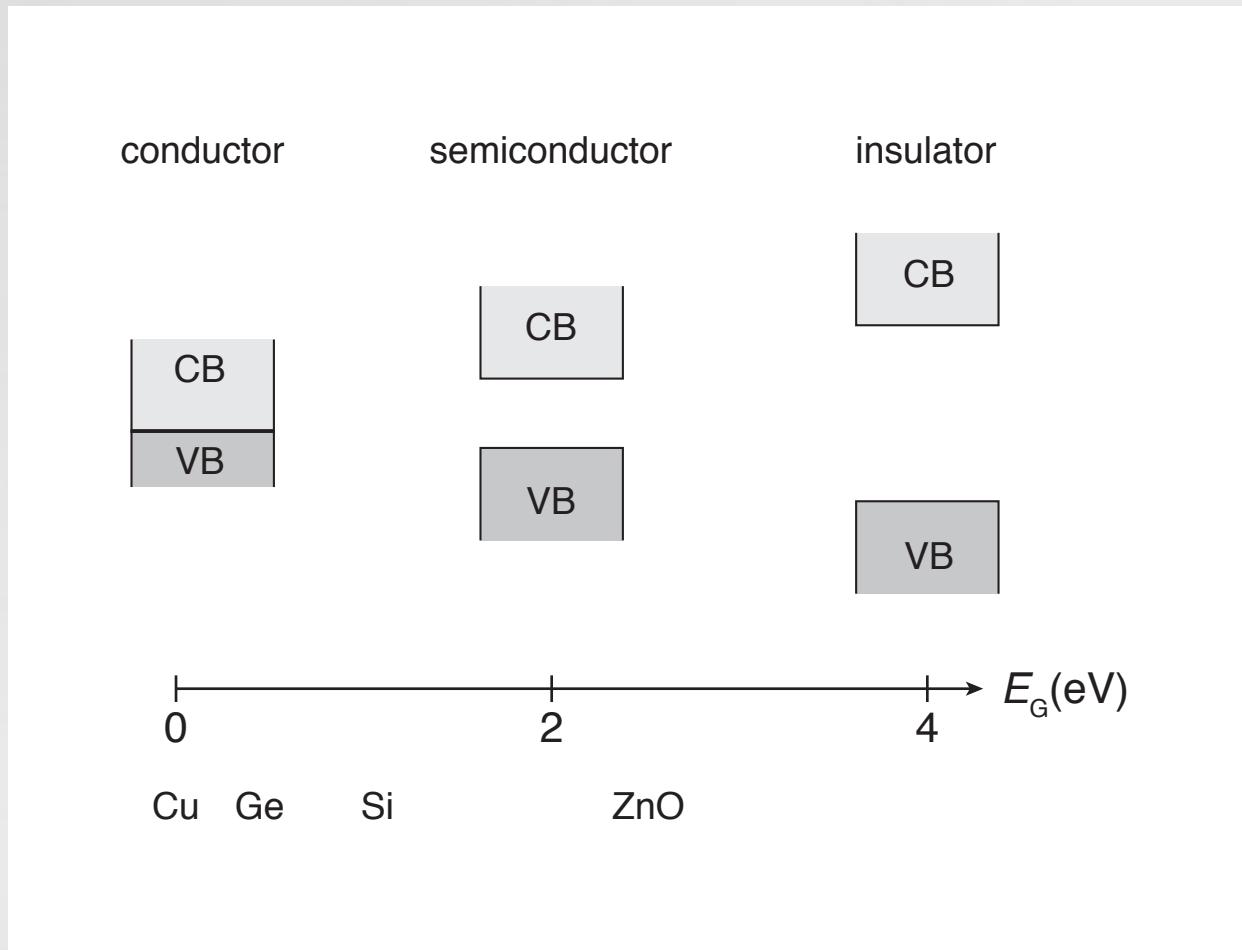
dopants:

I	II	III	IV	V	VI	VII	VIII										
H	Be	B	C	N	O	F	He										
Li	Mg	Al	Si	P	S	Cl	Ne										
Na		Ga	Ge	As	Se	Br	Ar										
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe

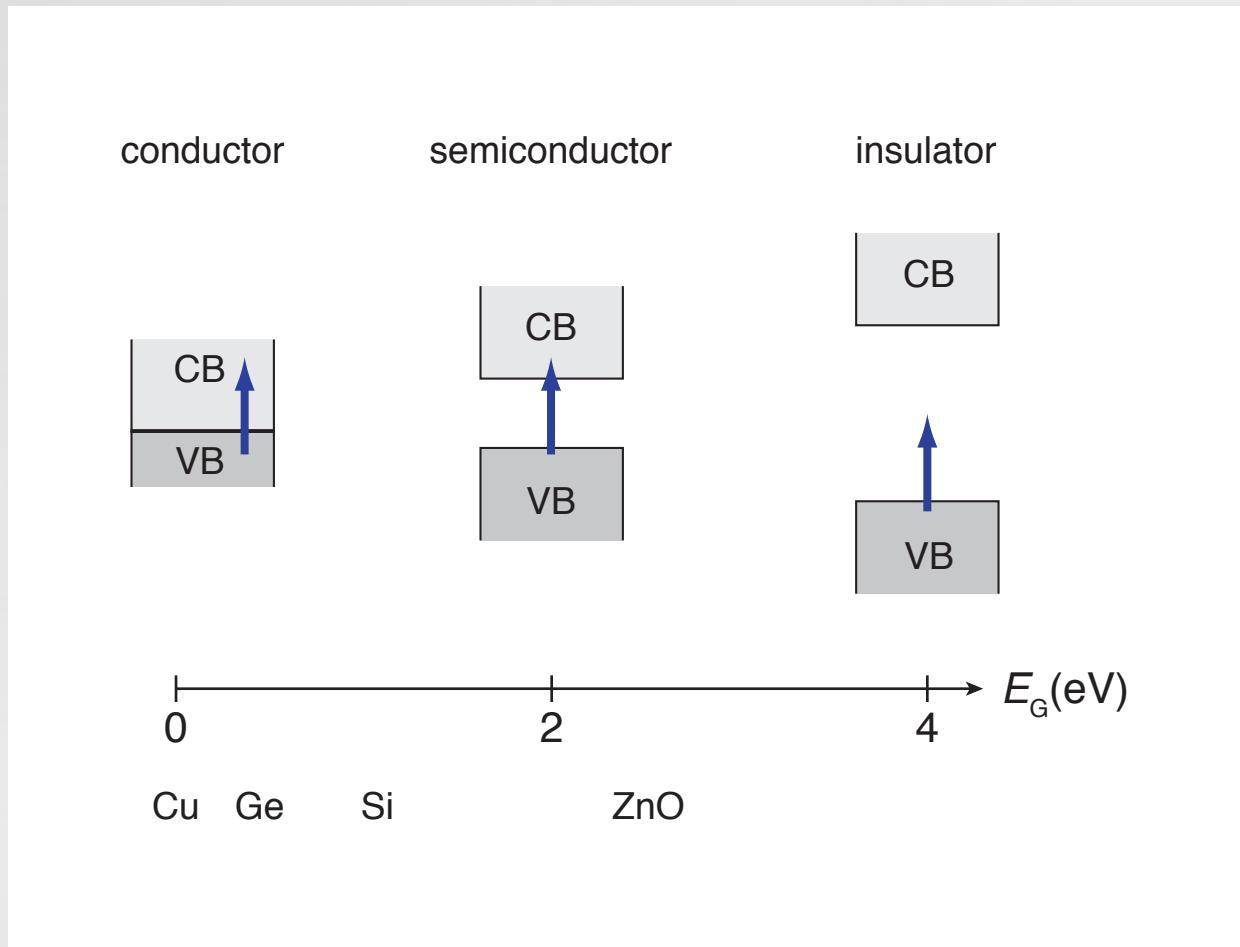
substrates:

Si Ge ZnO InP GaAs

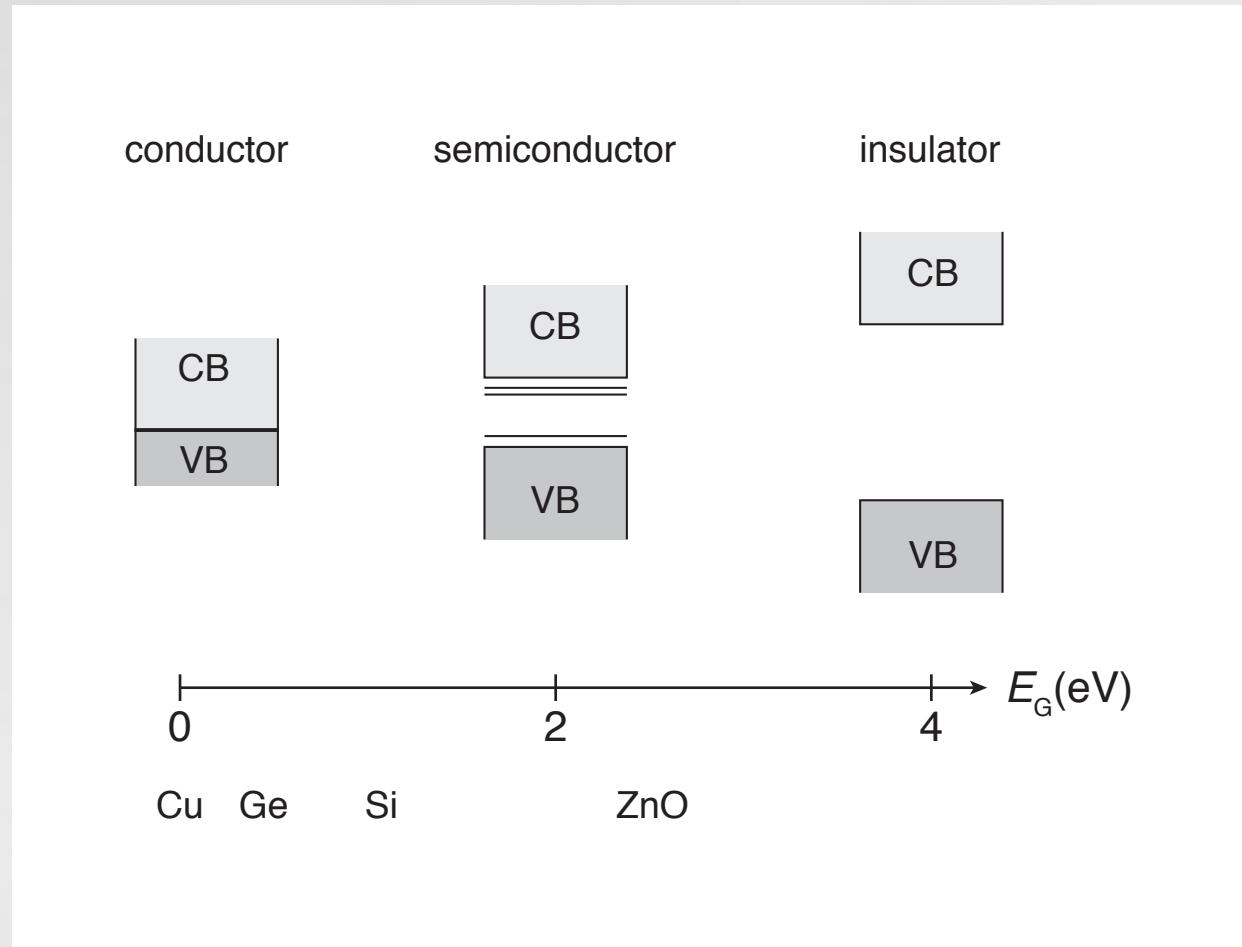
Ti Ag Al Cu Pd Rh Ta Pt TiO₂



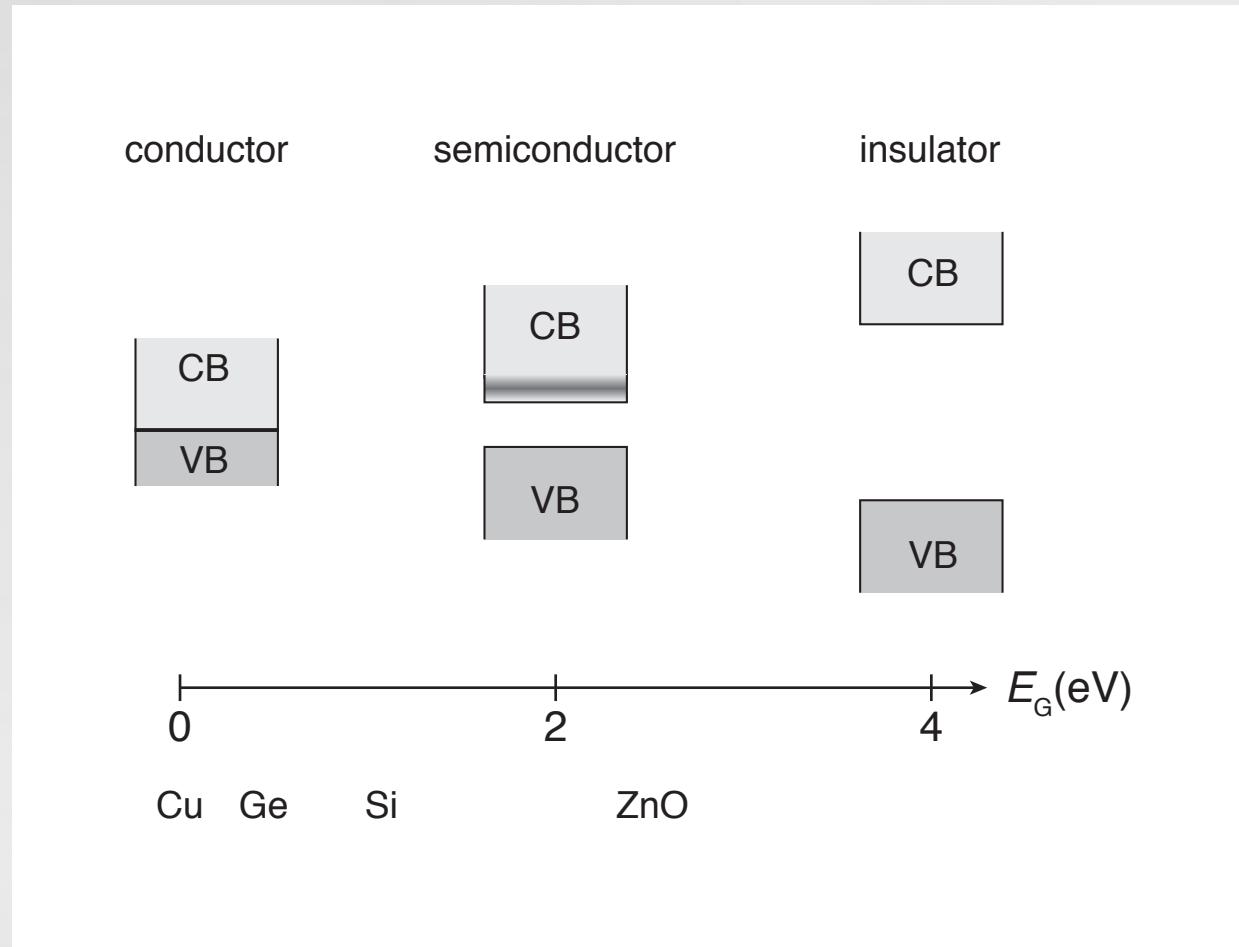
gap determines optical and electronic properties



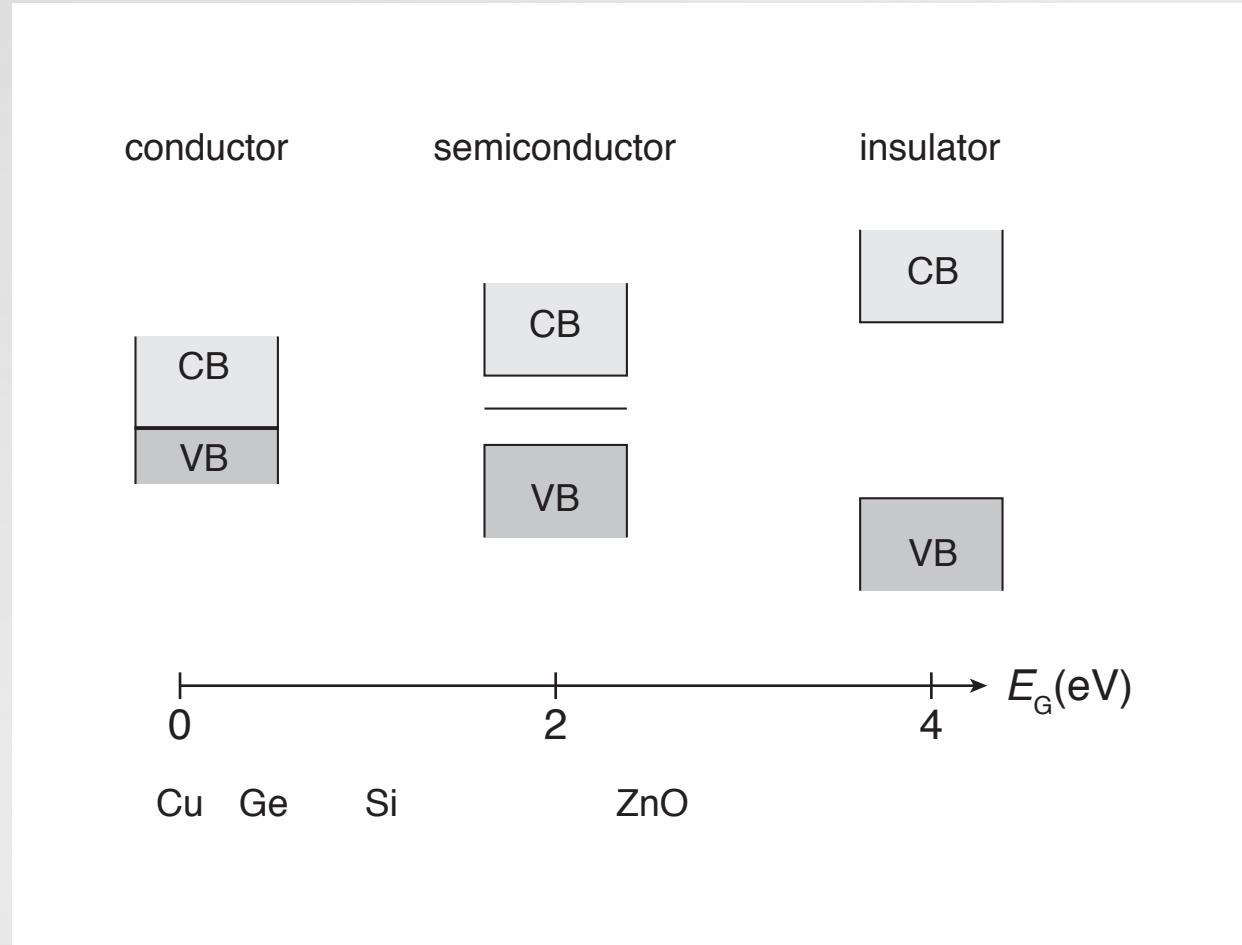
shallow-level dopants control electronic properties



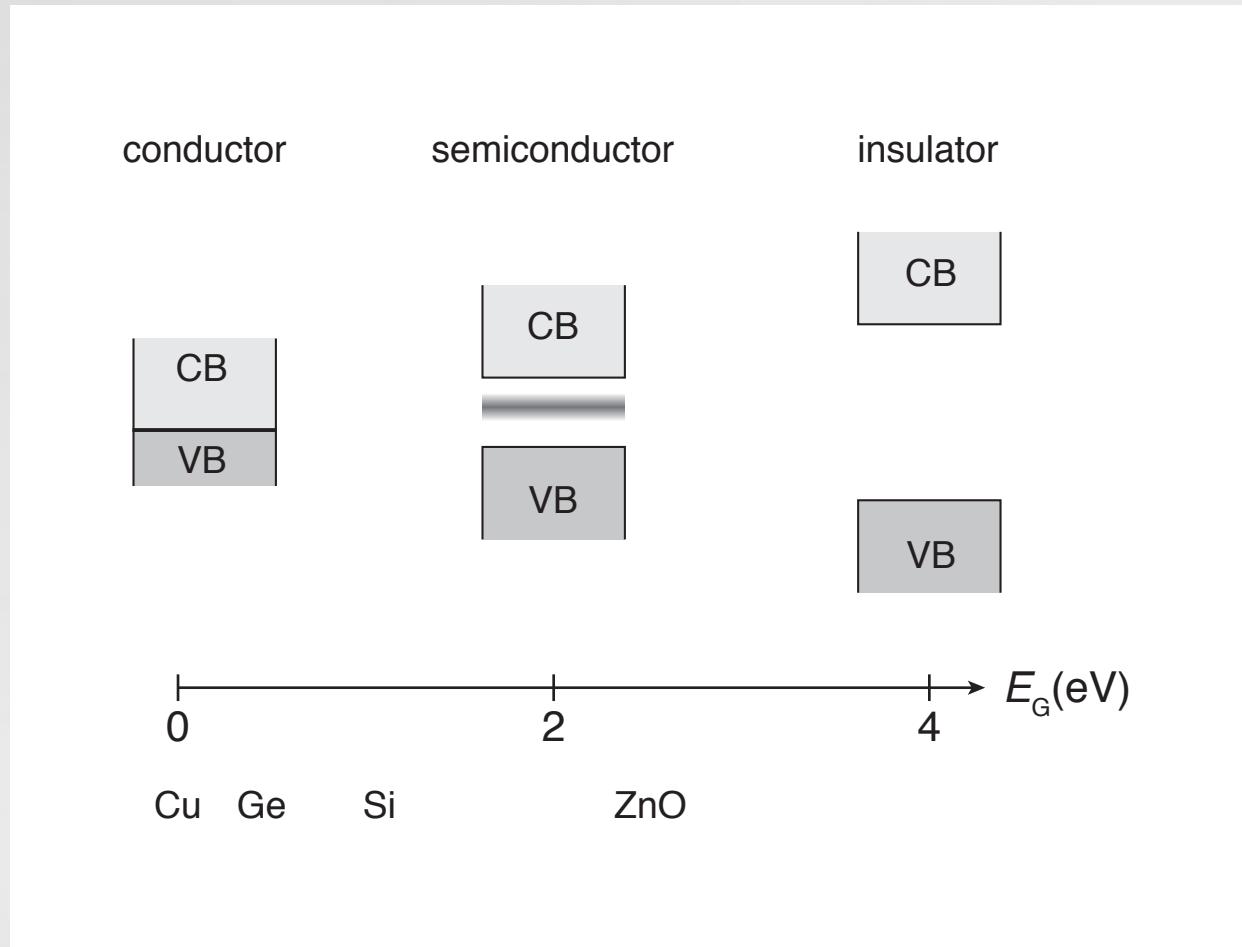
shallow-level dopants control electronic properties



deep-level dopants typically avoided



femtosecond laser-doping gives rise to intermediate band



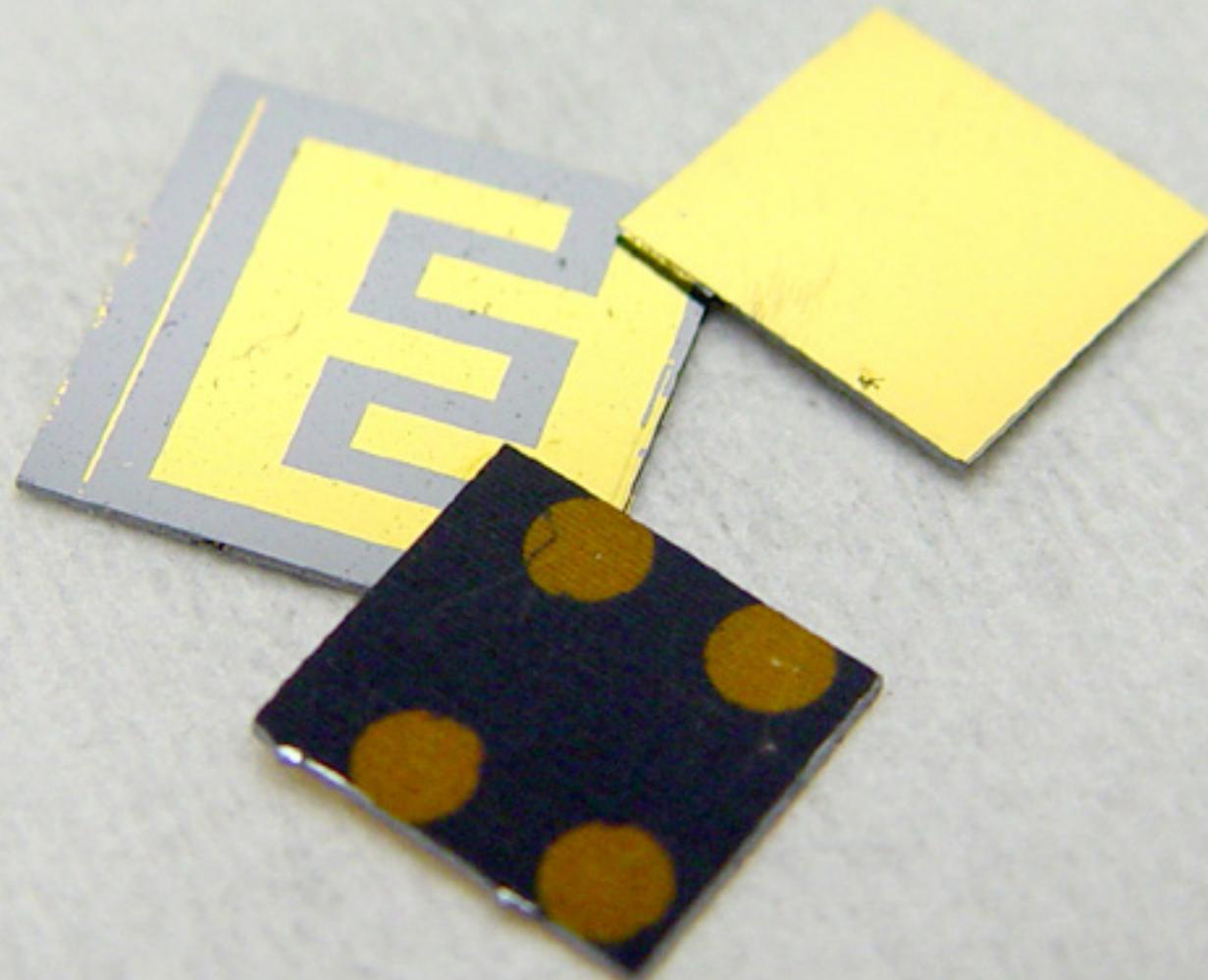
intermediate band formation in chalcogen-hyperdoped Si

dopants:

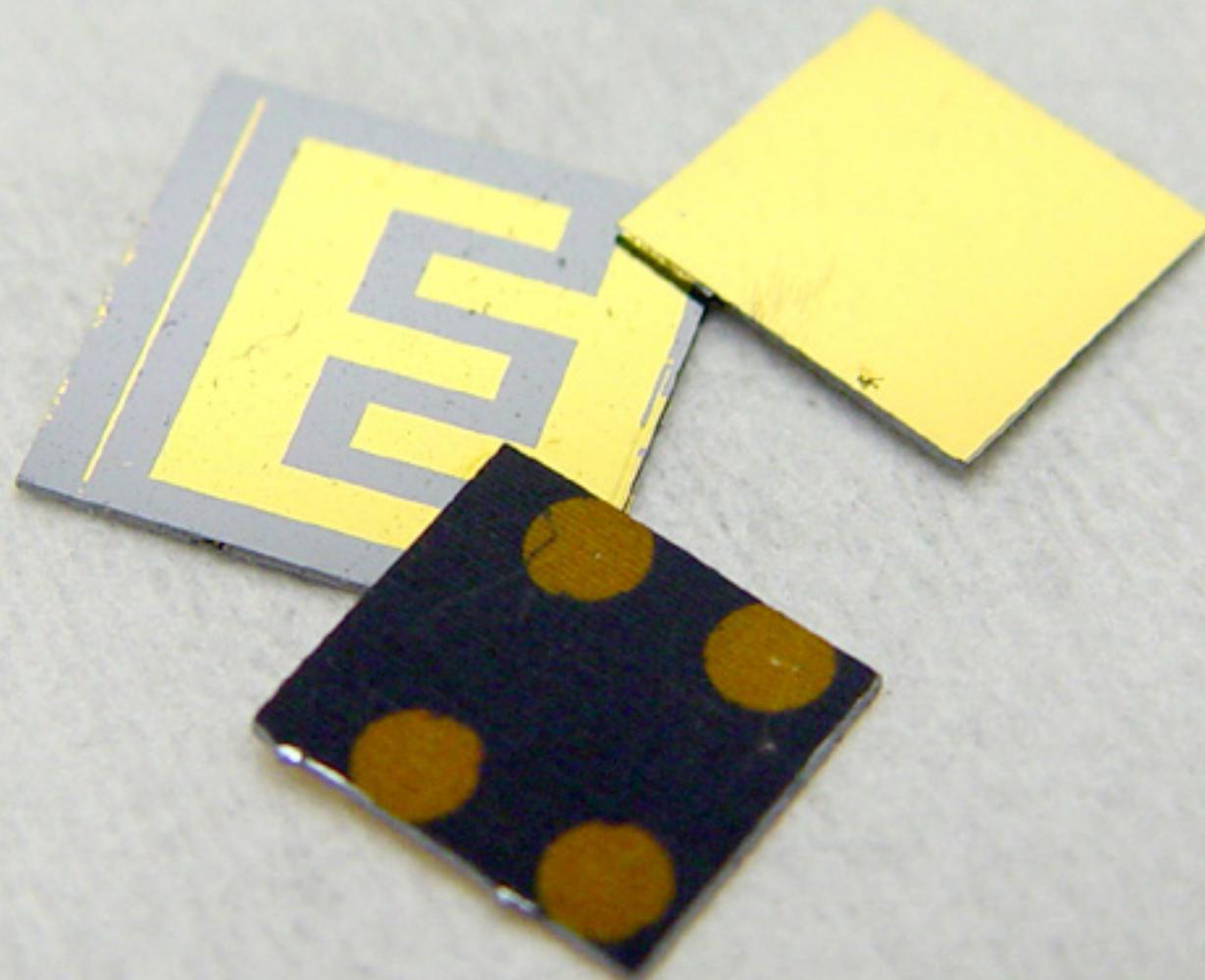
I															VIII
H															He
Li	Be														
Na	Mg														
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te
															I
															Xe

substrates:

Si

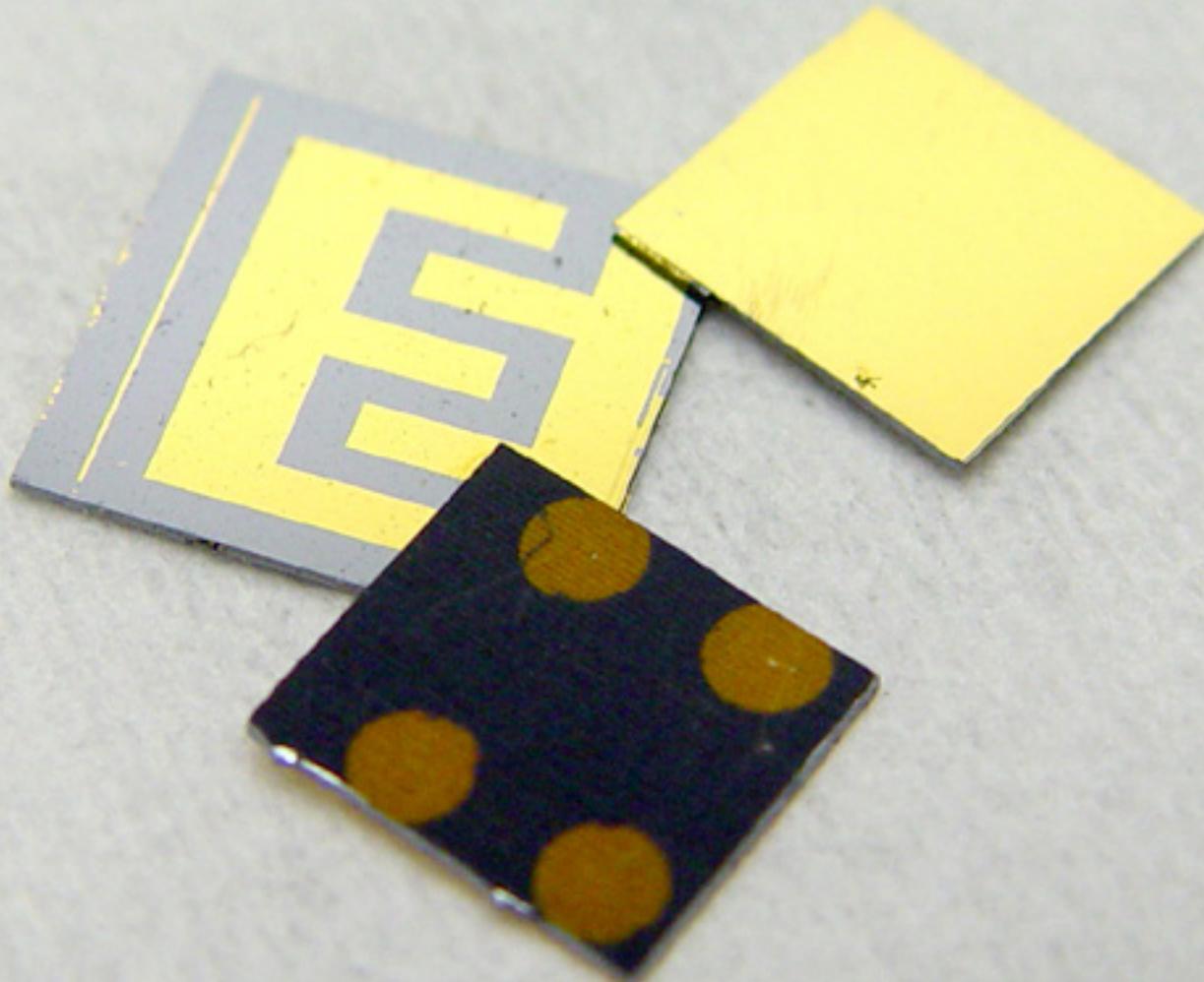


1 properties



1 properties

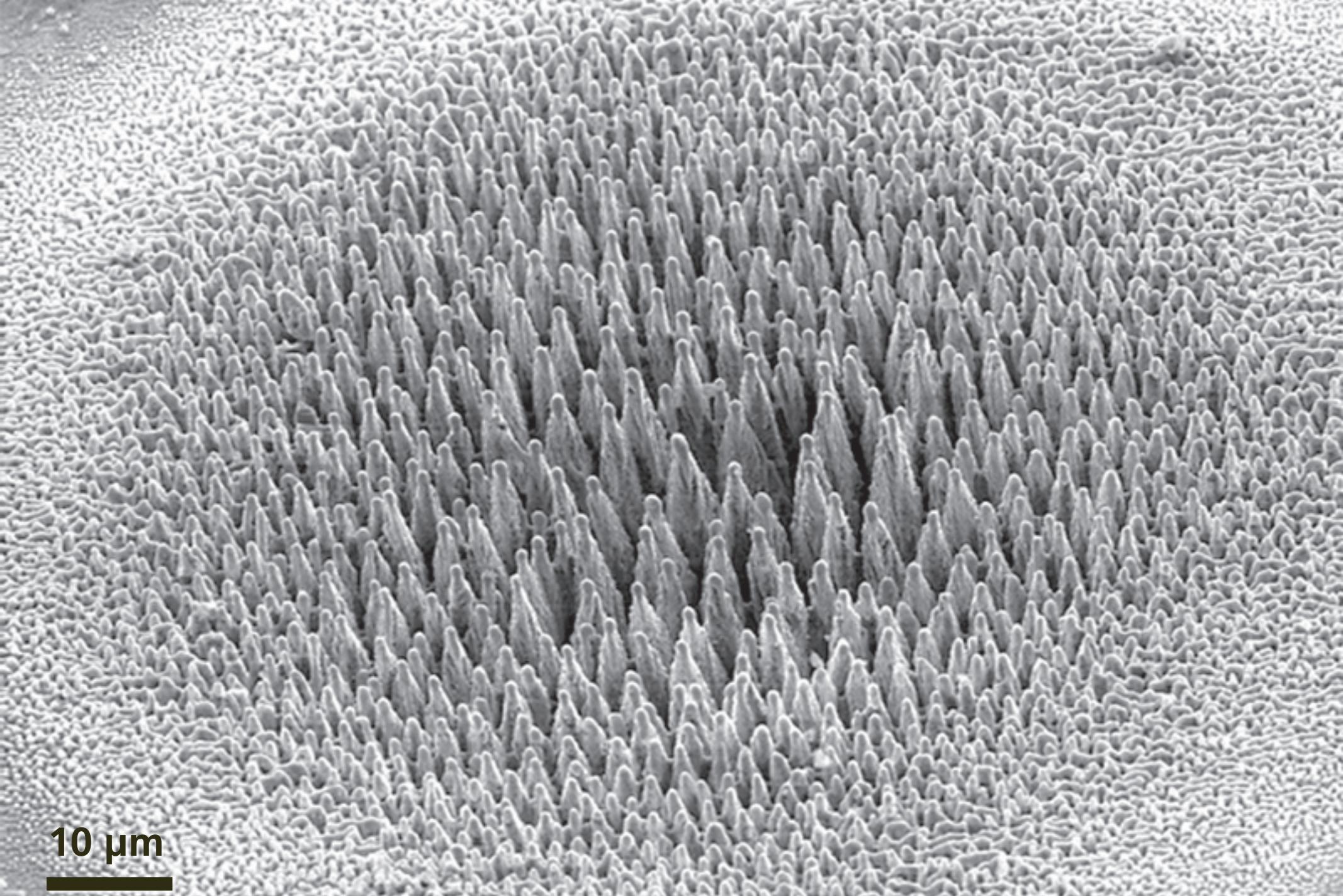
2 intermediate band



1 properties

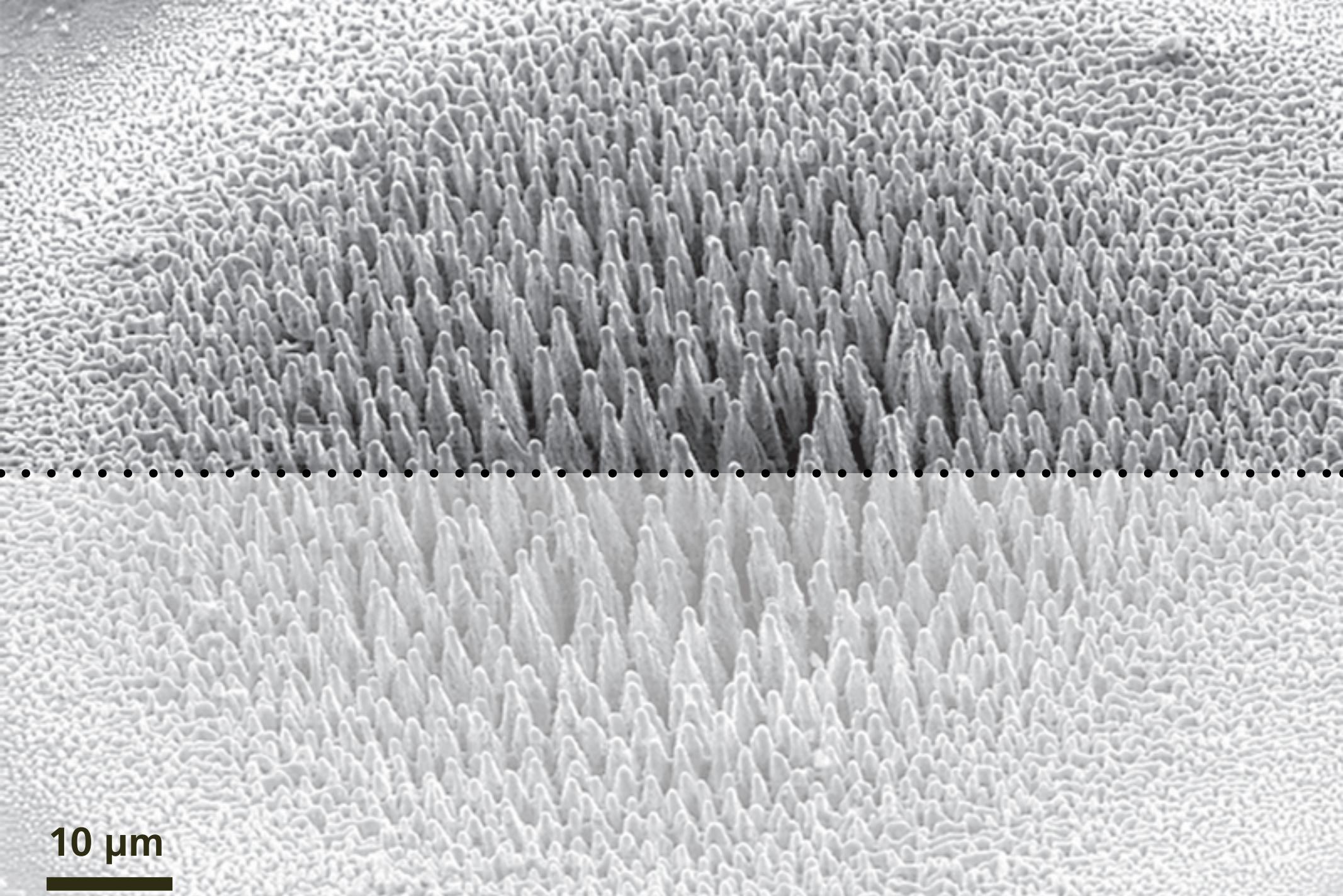
2 intermediate band

3 devices



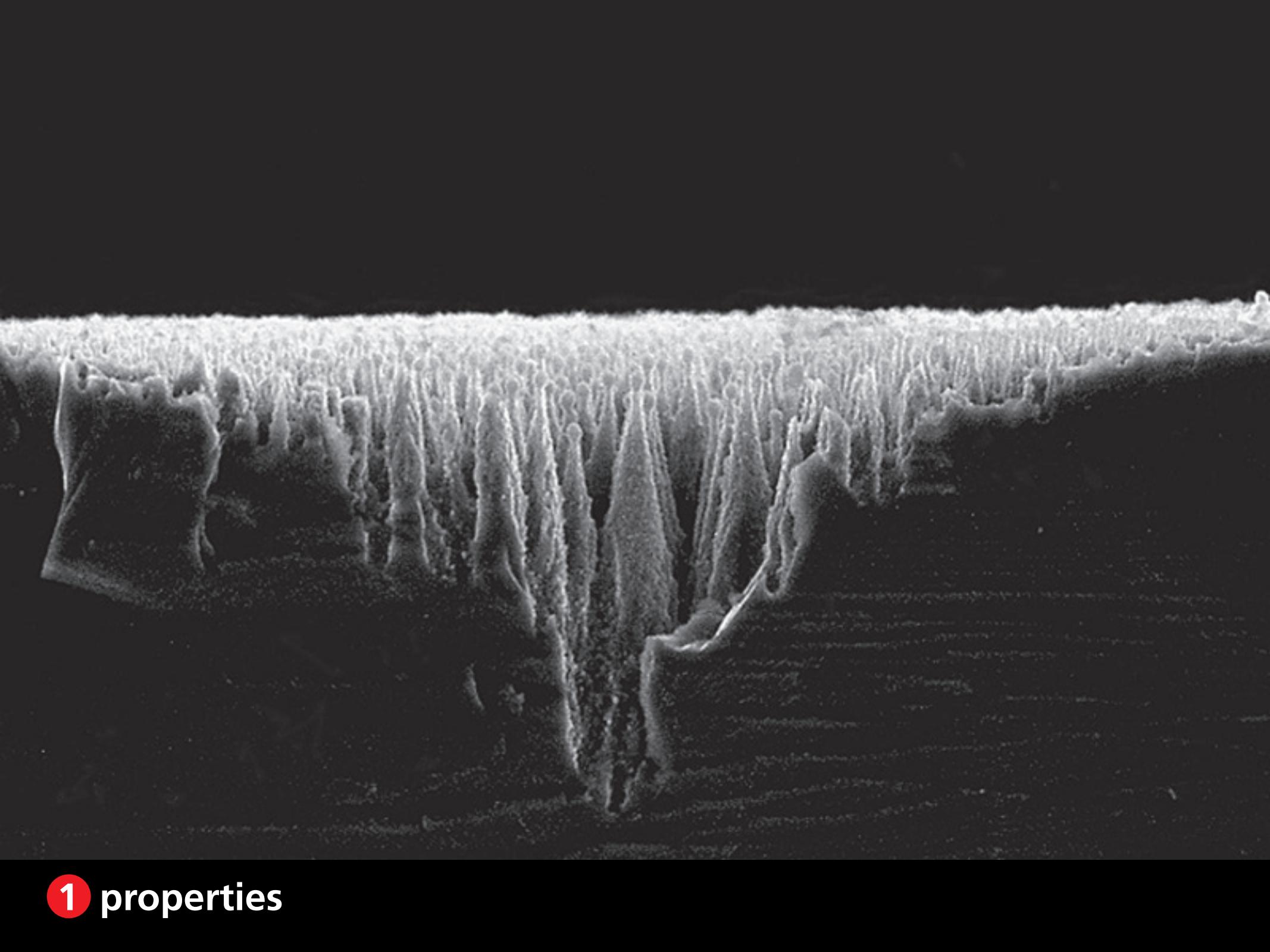
10 μm

1 properties

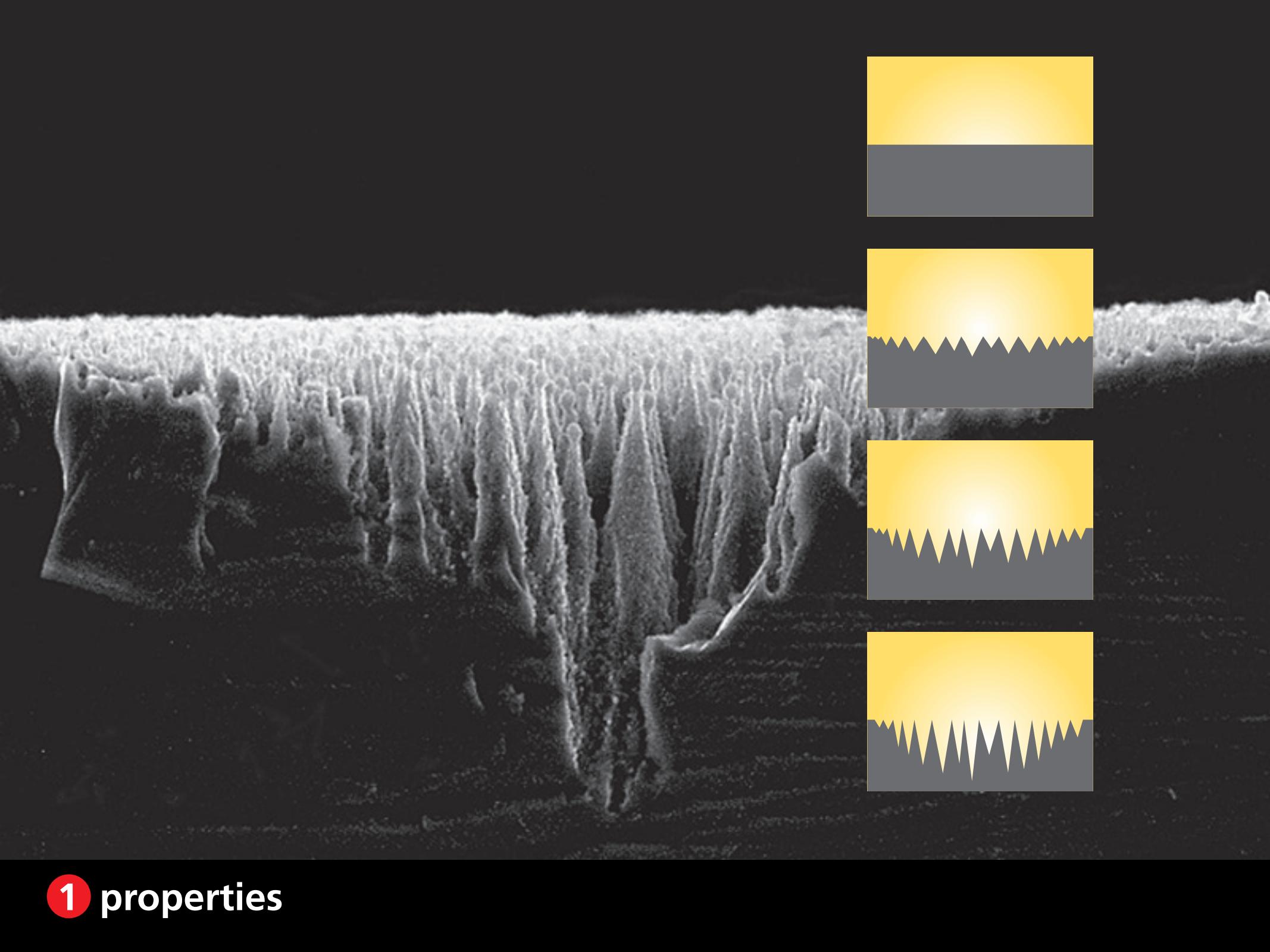


10 μm

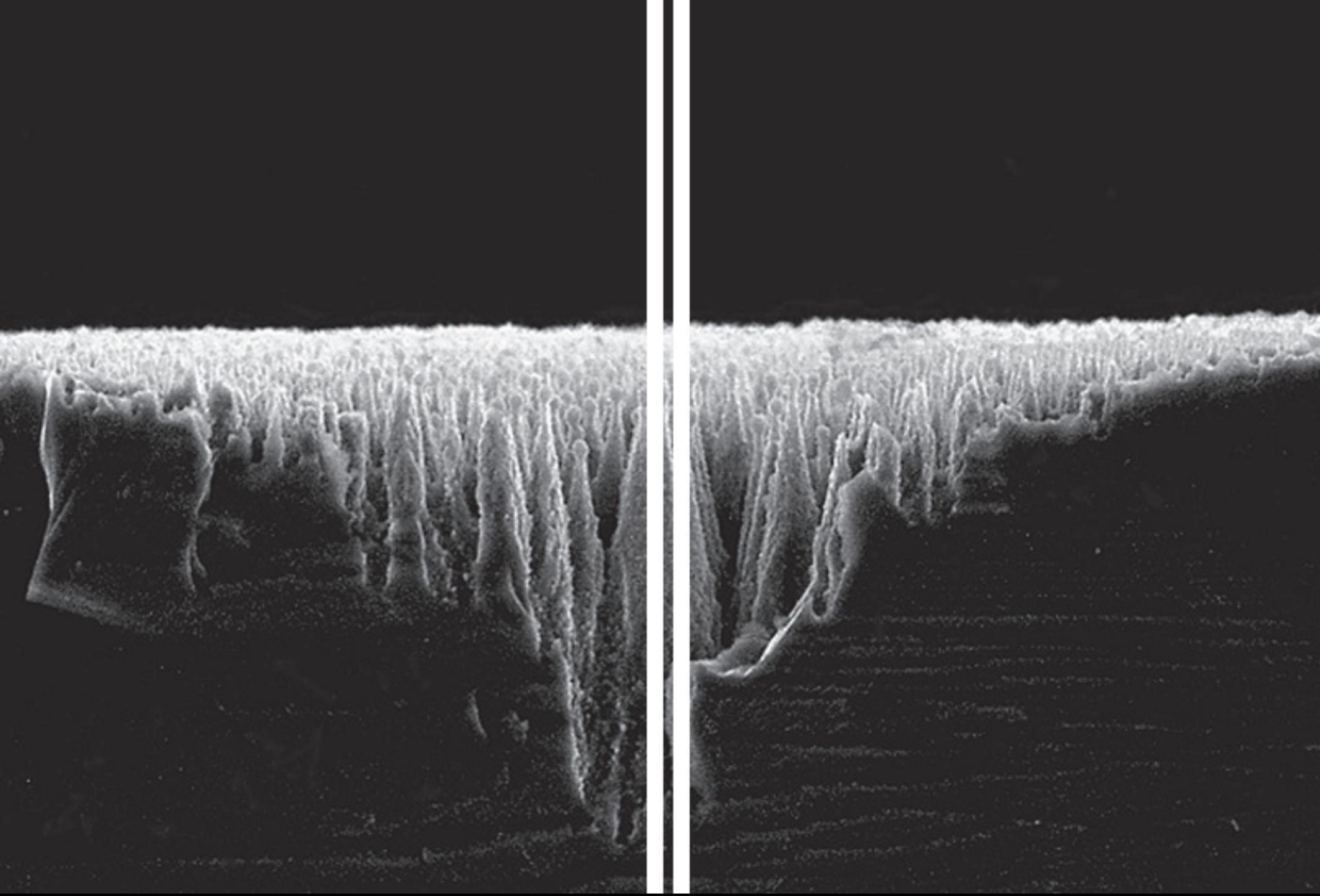
1 properties



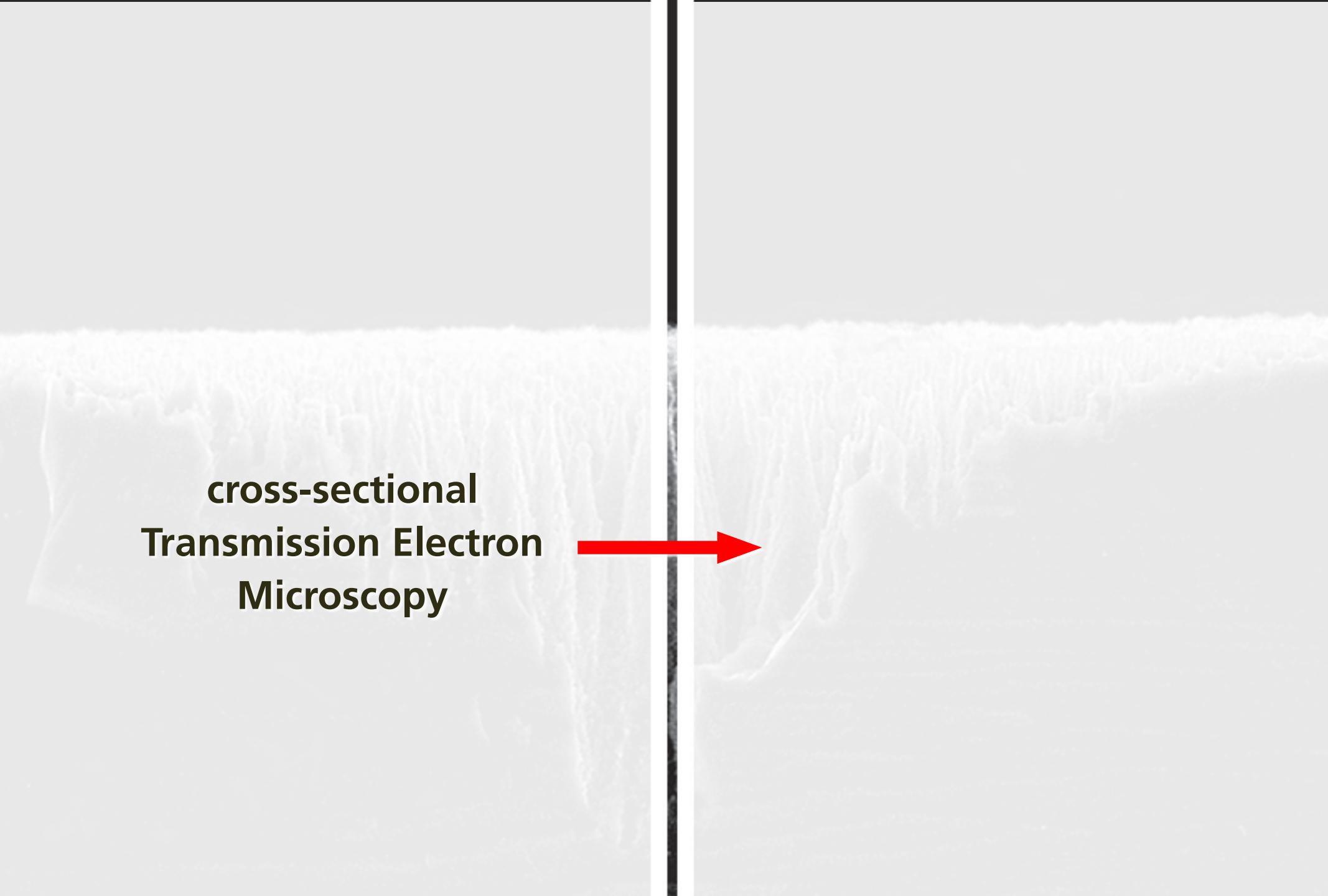
1 properties



1 properties



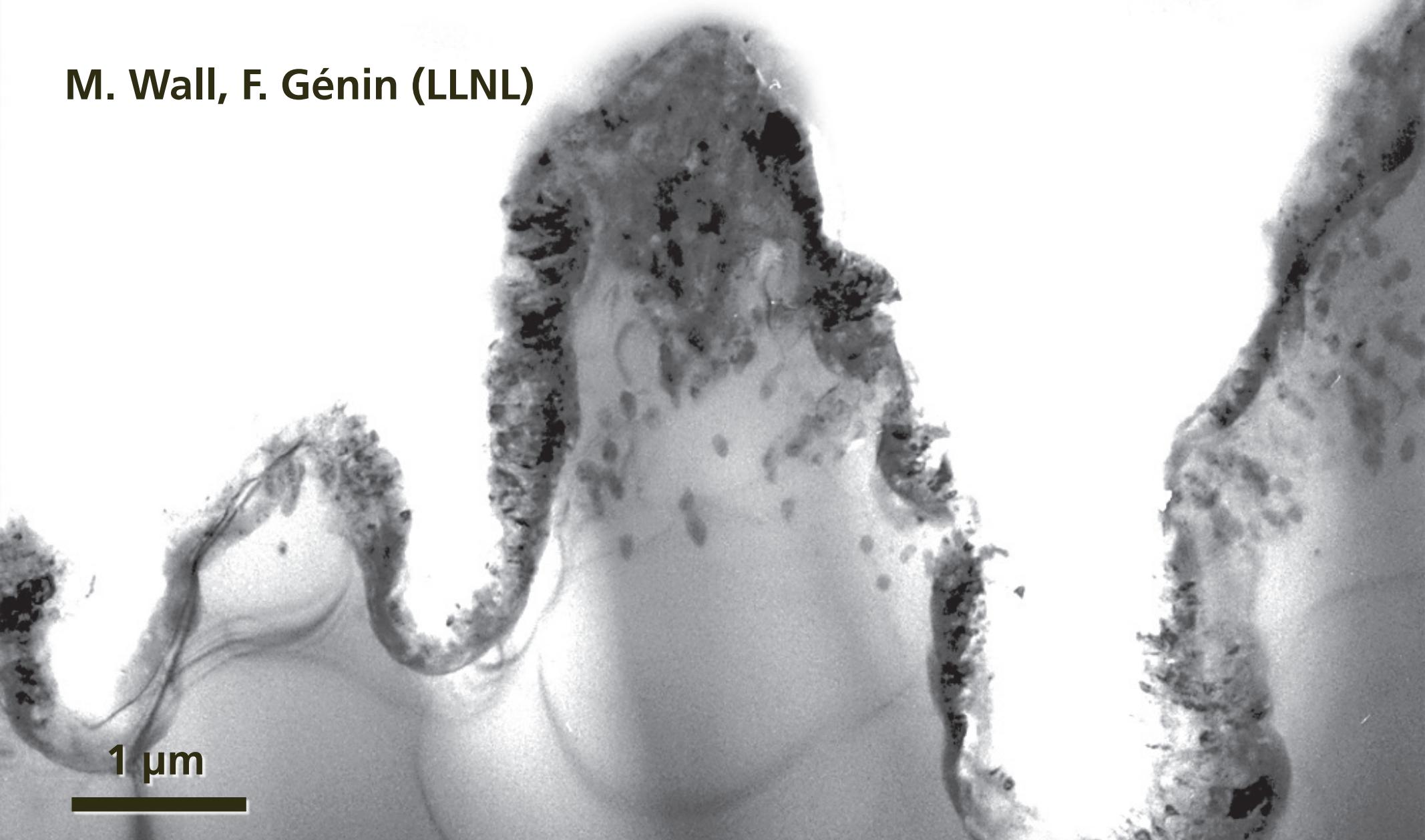
1 properties



**cross-sectional
Transmission Electron
Microscopy**

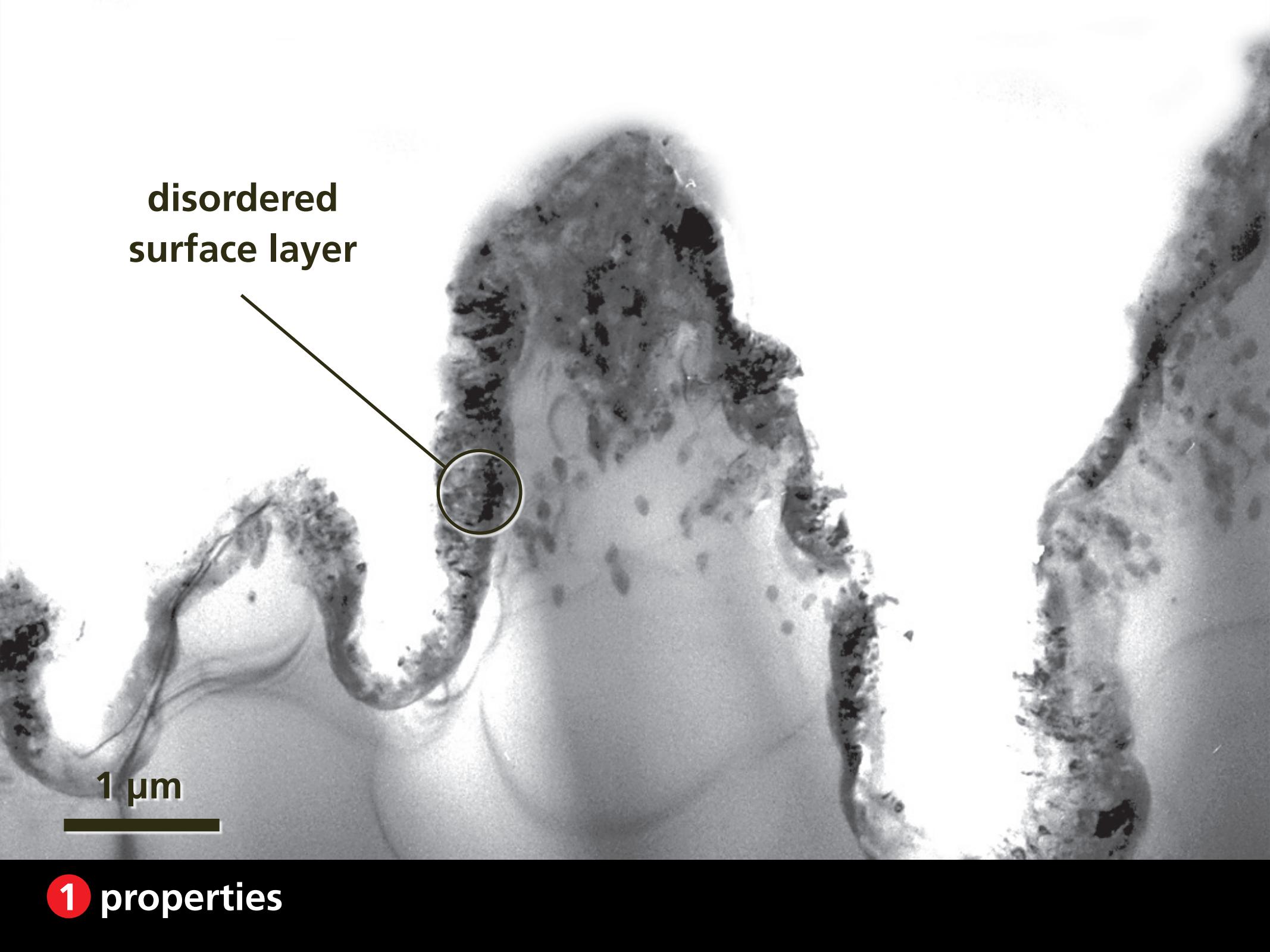


M. Wall, F. Génin (LLNL)



1 μm

1 properties

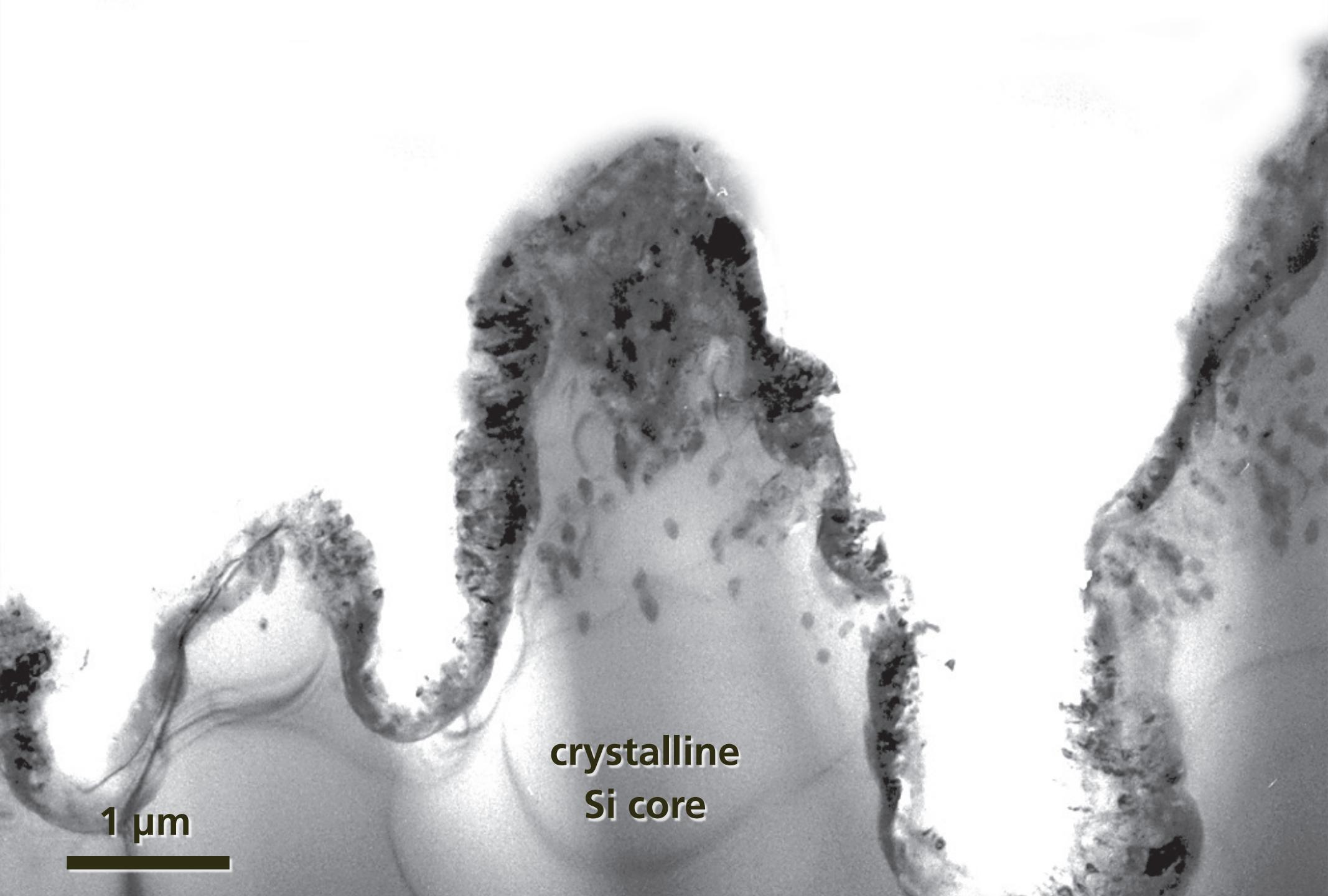


**disordered
surface layer**



1 μm

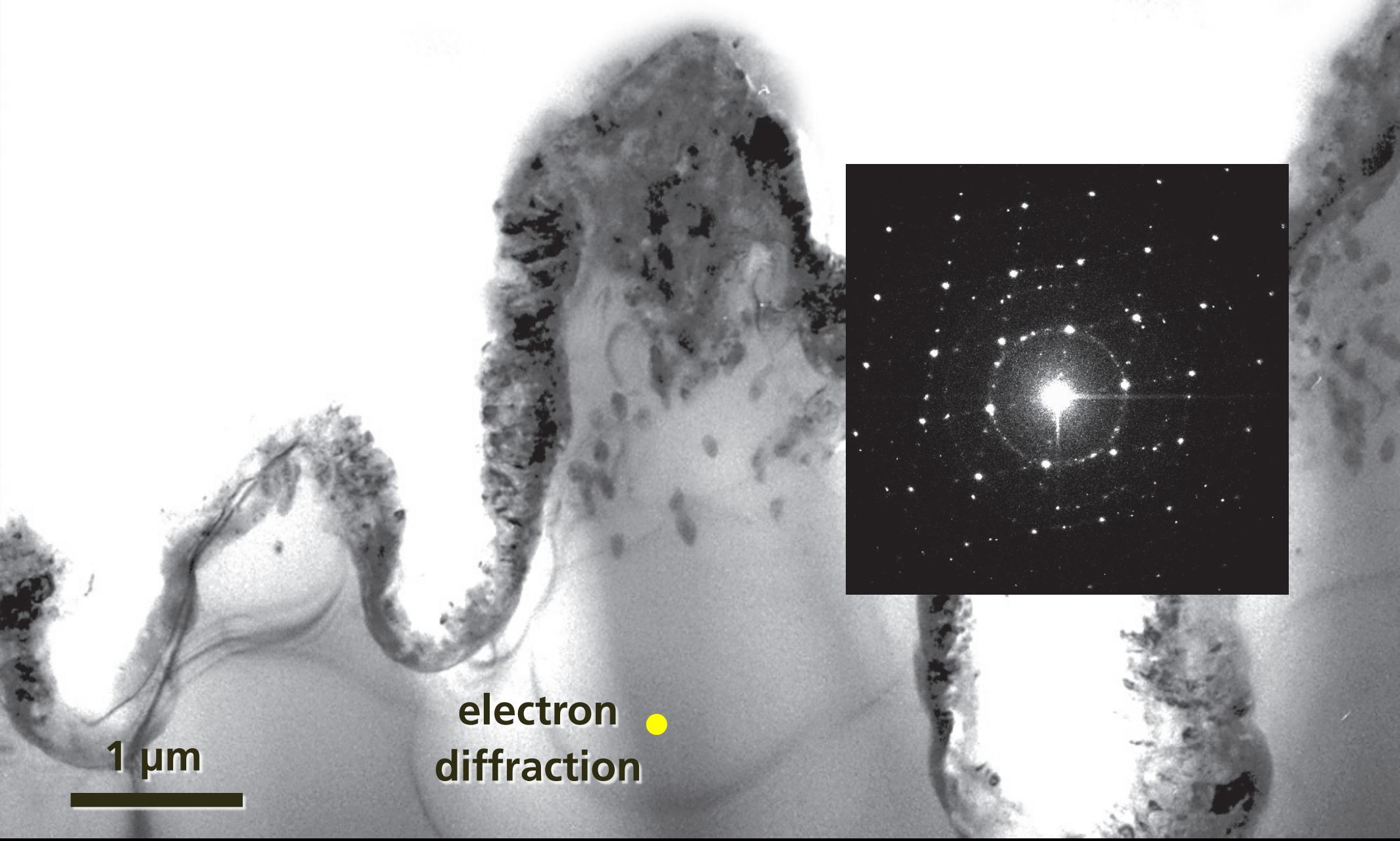
1 properties



1 μm

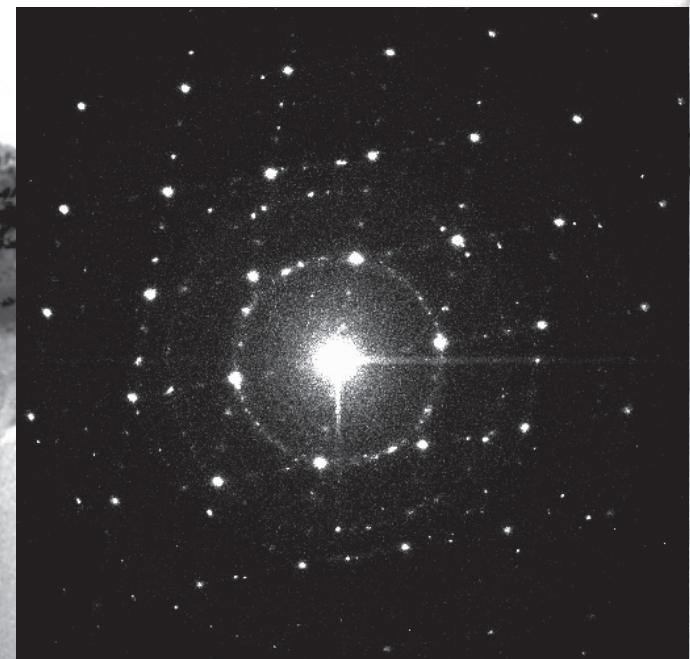
crystalline
Si core

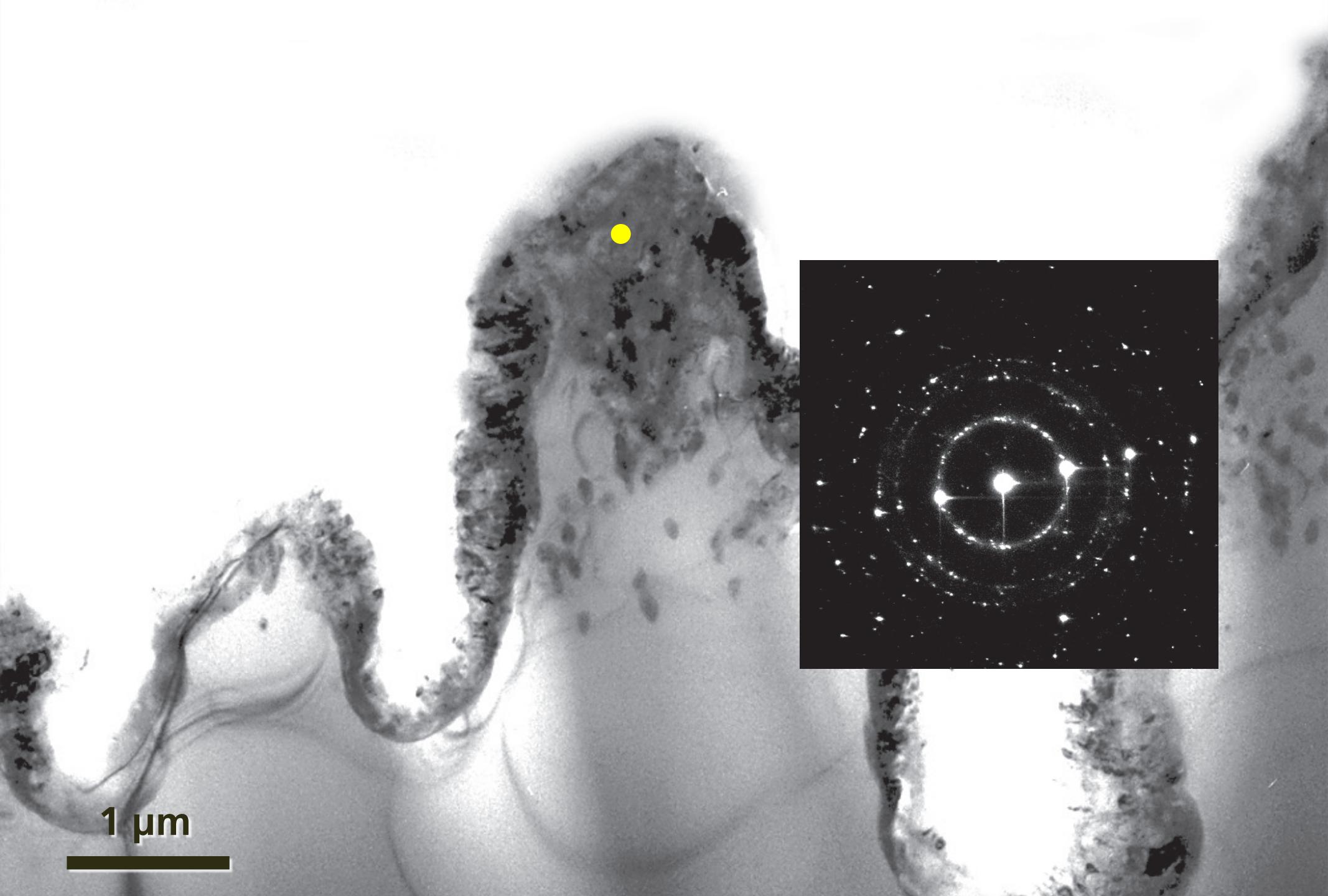
1 properties



electron
diffraction

1 properties





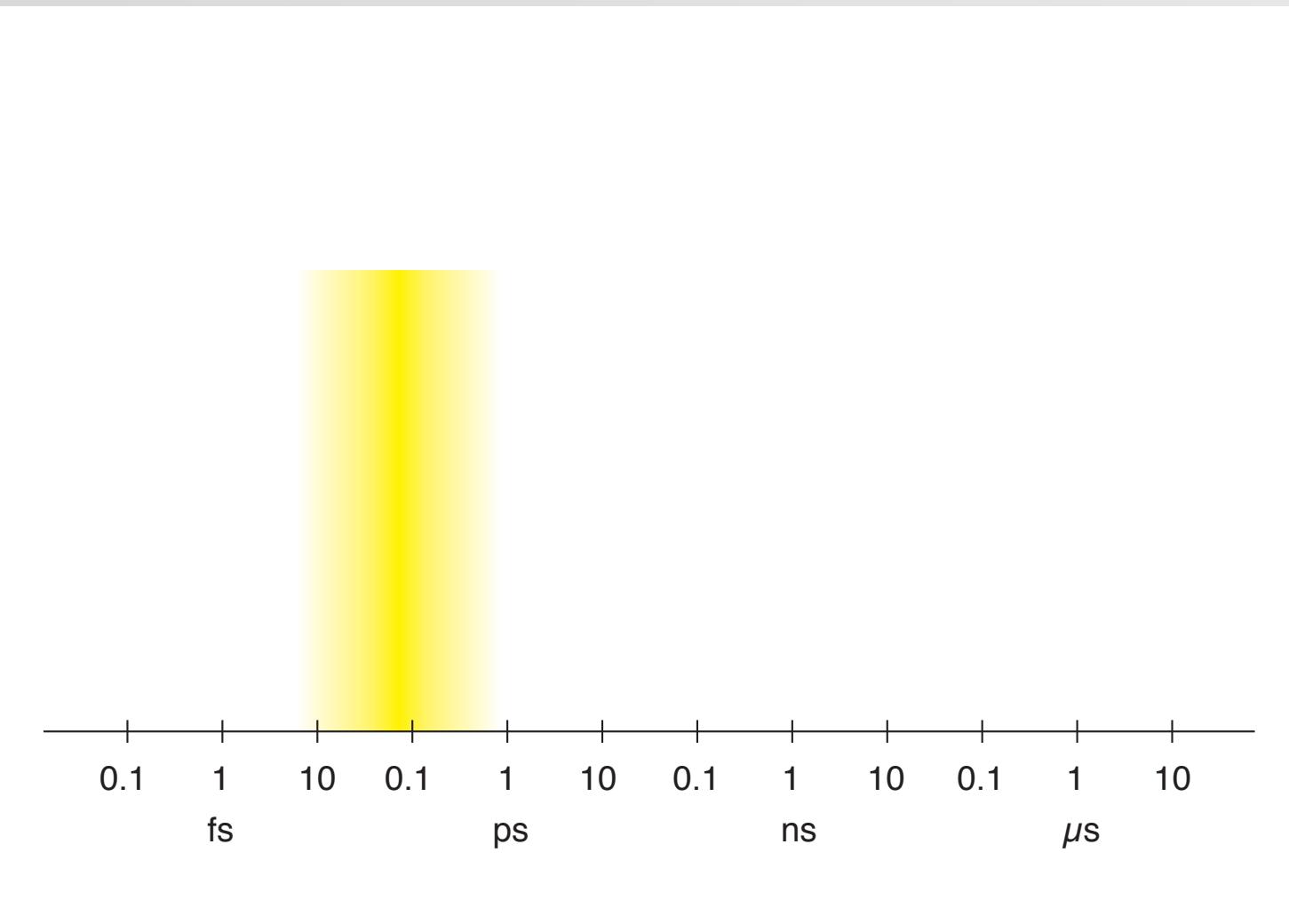
1 properties

- 300-nm disordered surface layer
- undisturbed crystalline core
- surface layer: polycrystalline Si with 1.6% sulfur

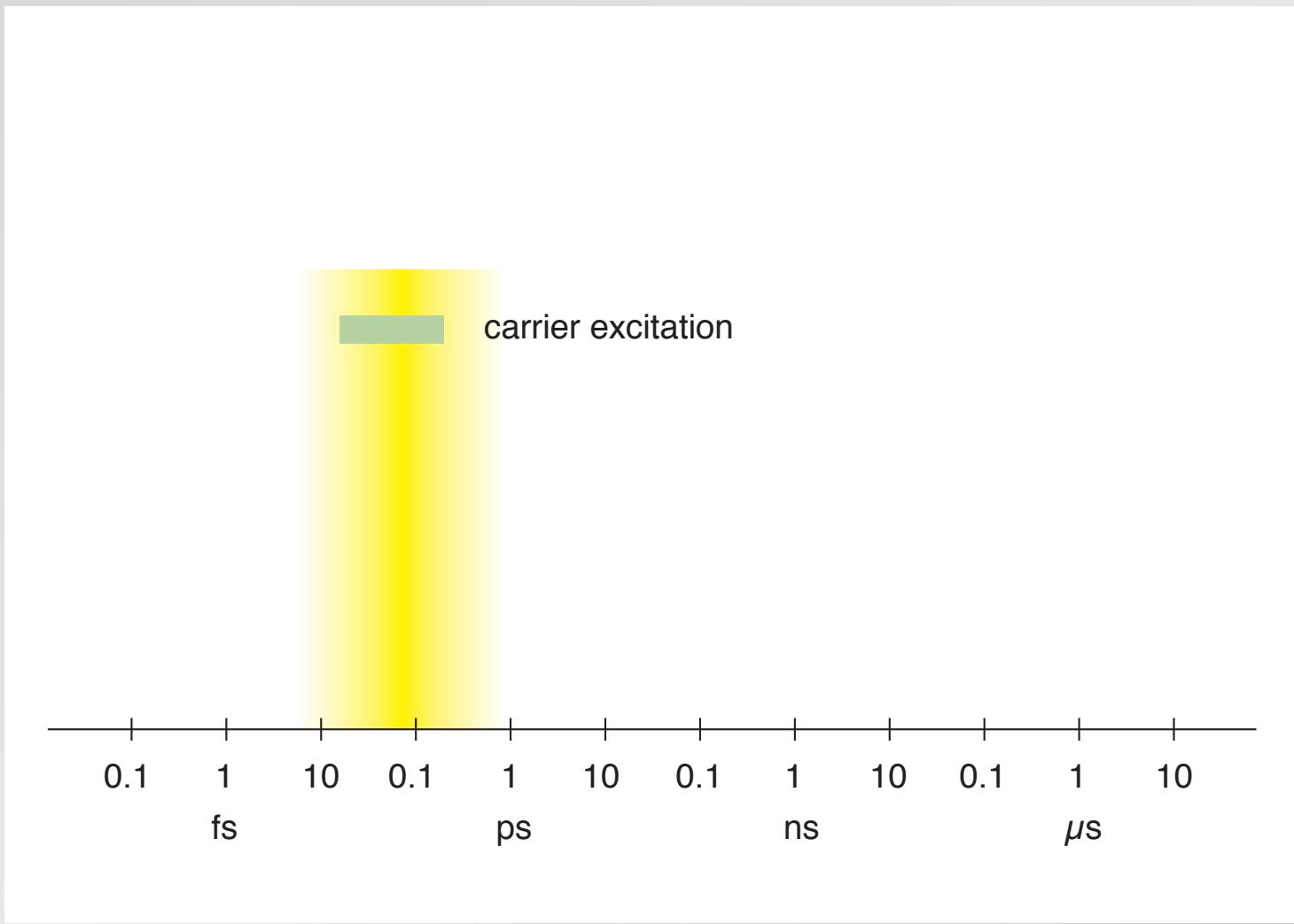
1 μm

two processes: melting and ablation

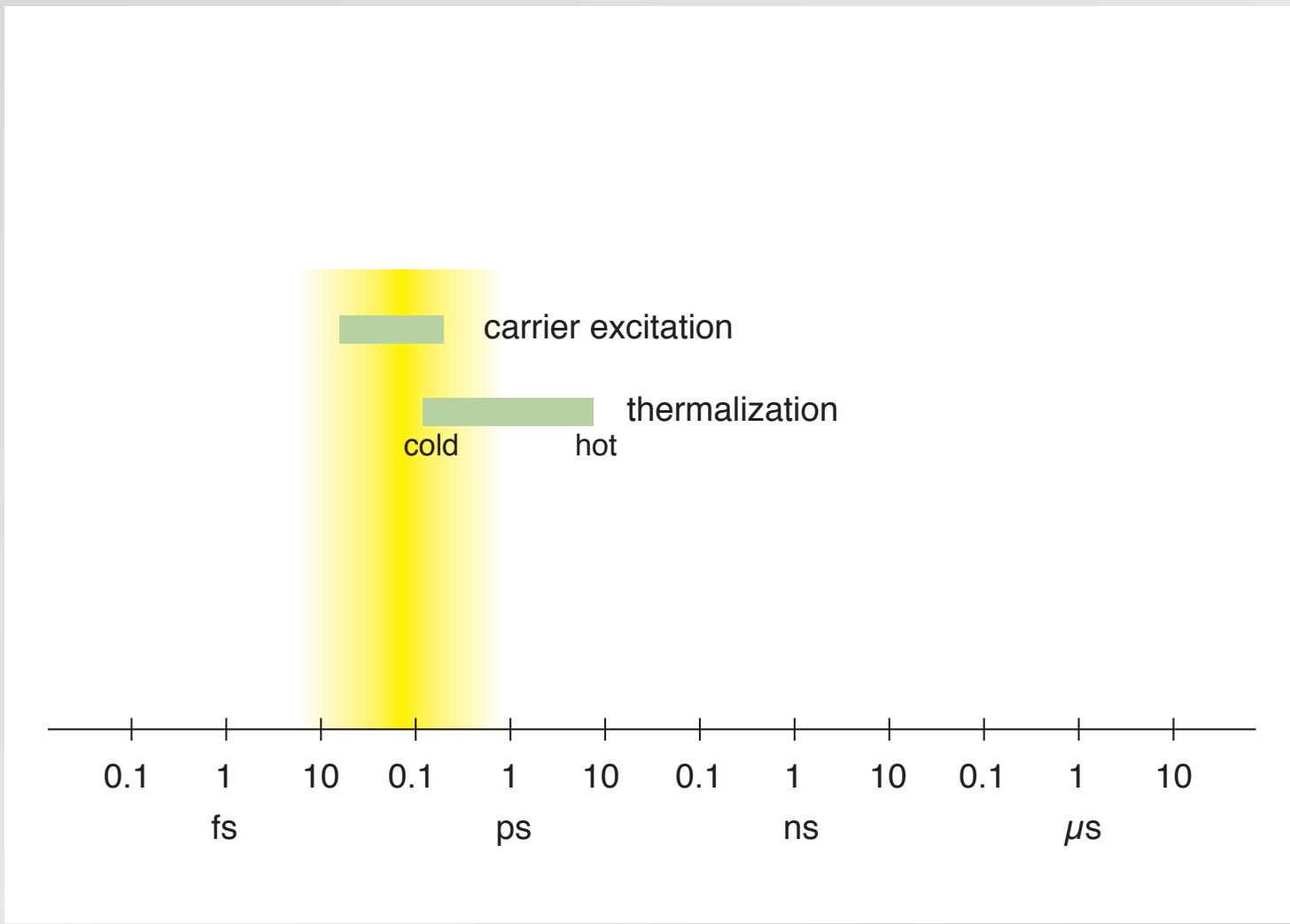
relevant time scales



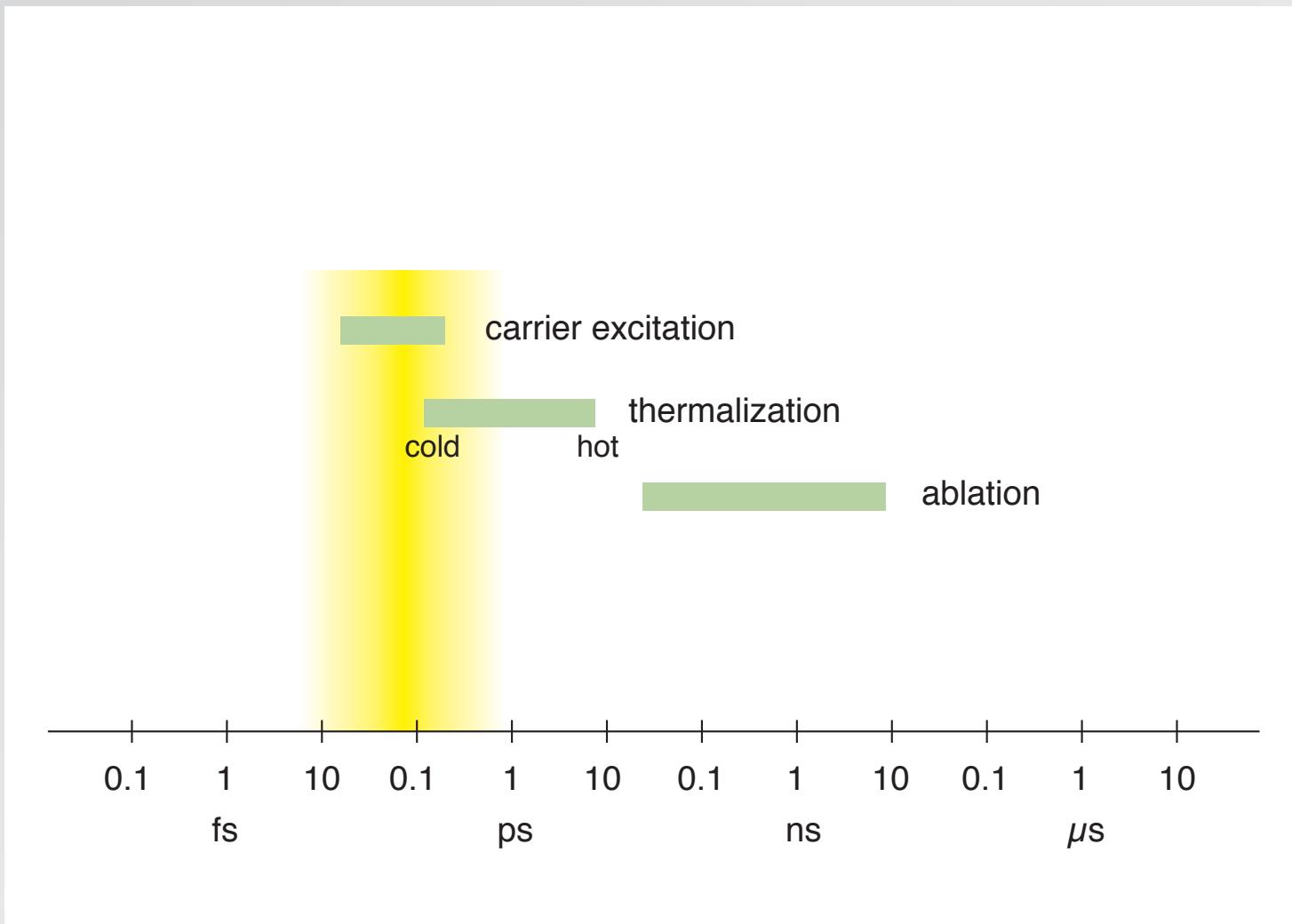
relevant time scales



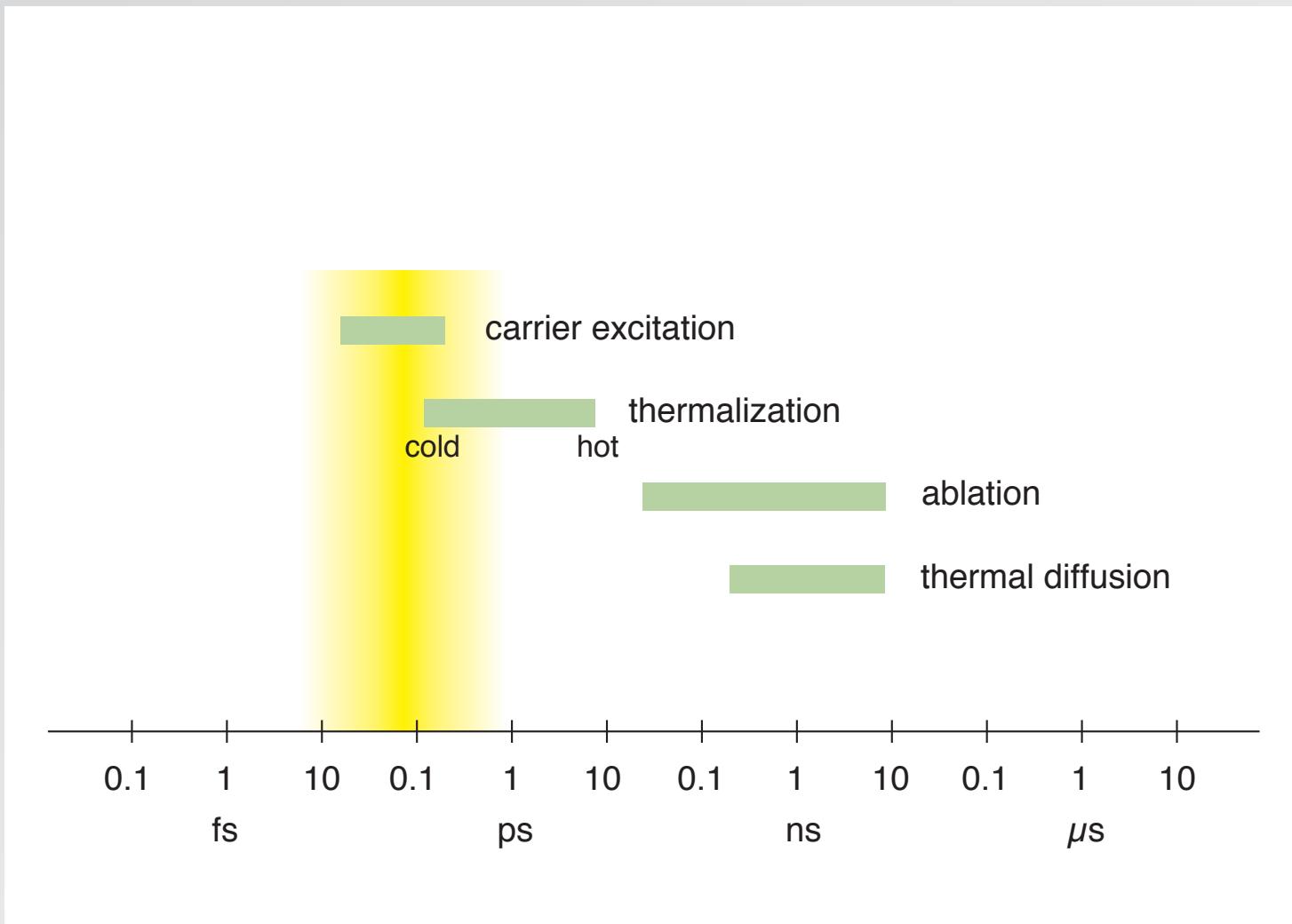
relevant time scales



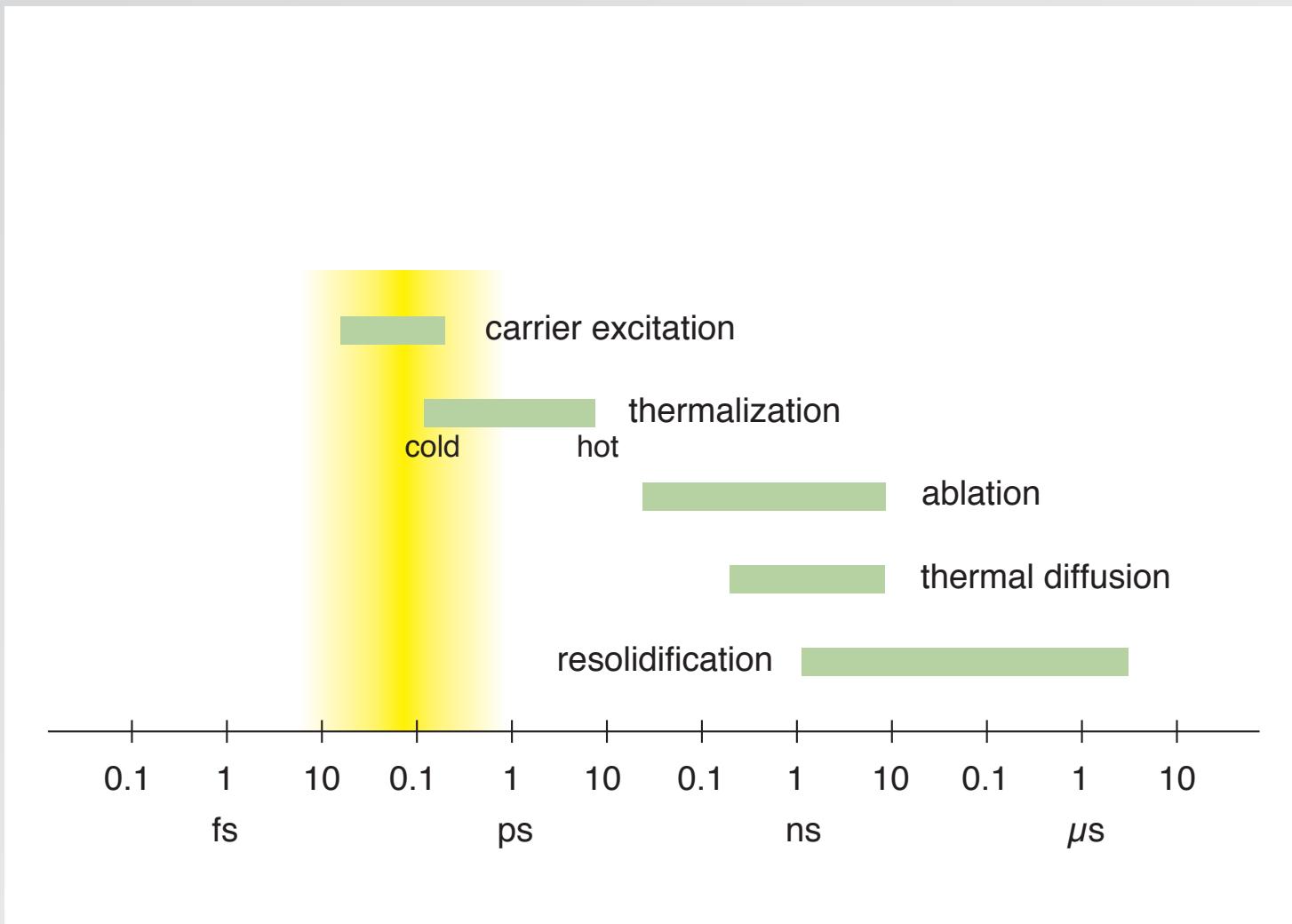
relevant time scales



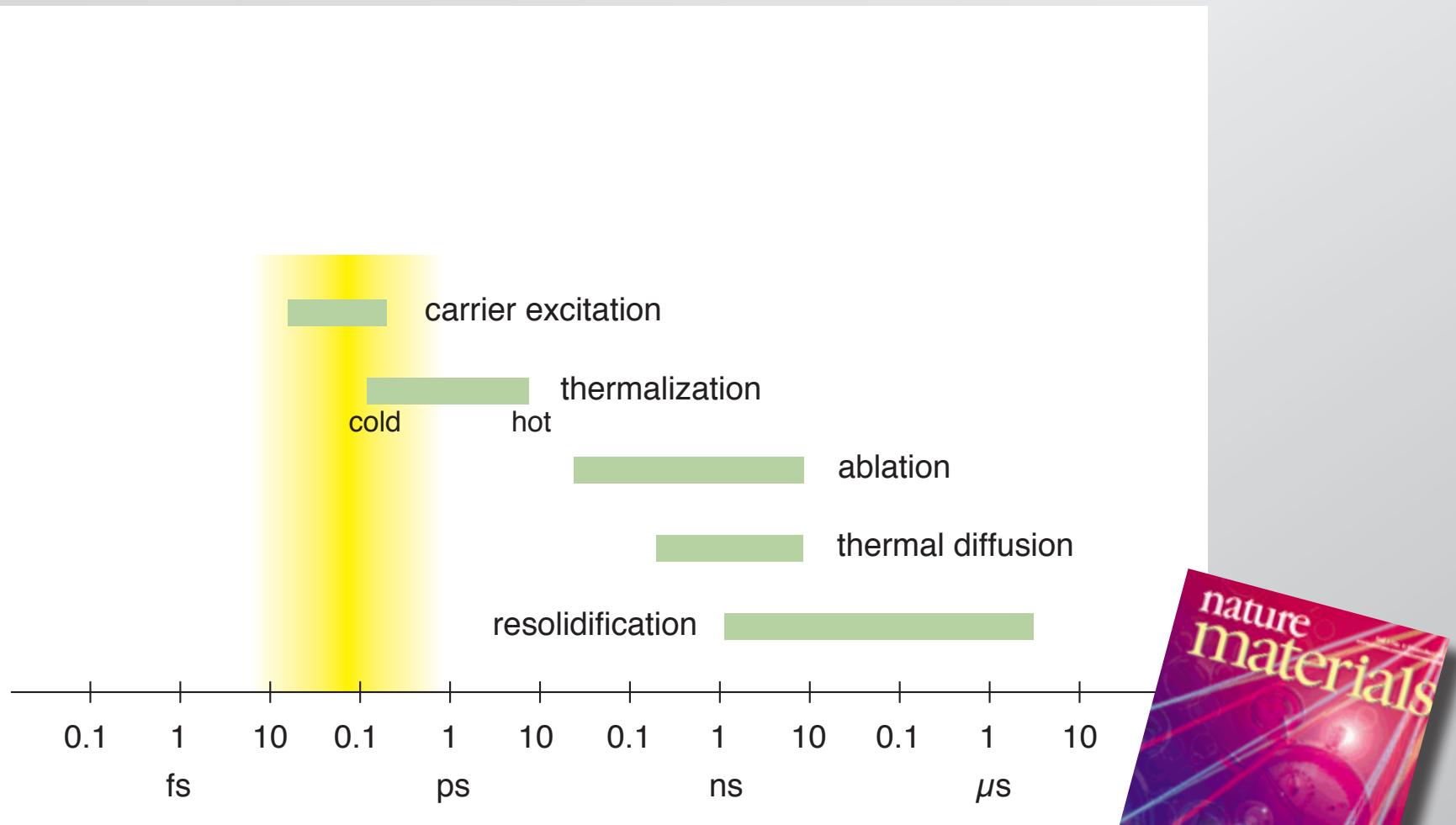
relevant time scales



relevant time scales



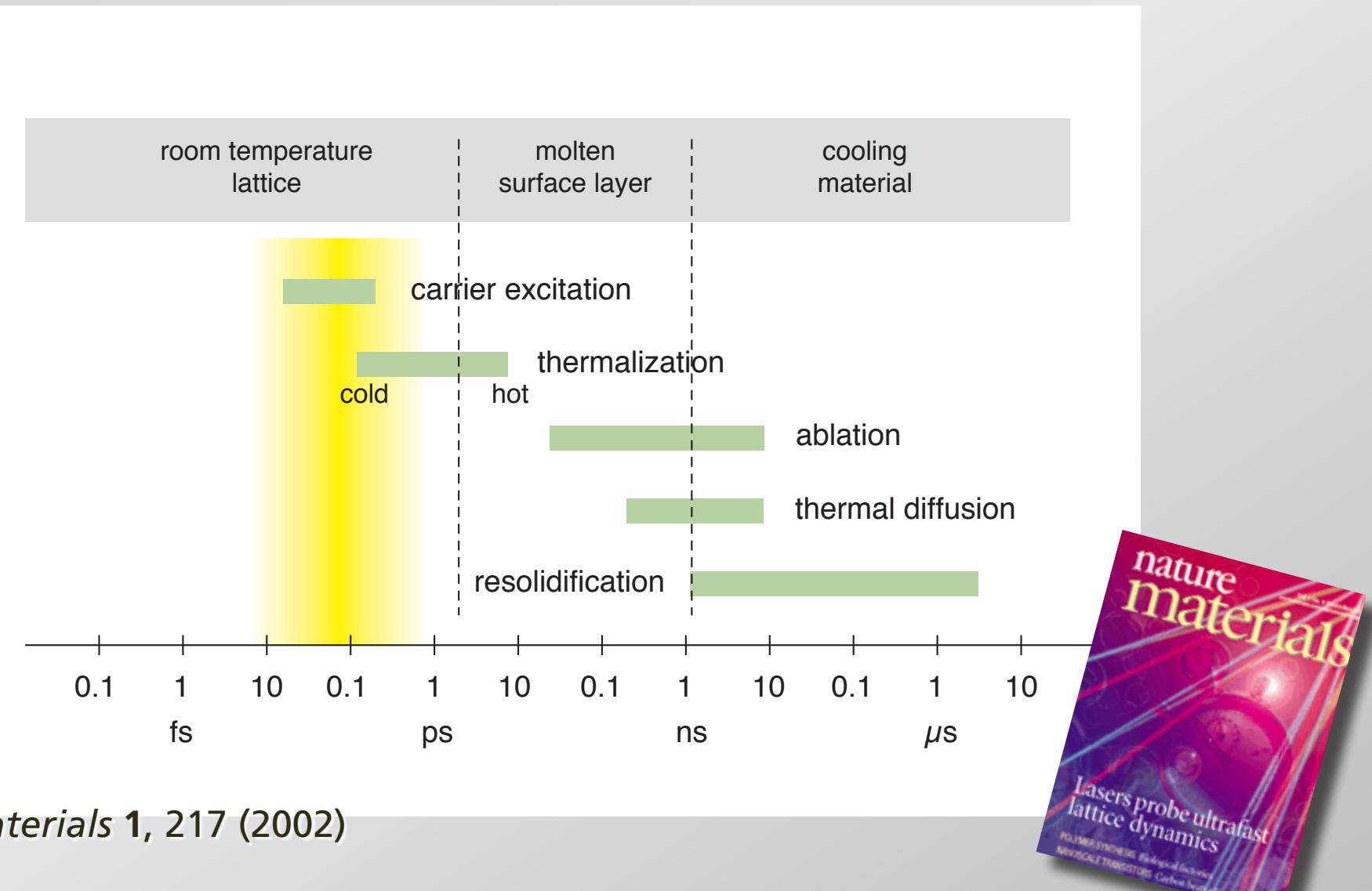
relevant time scales



Nature Materials 1, 217 (2002)

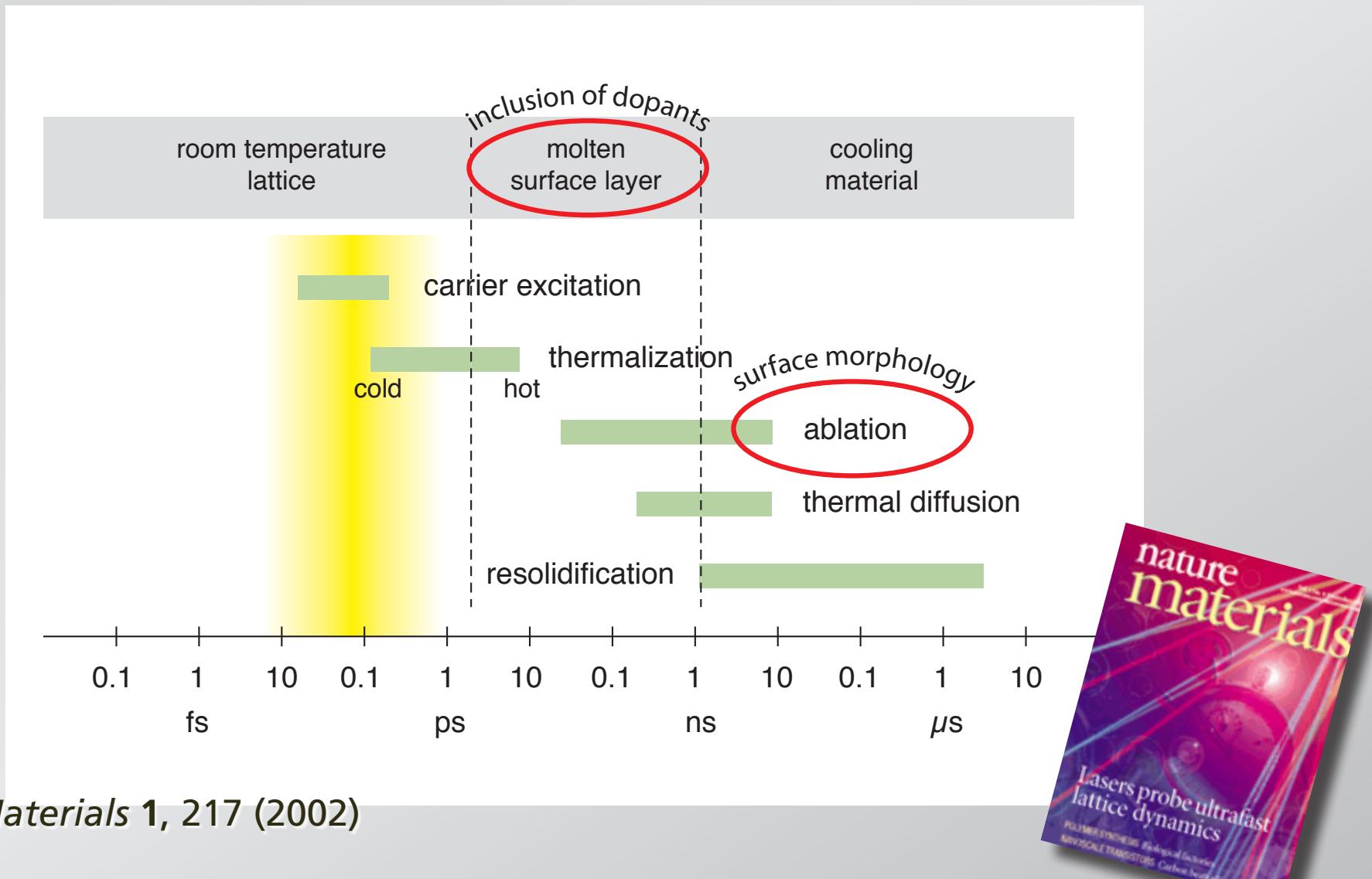


relevant time scales



Nature Materials 1, 217 (2002)

relevant time scales



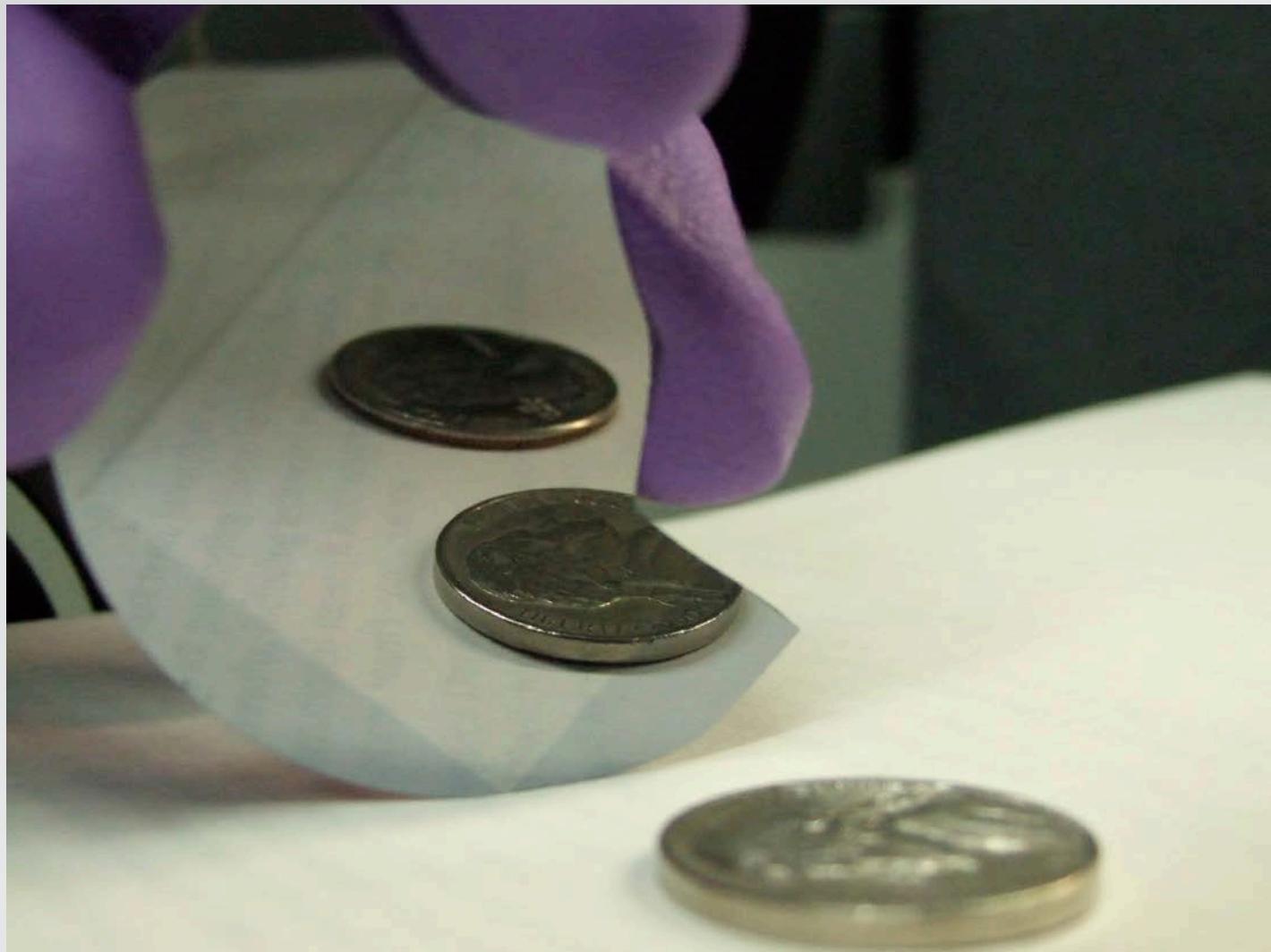
Nature Materials 1, 217 (2002)

different thresholds:

melting: 1.5 kJ/m^2

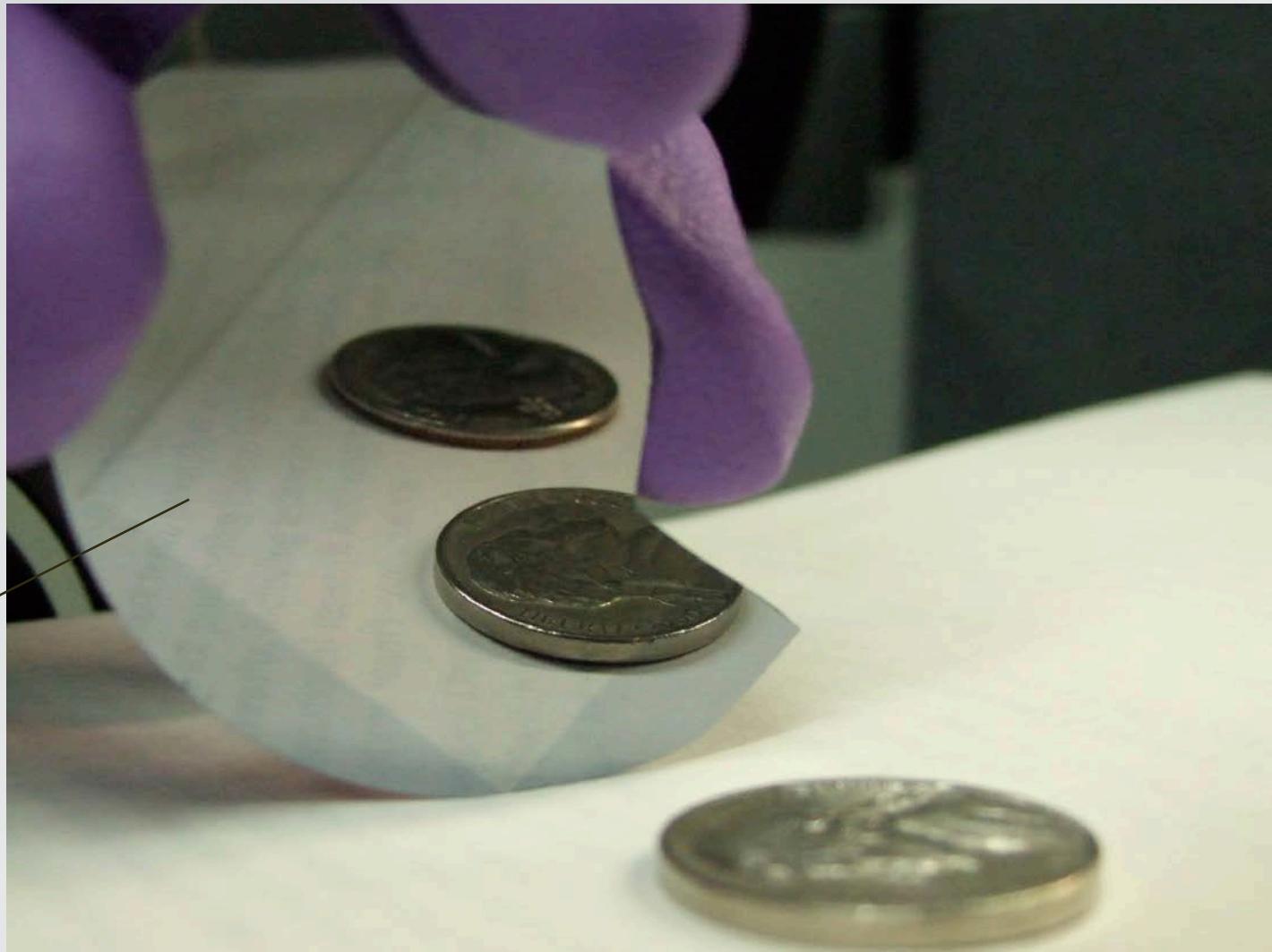
ablation: 3.1 kJ/m^2

decouple ablation from melting



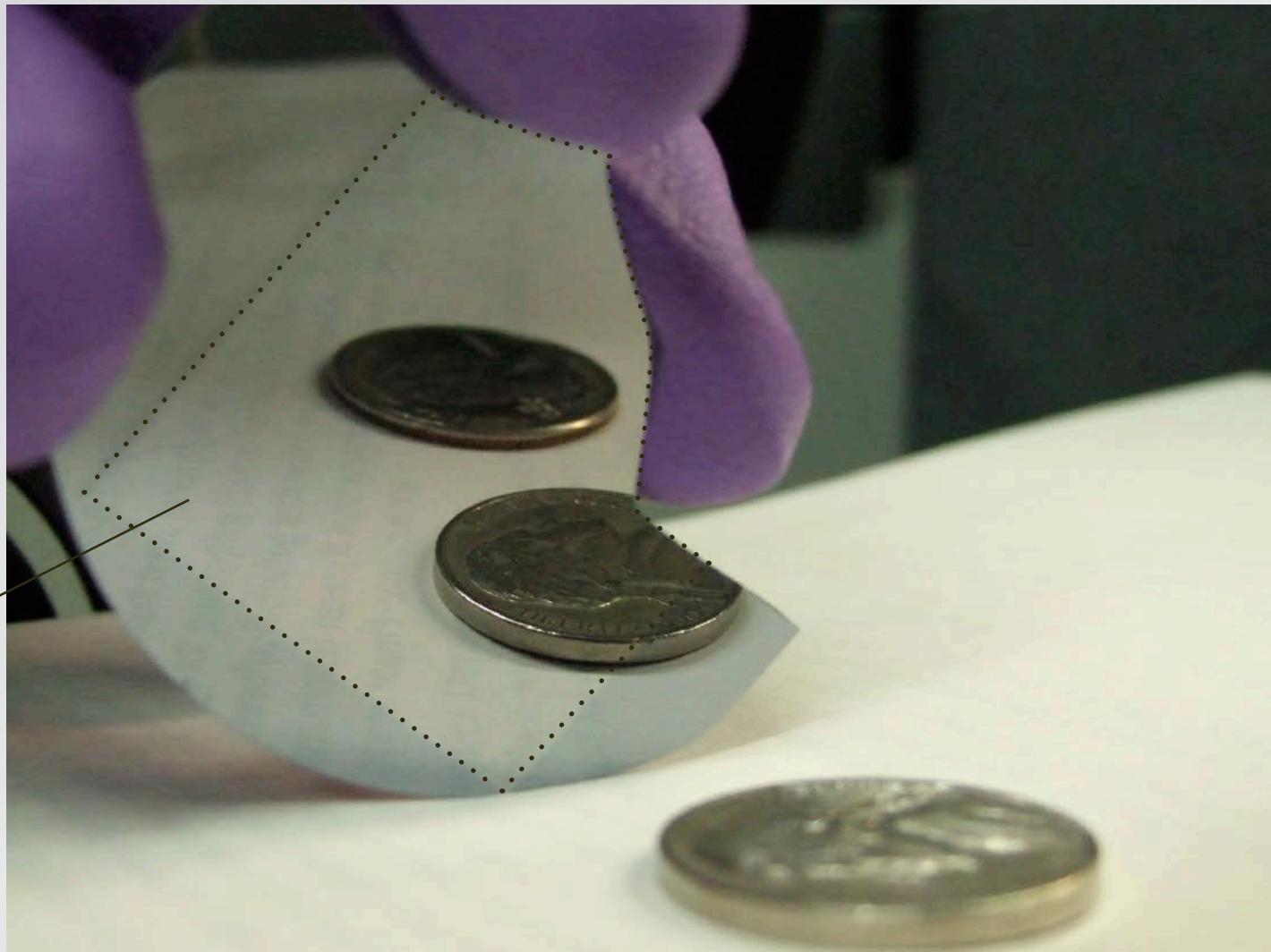
decouple ablation from melting

doped



1 properties

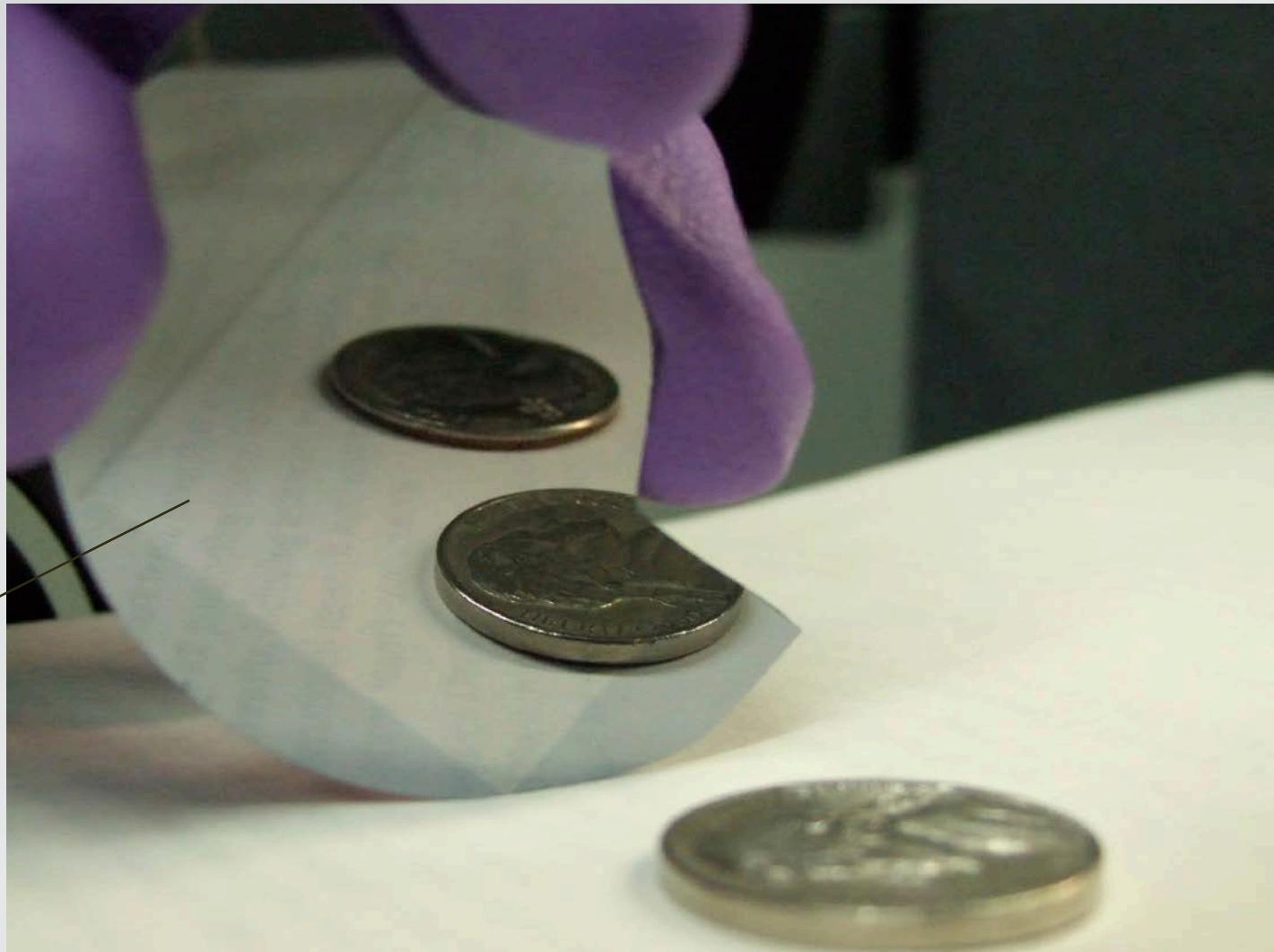
decouple ablation from melting



1 properties

decouple ablation from melting

doped

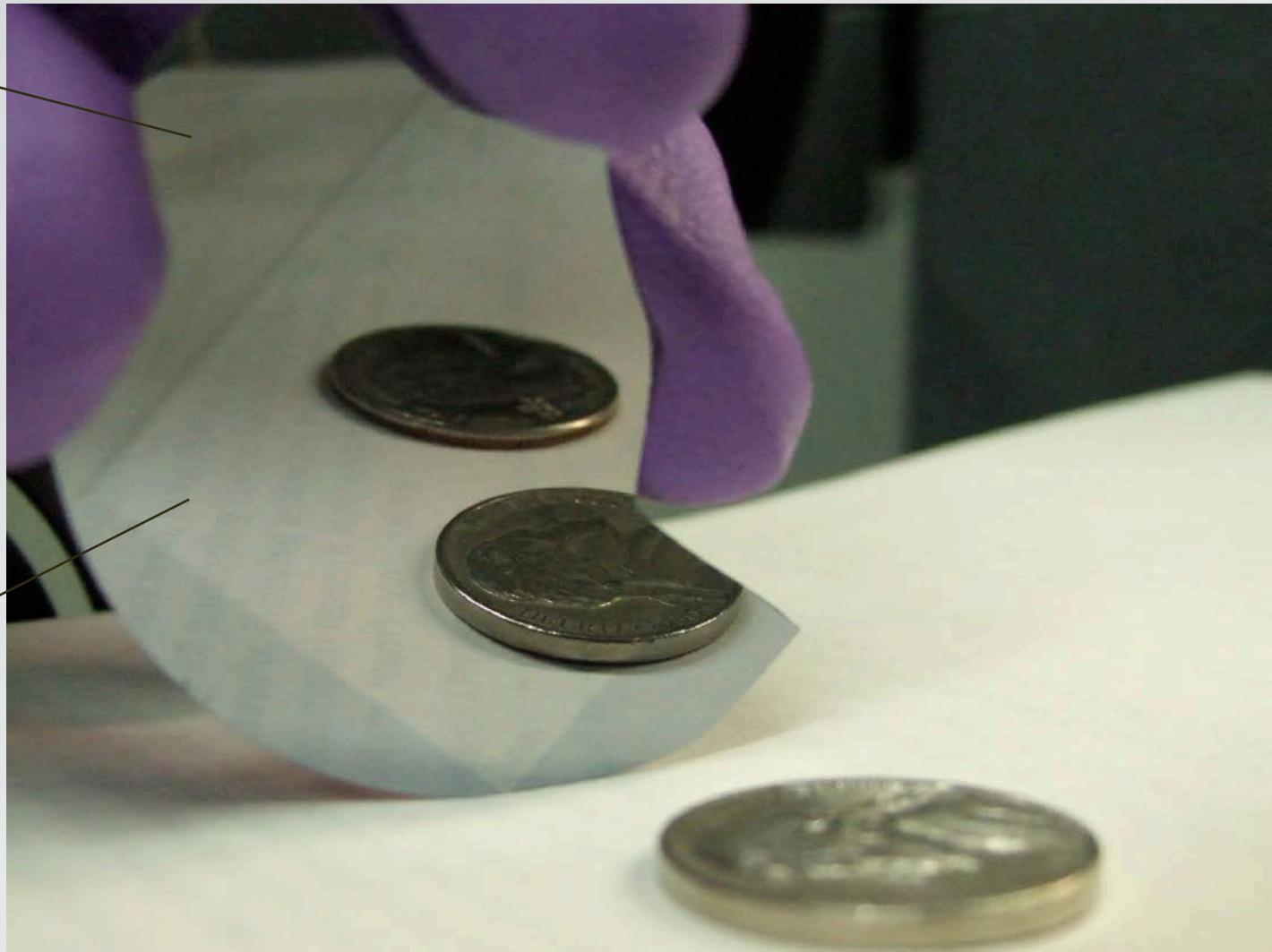


1 properties

decouple ablation from melting

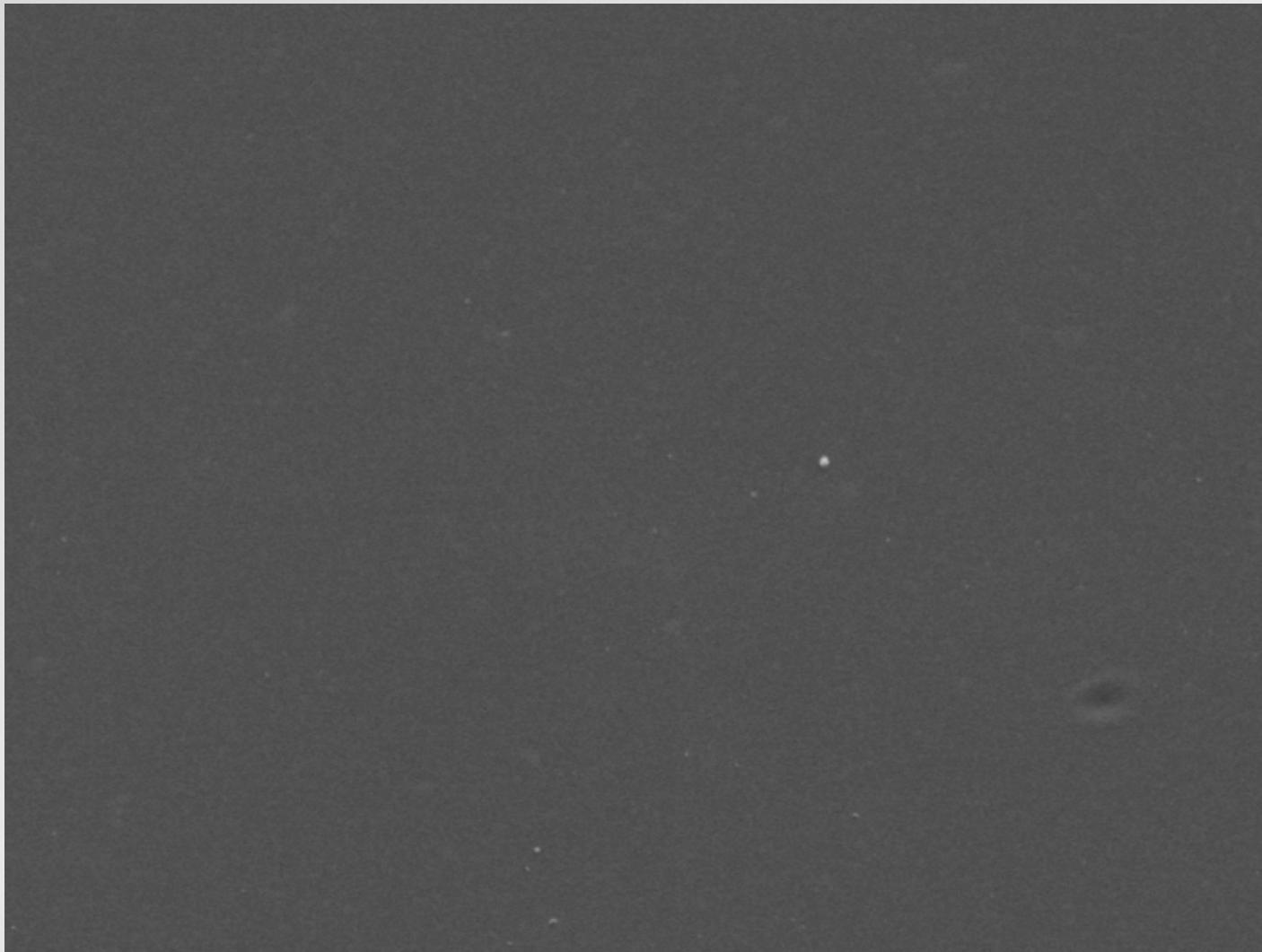
undoped

doped

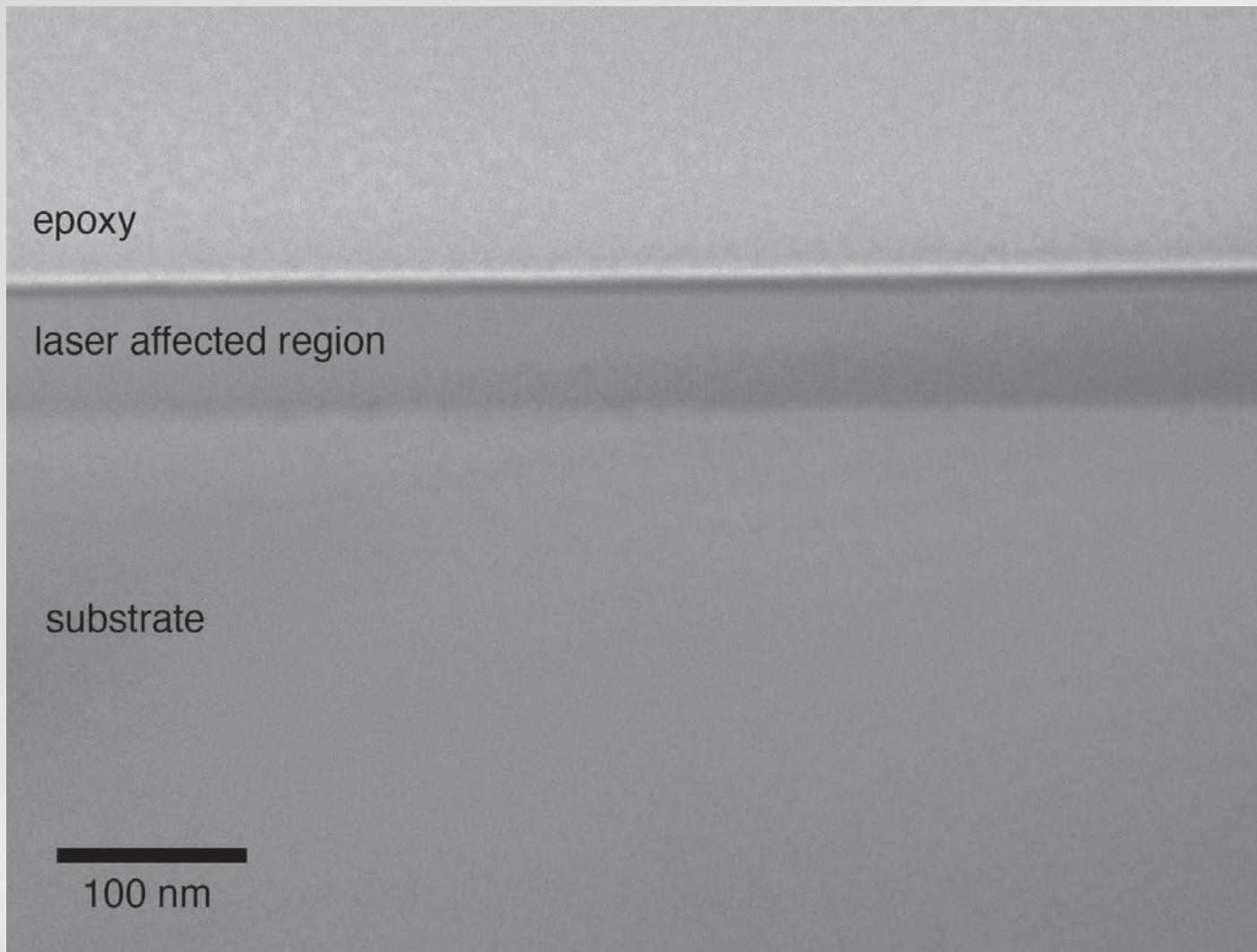


1 properties

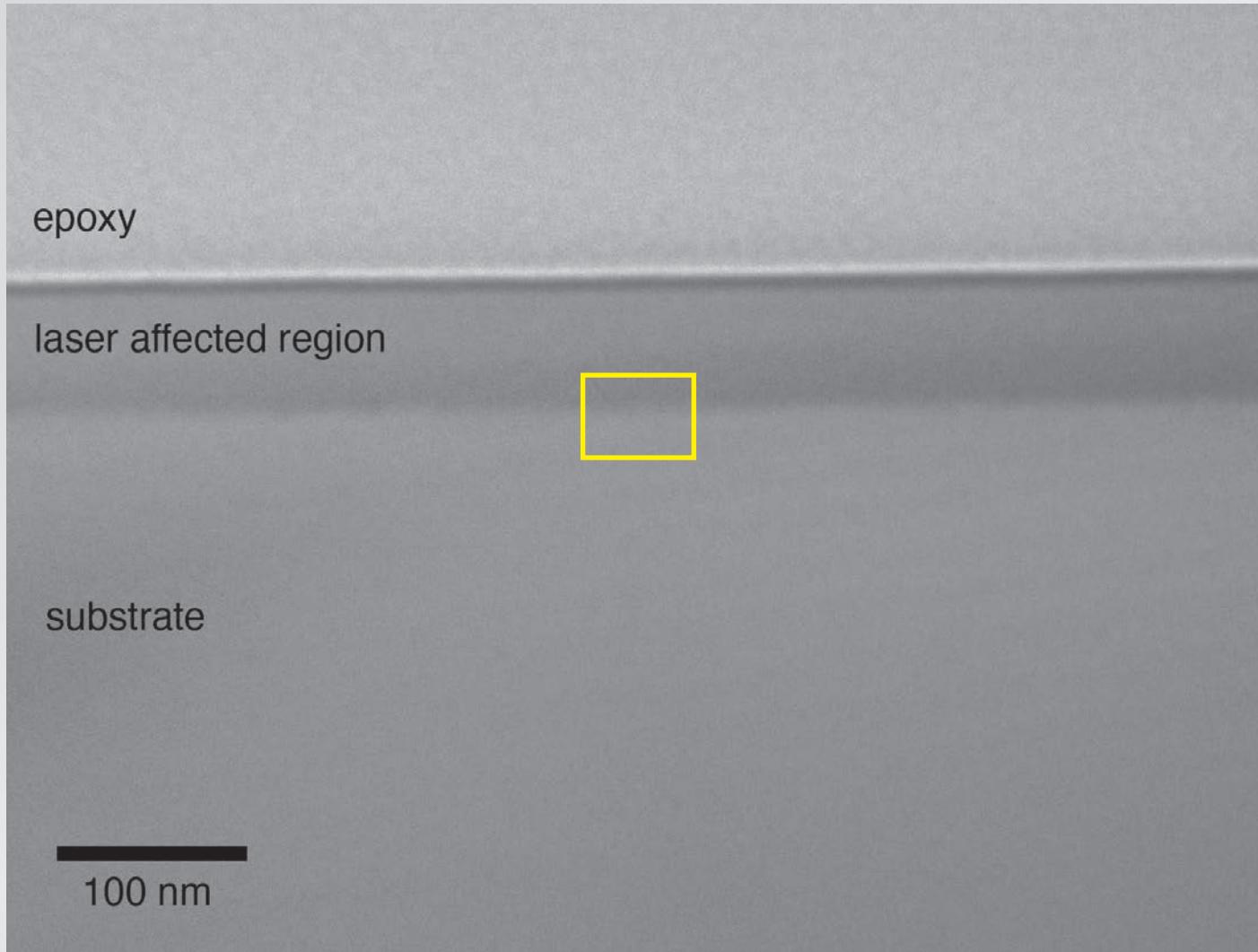
decouple ablation from melting



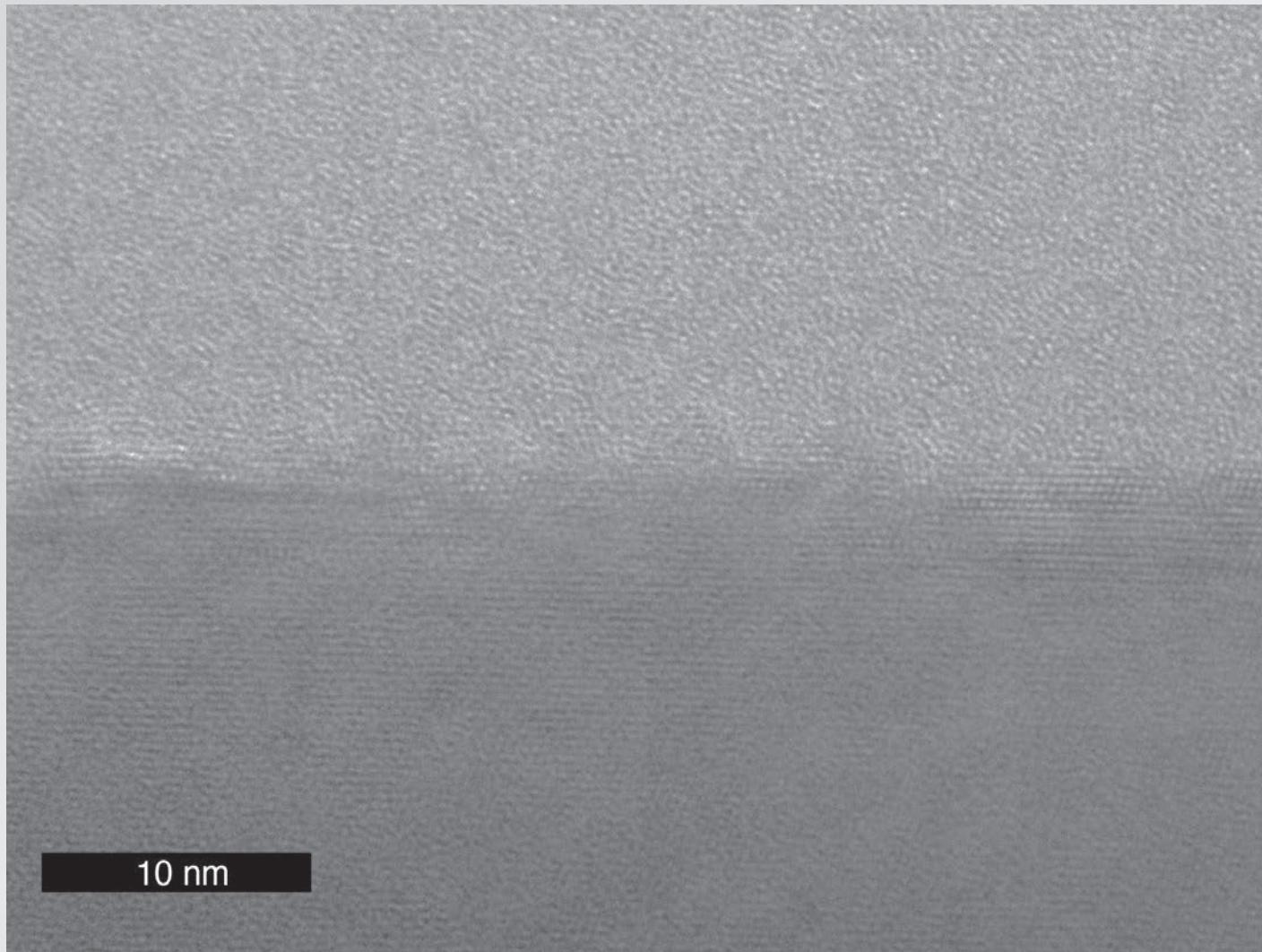
decouple ablation from melting



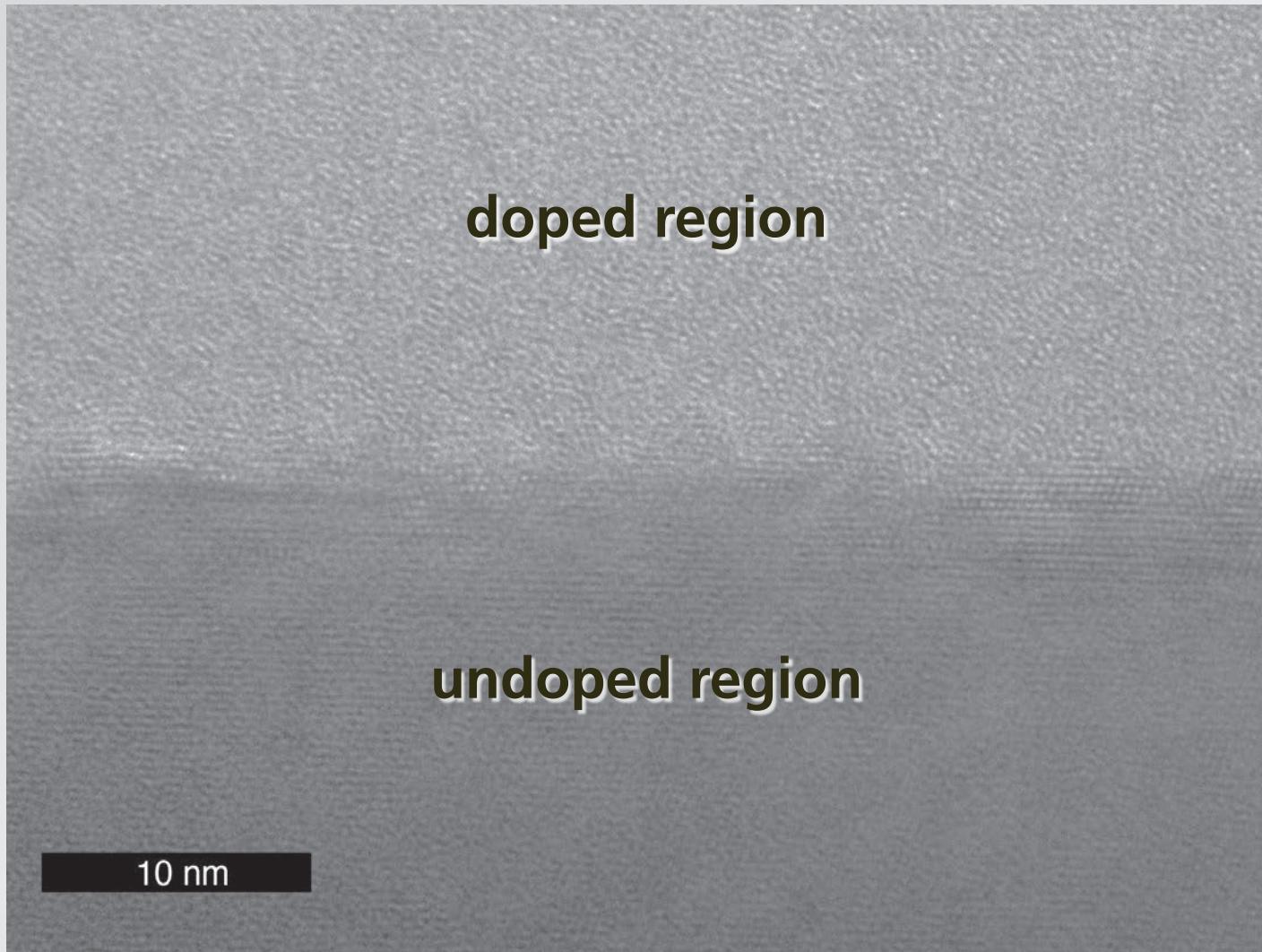
decouple ablation from melting



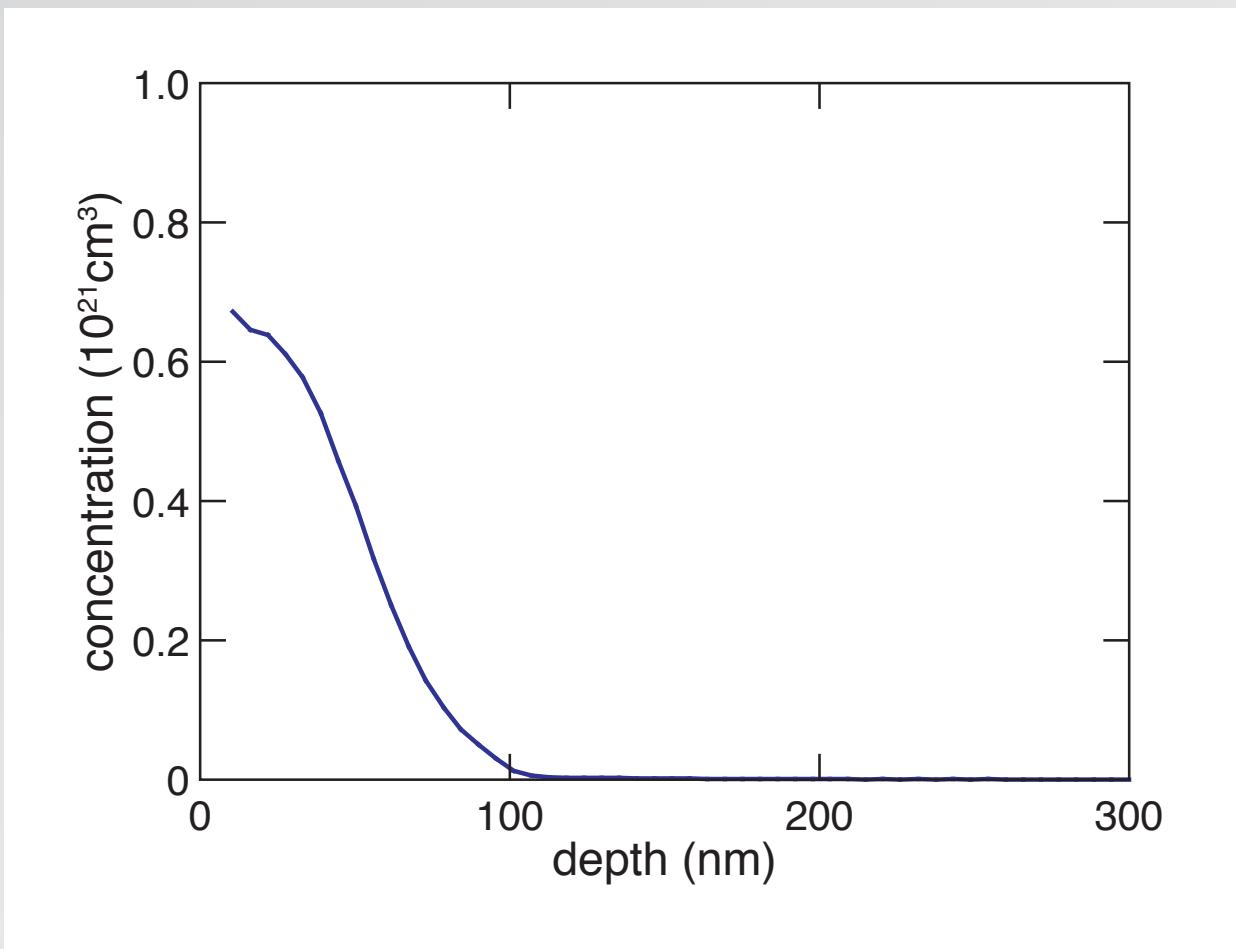
decouple ablation from melting



decouple ablation from melting



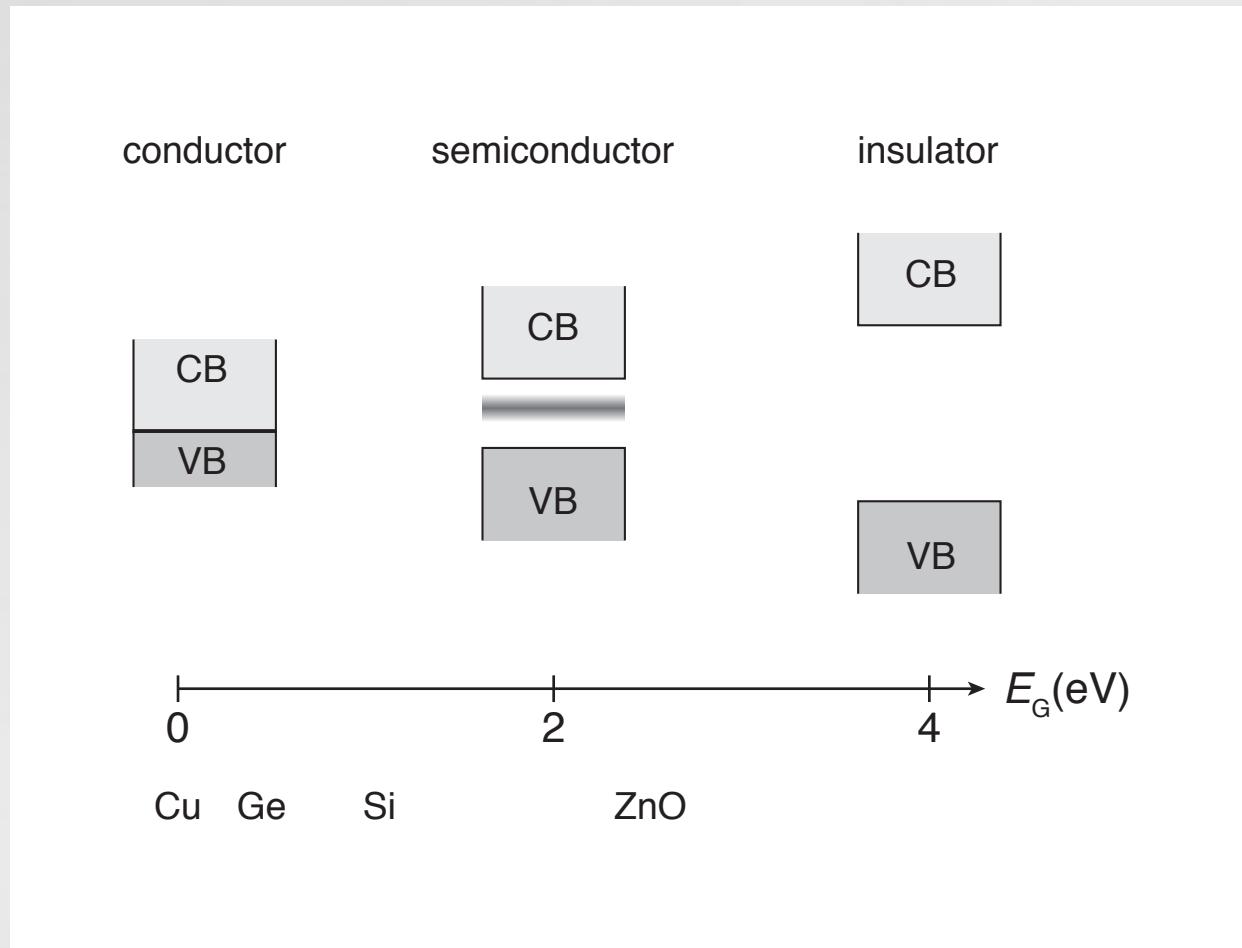
secondary ion mass spectrometry



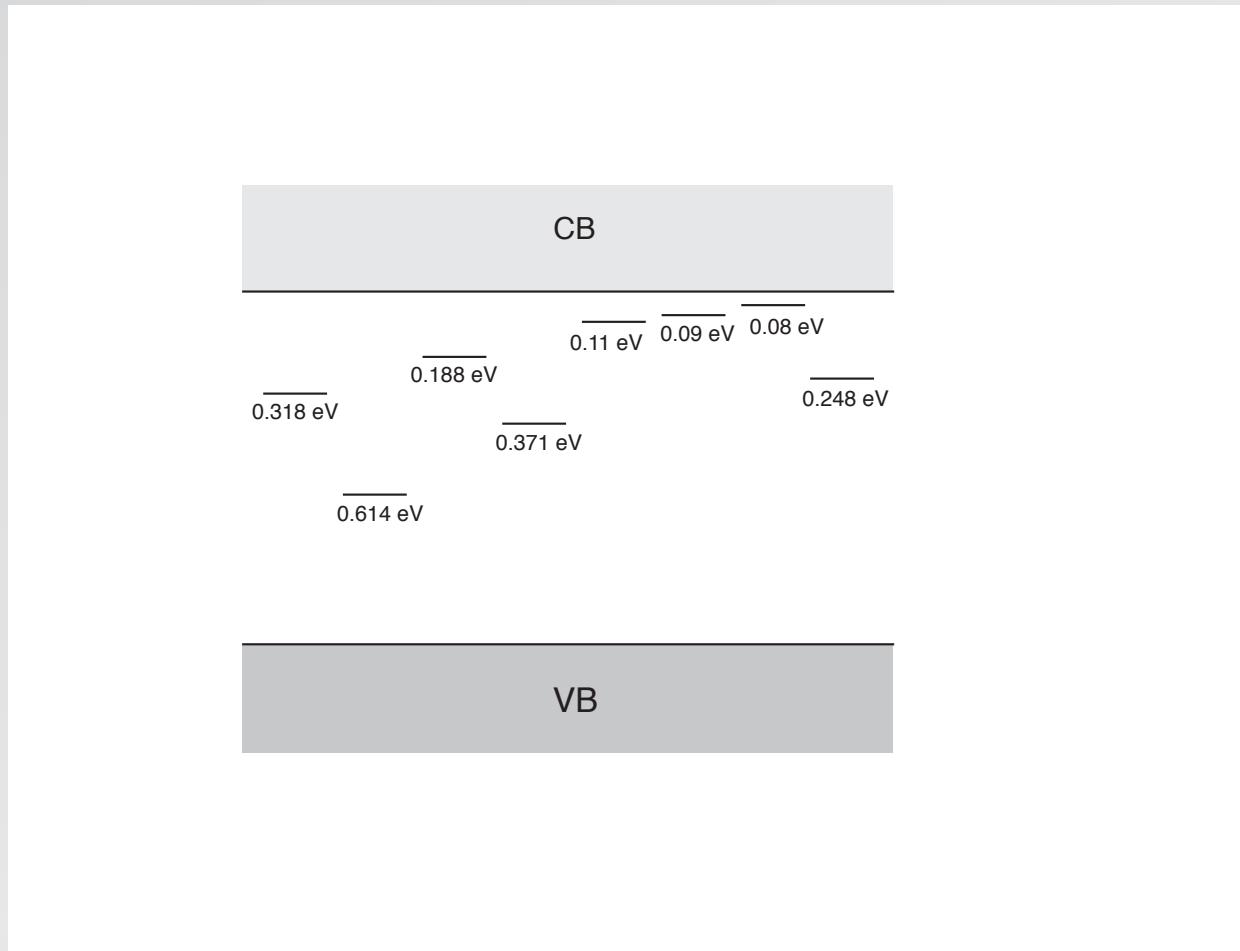
Things to keep in mind

- 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



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

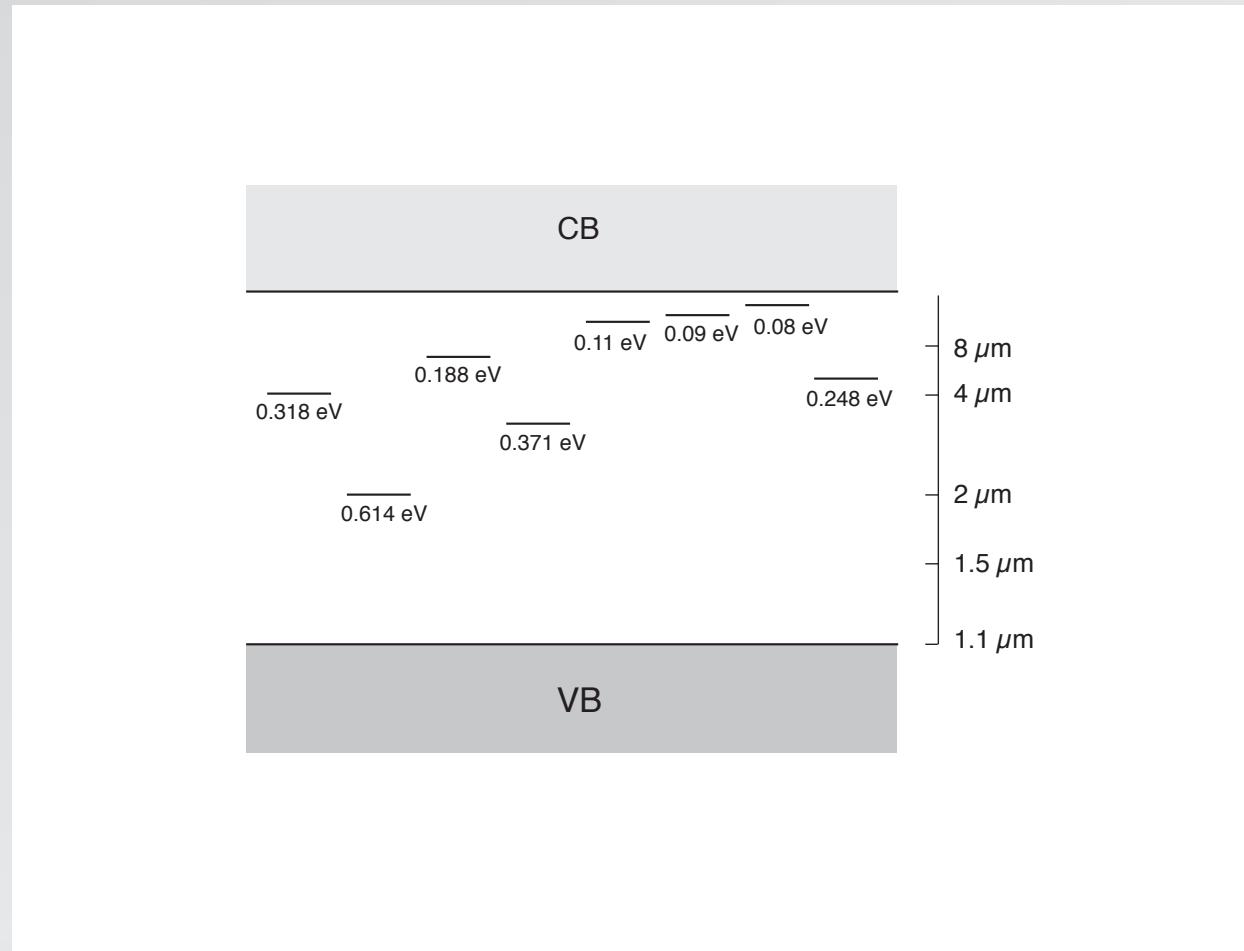


Janzén *et al.*, Phys. Rev. B 29, 1907 (1984)

1 properties

2 intermediate band

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

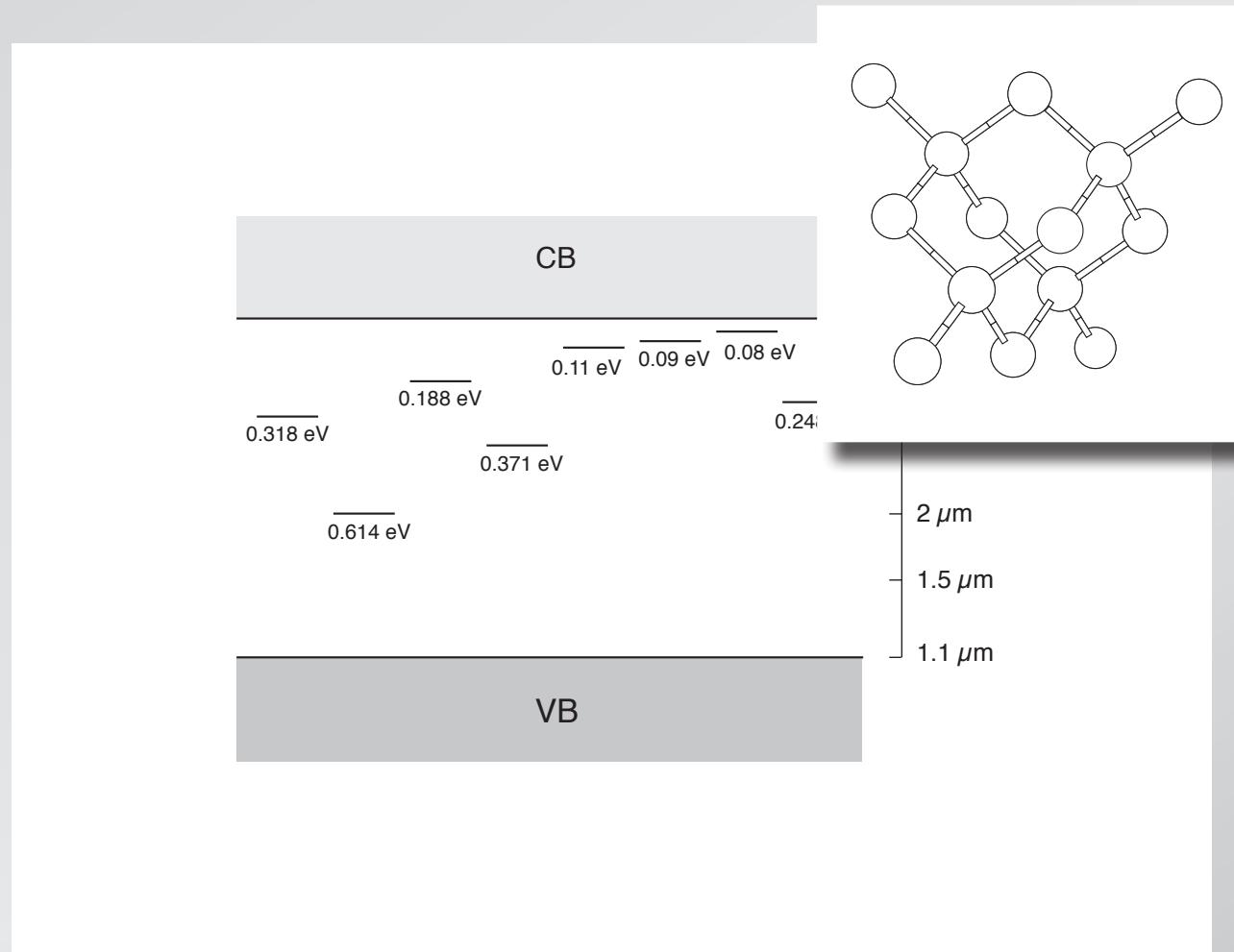


Janzén *et al.*, Phys. Rev. B 29, 1907 (1984)

1 properties

2 intermediate band

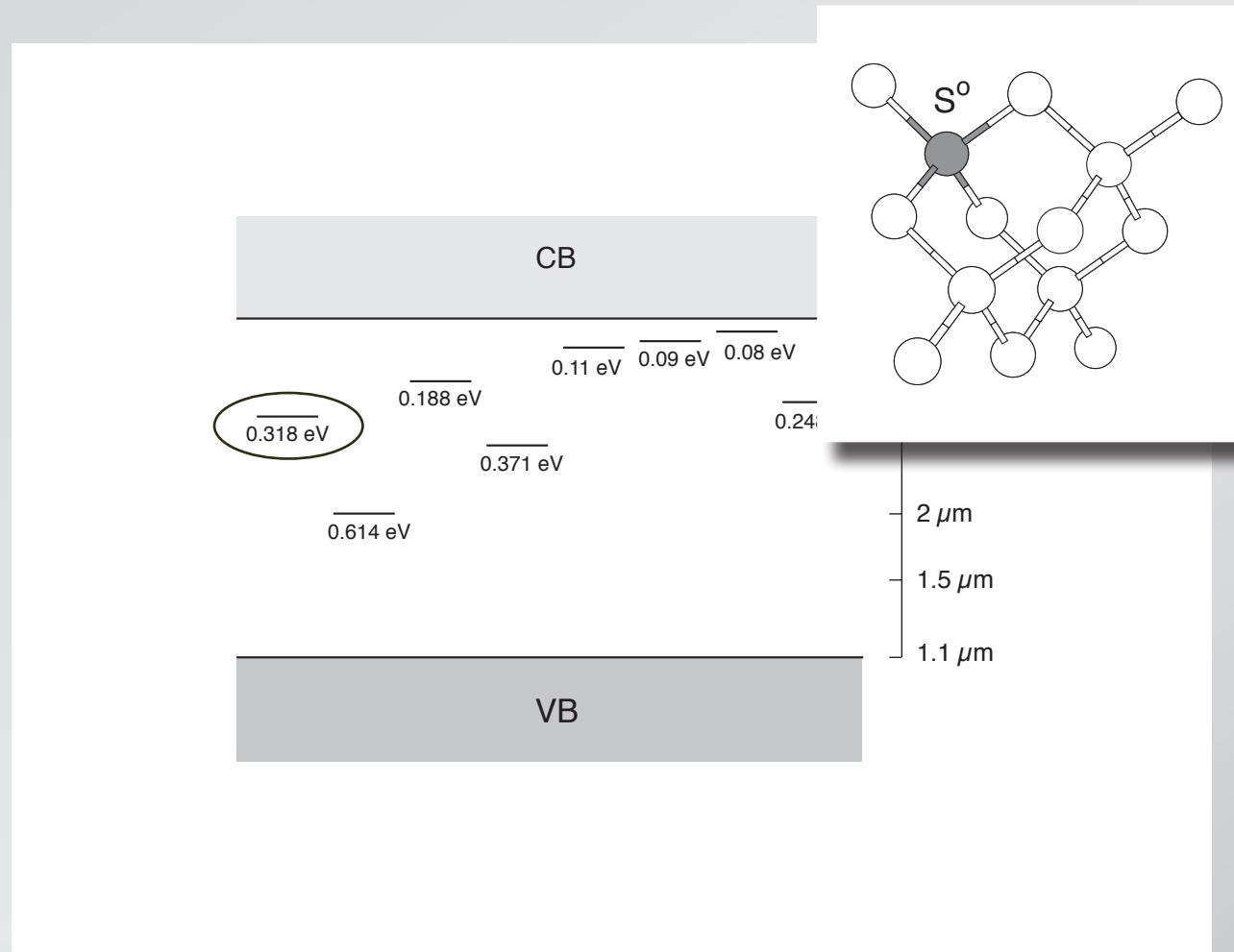
1 part in 10^6 sulfur introduces donor states in gap



Janzén *et al.*, Phys. Rev. B 29, 1907 (1984)

1 intermediate band

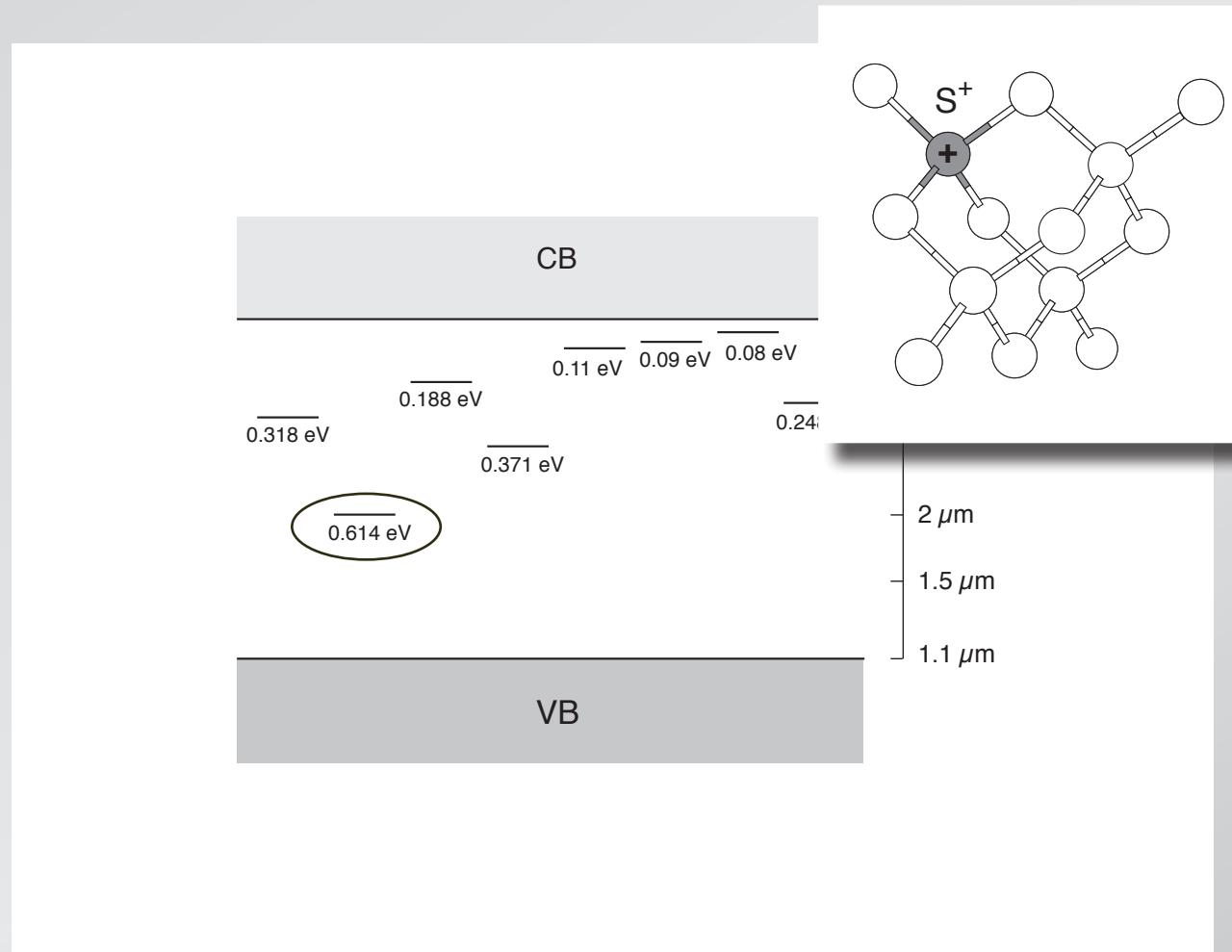
1 part in 10^6 sulfur introduces donor states in gap



Janzén et al., Phys. Rev. B 29, 1907 (1984)

1 intermediate band

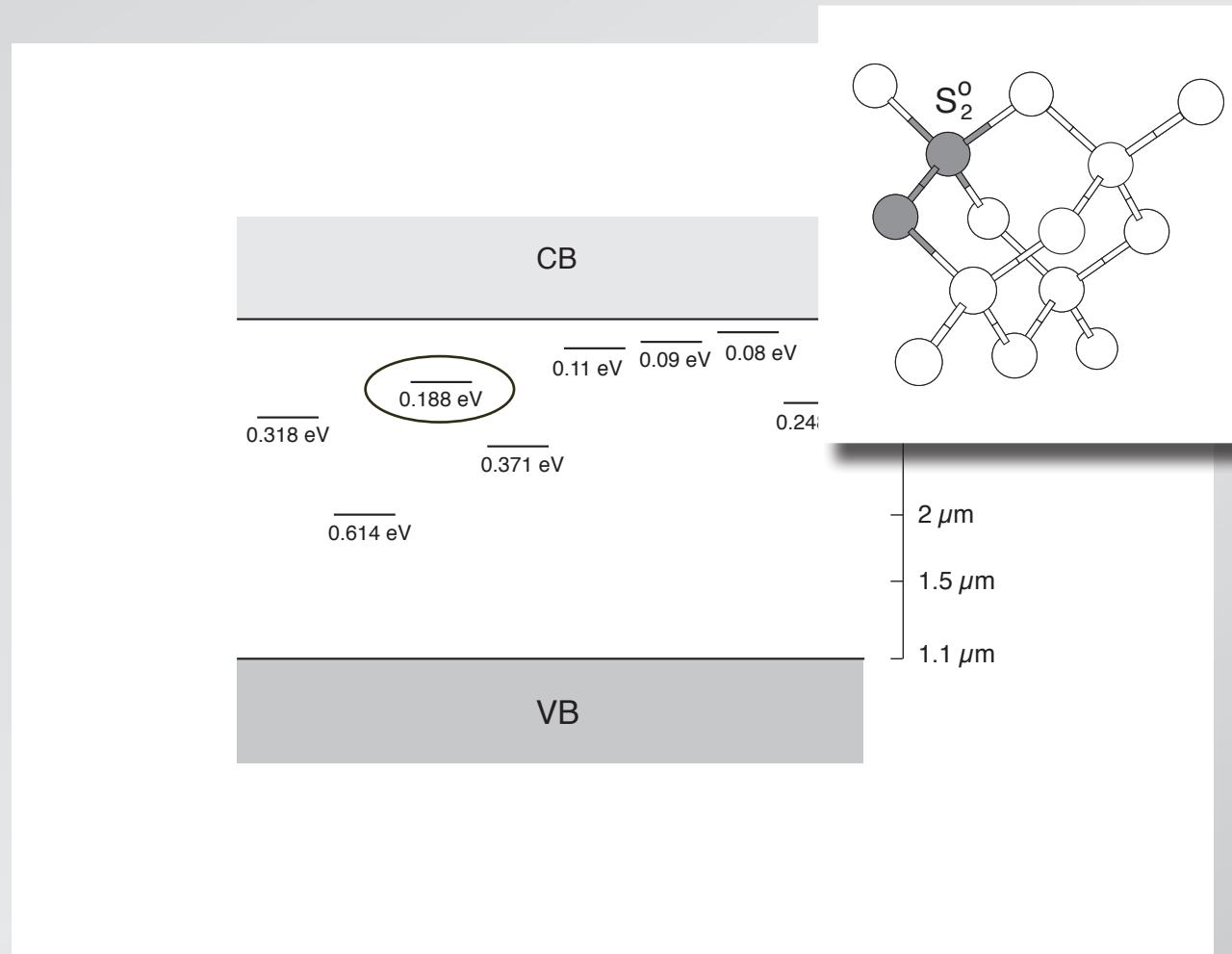
1 part in 10^6 sulfur introduces donor states in gap



Janzén et al., Phys. Rev. B 29, 1907 (1984)

1 intermediate band

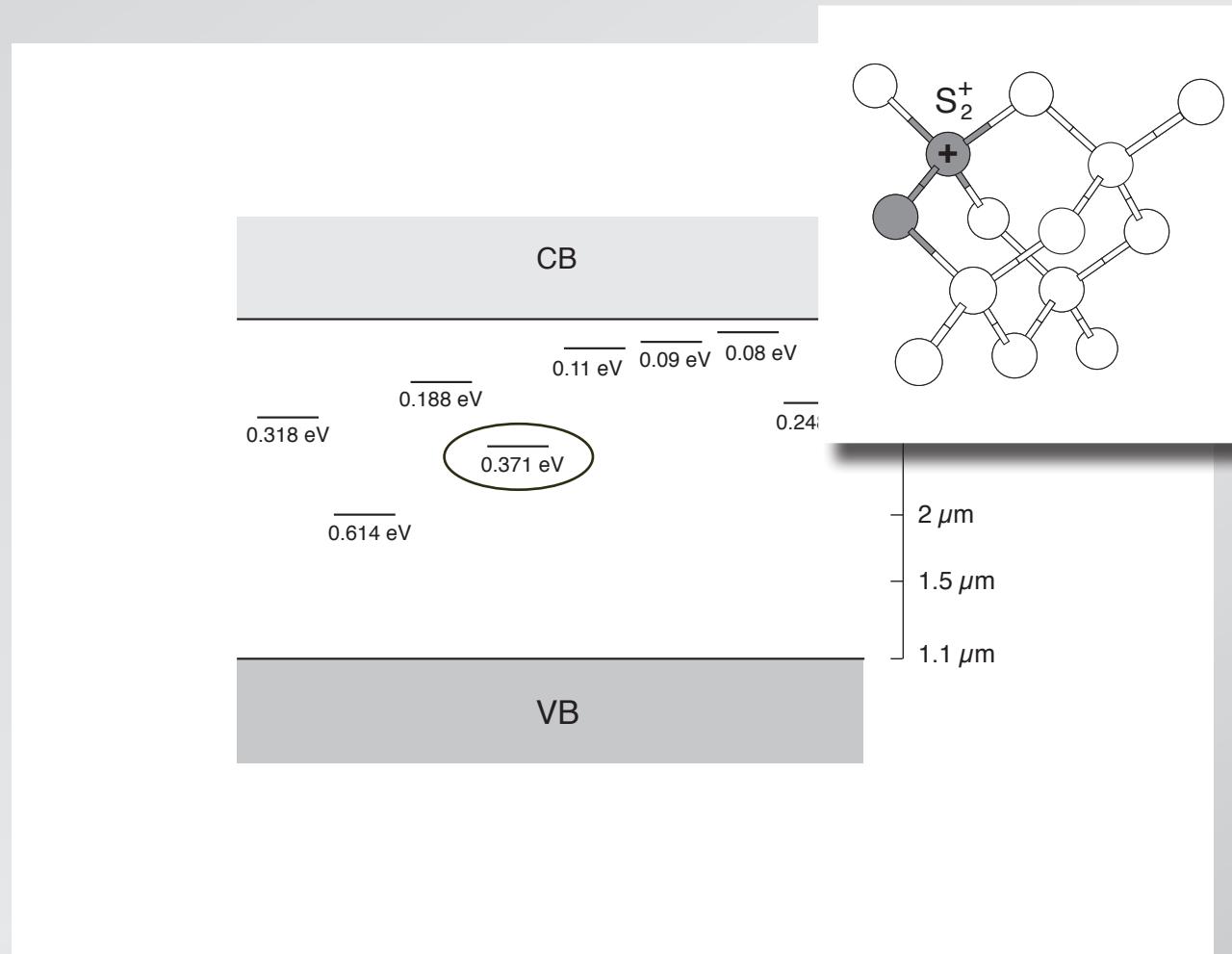
1 part in 10^6 sulfur introduces donor states in gap



Janzén *et al.*, Phys. Rev. B 29, 1907 (1984)

1 intermediate band

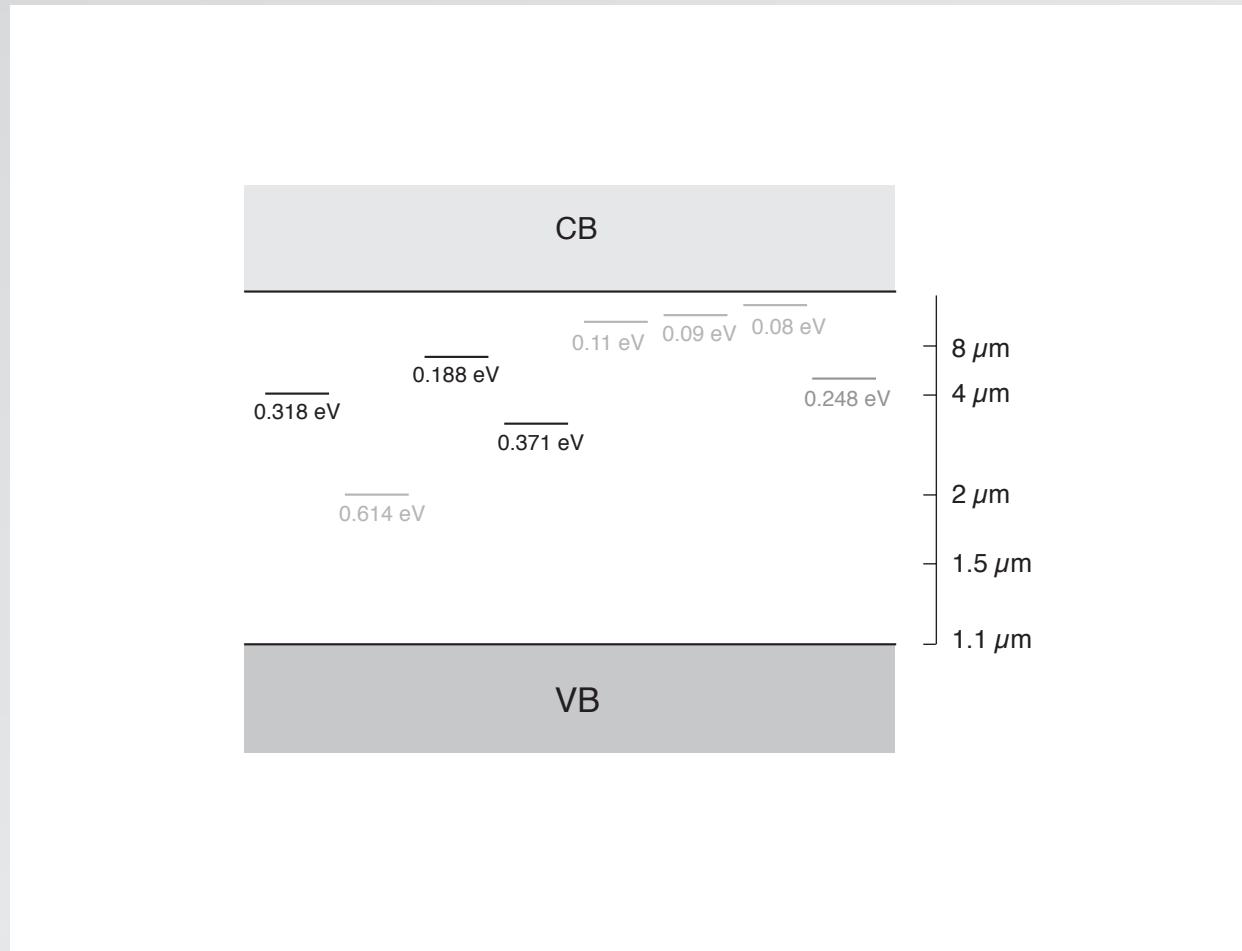
1 part in 10^6 sulfur introduces donor states in gap



Janzén et al., Phys. Rev. B 29, 1907 (1984)

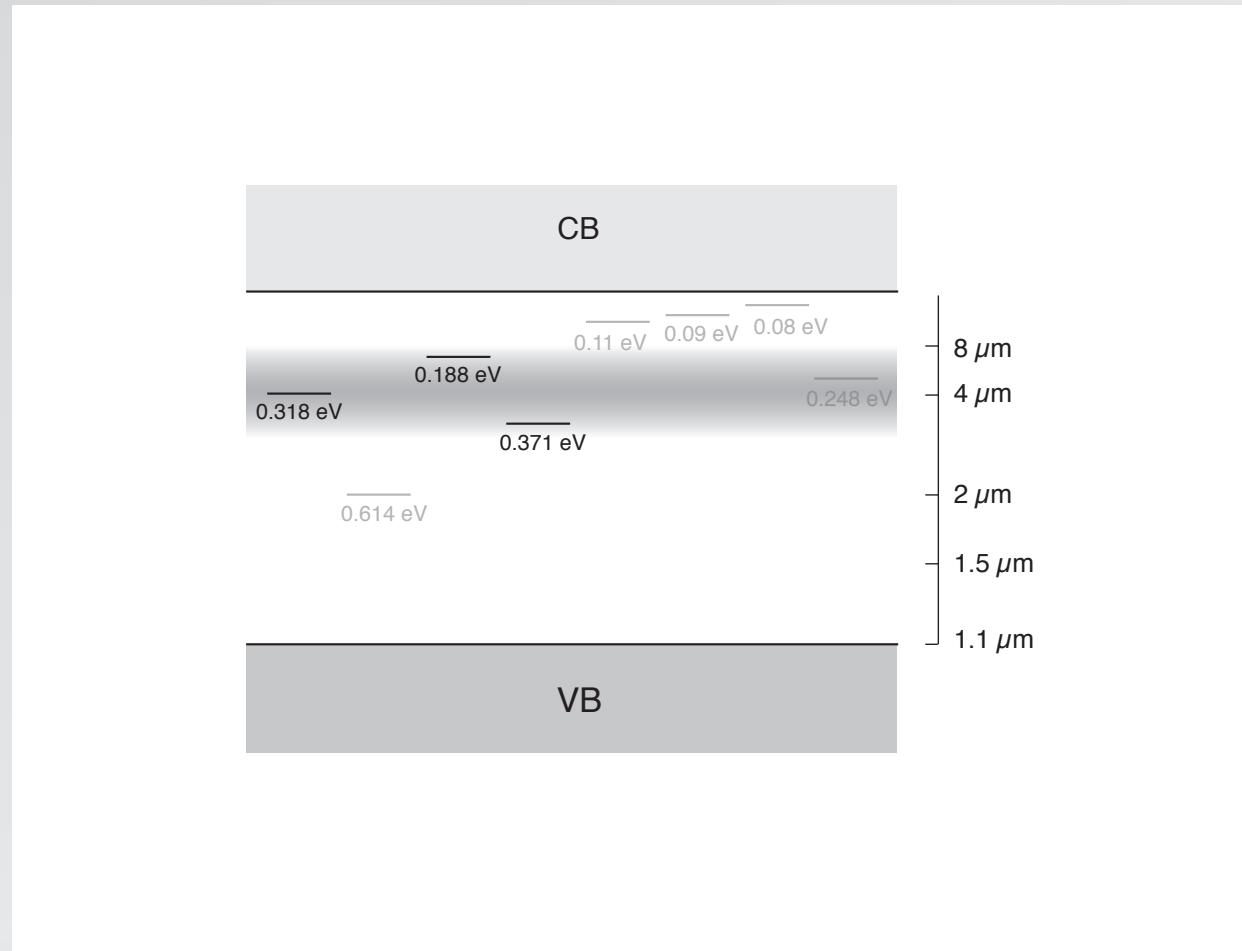
1 intermediate band

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



Janzén *et al.*, Phys. Rev. B 29, 1907 (1984)

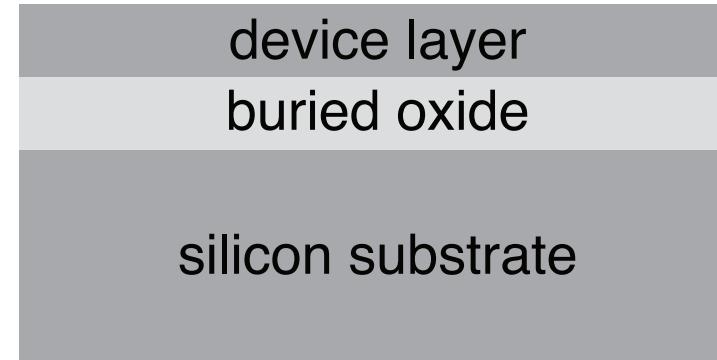
at high concentration states broaden into band



1 properties

2 intermediate band

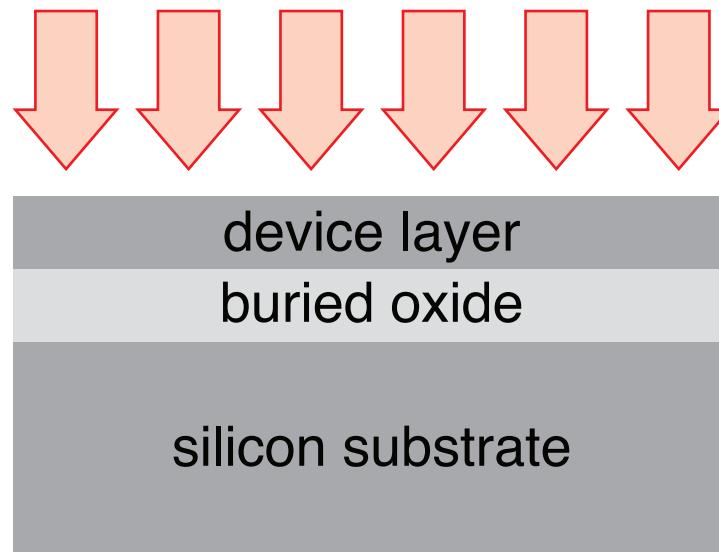
isolate surface layer for Hall measurements



1 properties

2 intermediate band

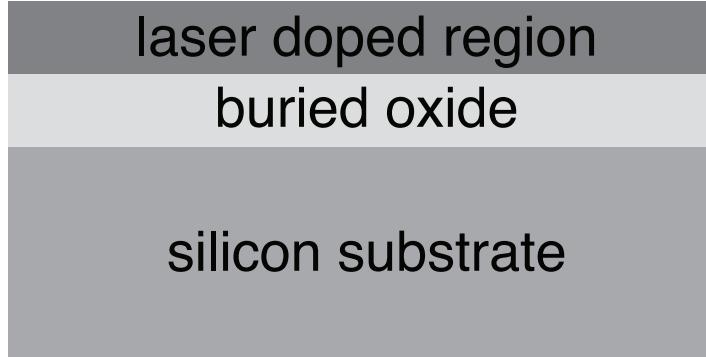
isolate surface layer for Hall measurements



1 properties

2 intermediate band

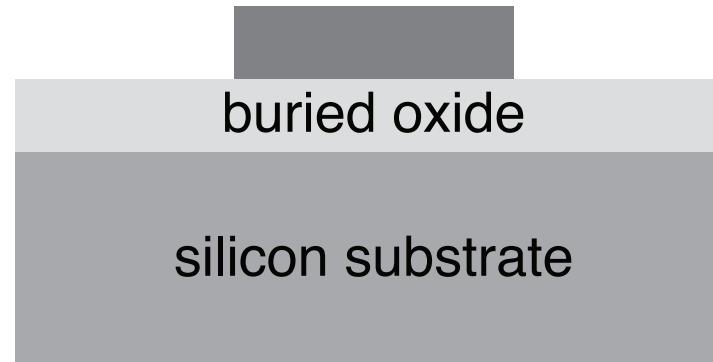
isolate surface layer for Hall measurements



1 properties

2 intermediate band

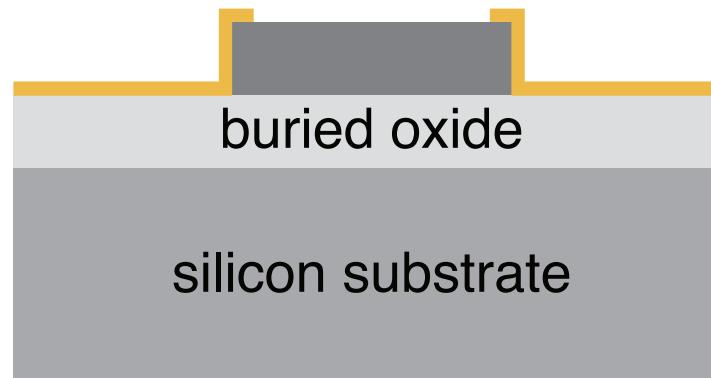
isolate surface layer for Hall measurements



1 properties

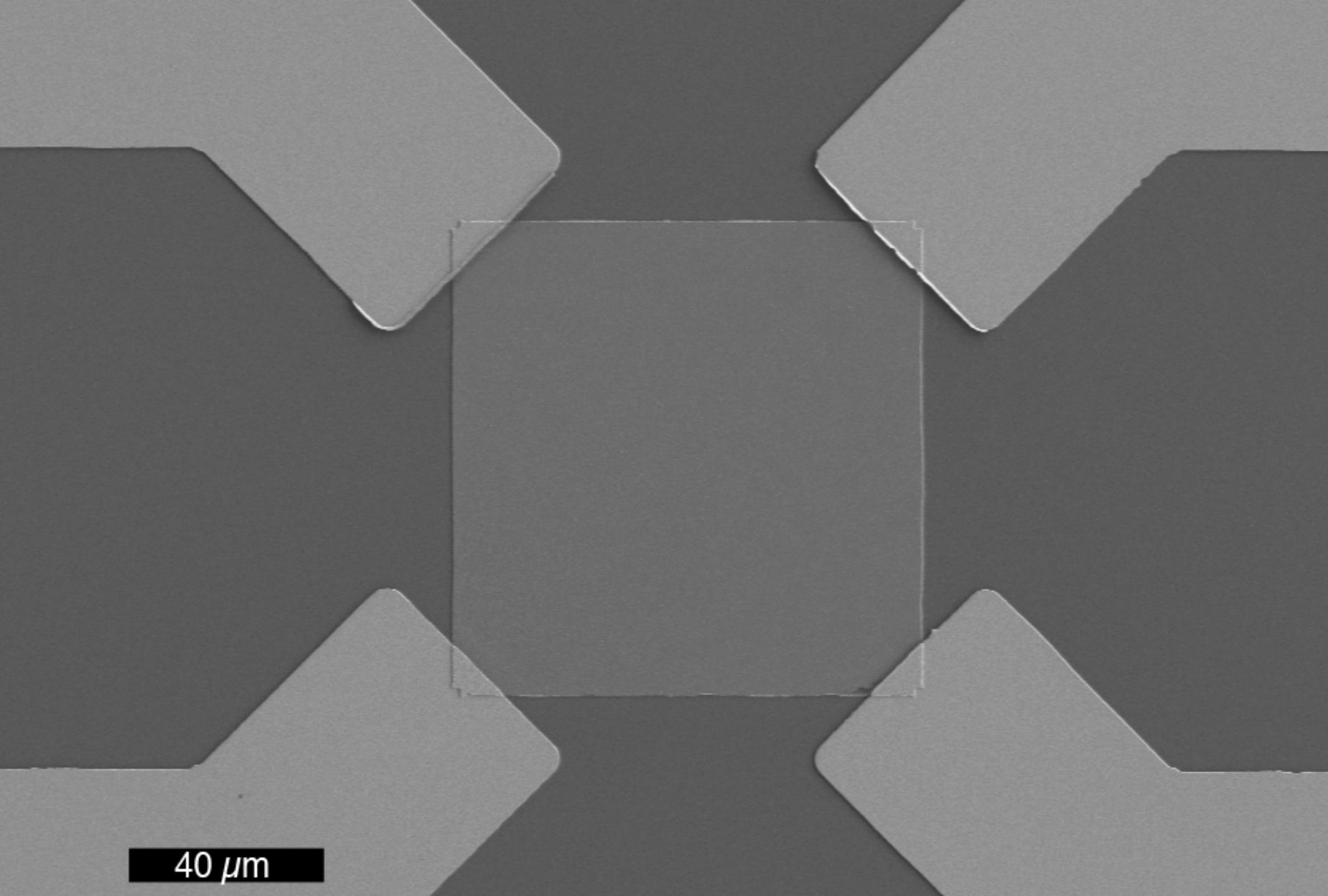
2 intermediate band

isolate surface layer for Hall measurements



1 properties

2 intermediate band

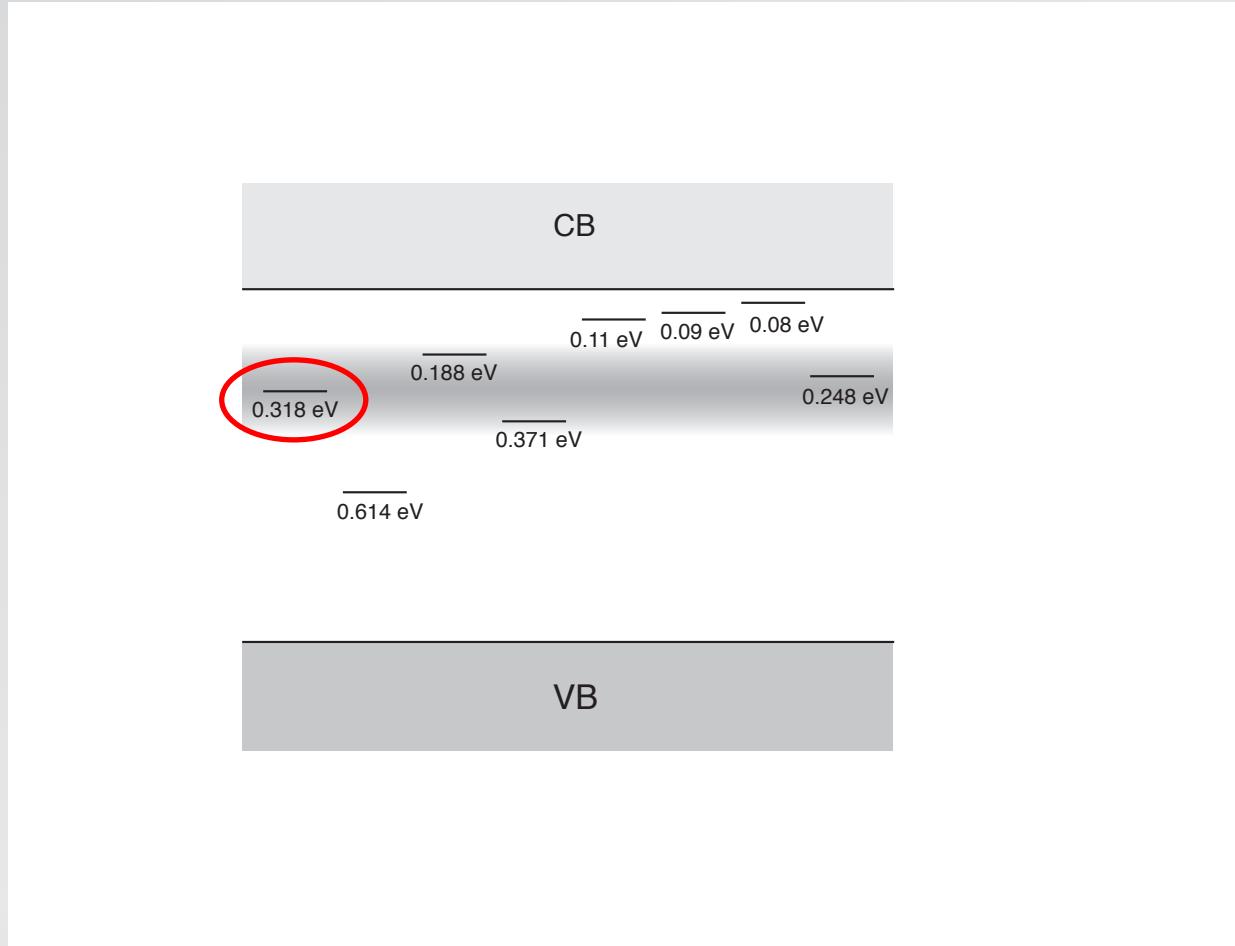


40 μm

1 properties

2 intermediate band

impurity (donor) band centered at 310 meV



1 properties

2 intermediate band

Insulator-to-Metal Transition in Selenium-Hyperdoped Silicon: Observation and Origin

Elif Ertekin,^{1,*} Mark T. Winkler,^{2,†} Daniel Recht,³ Aurore J. Said,³ Michael J. Aziz,³

Tonio Buonassisi,² and Jeffrey C. Grossman^{1,2,‡}

¹Department of Materials Science and Engineering, Massachusetts Institute of Technology, Cambridge Massachusetts 02139, USA
²Department of Mechanical Engineering, Massachusetts Institute of Technology, Cambridge Massachusetts 02139, USA
³Harvard School of Engineering and Applied Sciences, Cambridge Massachusetts 02138, USA

(Received 14 October 2011; published 11 January 2012)

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.

PACS numbers: 71.30.+h, 61.72.sd, 73.61.Cw, 78.20.Bh

Of all the experimentally measurable physical properties of materials, electronic conductivity exhibits the largest variation, spanning a factor of 10^{31} 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]. Hyperdoping (doping beyond the solubility limit) creates IMTs in semiconductors. In this Letter, we identify a new materials playground to explore defect-mediated IMTs in silicon hyperdoped with selenium to concentrations exceeding 10^{20} cm^{-3} (compared to the equilibrium solubility limit [3] of about 10^{16} cm^{-3}) and we detail the nature of the transition with both simulation and most resembles the IMT in phosphorus-doped silicon [14]. Additionally, we

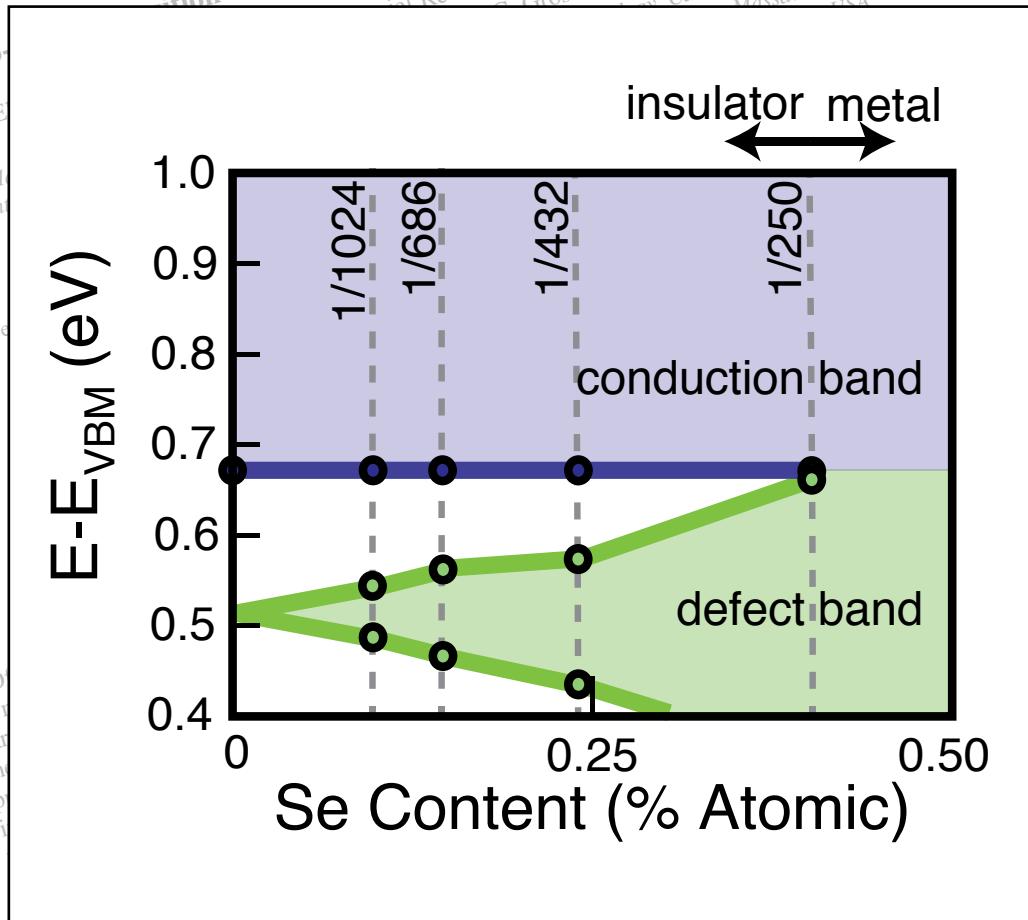
silicon appears to justify such interest. While isolated S and Se dopants are well-established deep double donors in silicon [3,14], the enhanced optical properties of hyperdoped silicon (in which these chalcogenic impurities are present at much higher concentrations) are not yet well understood. Further, 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 chalcogens [17].

We prepared Se-doped silicon (Se:Si) samples using ion implantation followed by nanosecond pulsed-laser melting (PLM) and rapid resolidification. The PLM process enables chalcogen doping with concentrations exceeding 1% atomic; such samples exhibit unexplained optical properties including broad, featureless absorption of photons with energy lower than the band gap of silicon [9]. Silicon substrates (boron doped, $\rho \approx 25 \Omega \text{ cm}$) were ion implanted with Se to nominal doses of 3×10^{15} and $1 \times 10^{16} \text{ cm}^2$ using an ion beam energy of 176 keV. The implanted samples were exposed to four laser pulses (fluences of 1.7, 1.7, 1.7 and 1.8 J cm^{-2}). This fluence regimen results in a slightly shallower dopant profile, and higher Se concentration, than reported previously [18]. The 1.8 J cm^{-2} laser pulse, which is electrically isolated from the previous pulse, creates a melt layer $1 \mu\text{m}$ thick as measured

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DFT calculations

PRL 108, 026401 (2012)
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13 JANUARY 2012
Formation in Selenium-Hyperdoped Silicon: Observation and Origin
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Ertekin et al., Phys. Rev. Lett. 108, 026401 (2012)

1 properties

2 intermediate band

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

JOURNAL OF APPLIED PHYSICS 113, 213501 (2013)

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I. INTRODUCTION

Silicon hyperdoped with chalcogens can be synthesized by pulsed laser irradiation in a sulfur-bearing atmosphere,^{1,2} ion implantation followed by pulsed laser melting,^{3,4} or pulsed laser mixing.⁵ This material has attracted interest because of its sub band gap absorption and has been studied as a candidate for infrared (IR) photodetectors^{6–8} and efficient solar cells.^{9–11} In addition, observations of carrier lifetime recovery for sufficiently high concentrations of titanium interstitials has aroused similar interest in this material.^{12,13} Hyperdoping has been shown to cause an increase in carrier density^{14–16} However,

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 $^{32}\text{S}^-$, 176 keV $^{80}\text{Se}^+$, or 245 keV $^{130}\text{Te}^+$ to doses of 1×10^{16} ions/cm². The dose of $^{32}\text{S}^-$ was varied from 3×10^{14} to 1×10^{16} ions/cm² and pre-amorphized by 85 keV Si^- to doses of 3×10^{15} ions/cm² when the $^{32}\text{S}^-$ 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 $1.7 \text{ J}/\text{cm}^2$. Time-resolved reflectivity of a fourth laser shot at $1.8 \text{ J}/\text{cm}^2$. The laser fluence was calibrated by comparing the melt duration. The laser fluence was calibrated by comparing the melt duration with numerical solutions to the one-dimensional heat equation.¹⁸ The details of the sample preparation method and the details of the sample observed by secondary ion mass spectrometry are reported elsewhere.³ For samples, the procedure is the same

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

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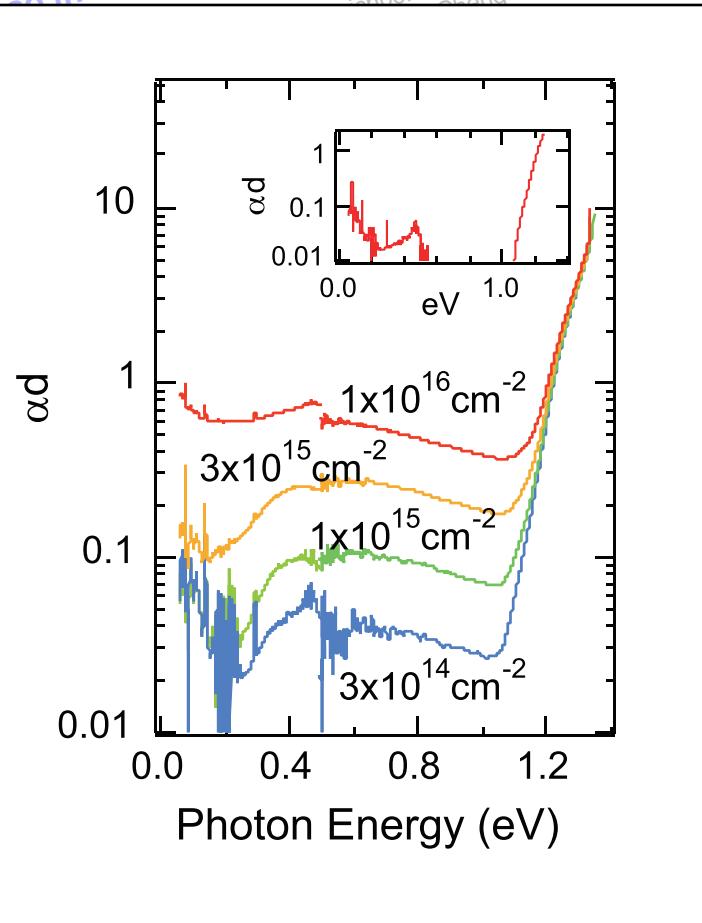
We report the emergence of a very broad infrared absorption band in silicon hyperdoped with chalcogens (S, Se, Te) at low concentrations (0.05–1.3%). The absorption band emerges at photon energies between 0.05 and 1.3 eV. Its strength increases with increasing chalcogen concentration. This is in contrast to the situation for the case of conventional ion implantation.

[http://dx.doi.org/10.1063/1.4828111]

I. INTRODUCTION

Silicon has been considered as a promising material for photovoltaic applications by pulsed laser irradiation followed by pulsed laser annealing.^{1–4} In addition, silicon implanted by pulsed laser mixing.⁵ This material has attracted attention because of its sub band gap absorption and has been studied as a candidate for infrared (IR) photodetectors^{6–8} and efficient solar cells.^{9,10} In addition, observations of carrier lifetime recovery for sufficient chalcogen hyperdoping has been shown to cause an intermediate band formation.^{11–13} However, it has aroused similar interest in the case of an intermediate bandgap formed by hyperdoping.^{14–16} However,

hyperdoped p type (001) Si wafers, resistivity was 176 keV ⁸⁰Se⁺, or 245 keV ¹³⁰Te⁺ to 1.76 × 10¹⁶ ions/cm². The dose of ³²S[−] was varied to doses of 3 × 10¹⁵ ions/cm² when the ³²S[−] dose is not greater than 1 × 10¹⁵ 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 488 nm Ar⁺ ion laser was used to measure the melt duration. The laser fluence was calibrated by comparing the melt duration with numerical solutions to the one-dimensional heat equation.¹⁸ The details of the sample preparation method and the details of the sample observed by secondary ion mass spectrometry are reported elsewhere.³ For samples, the procedure is the same



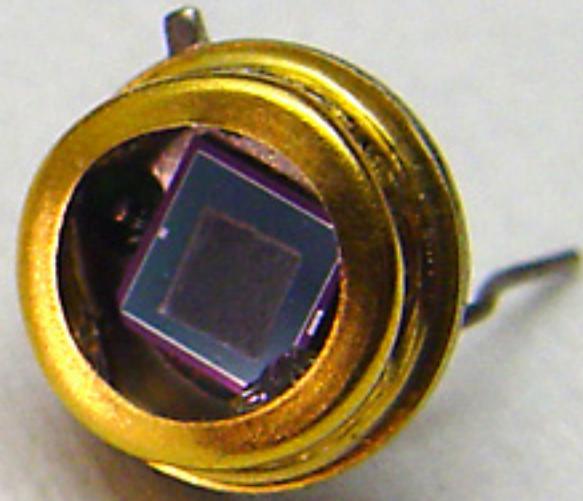
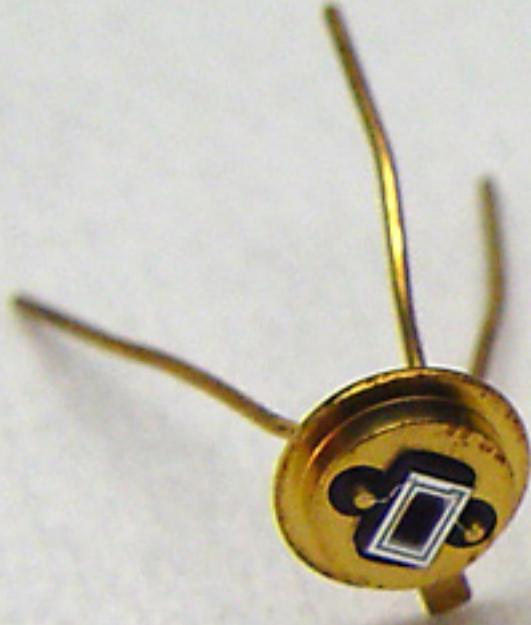
Umezu et al., J. Appl. Phys. 113, 213501 (2013)

1 properties

2 intermediate band

Things to keep in mind

- IR absorption rolls off around $8 \mu\text{m}$
- 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

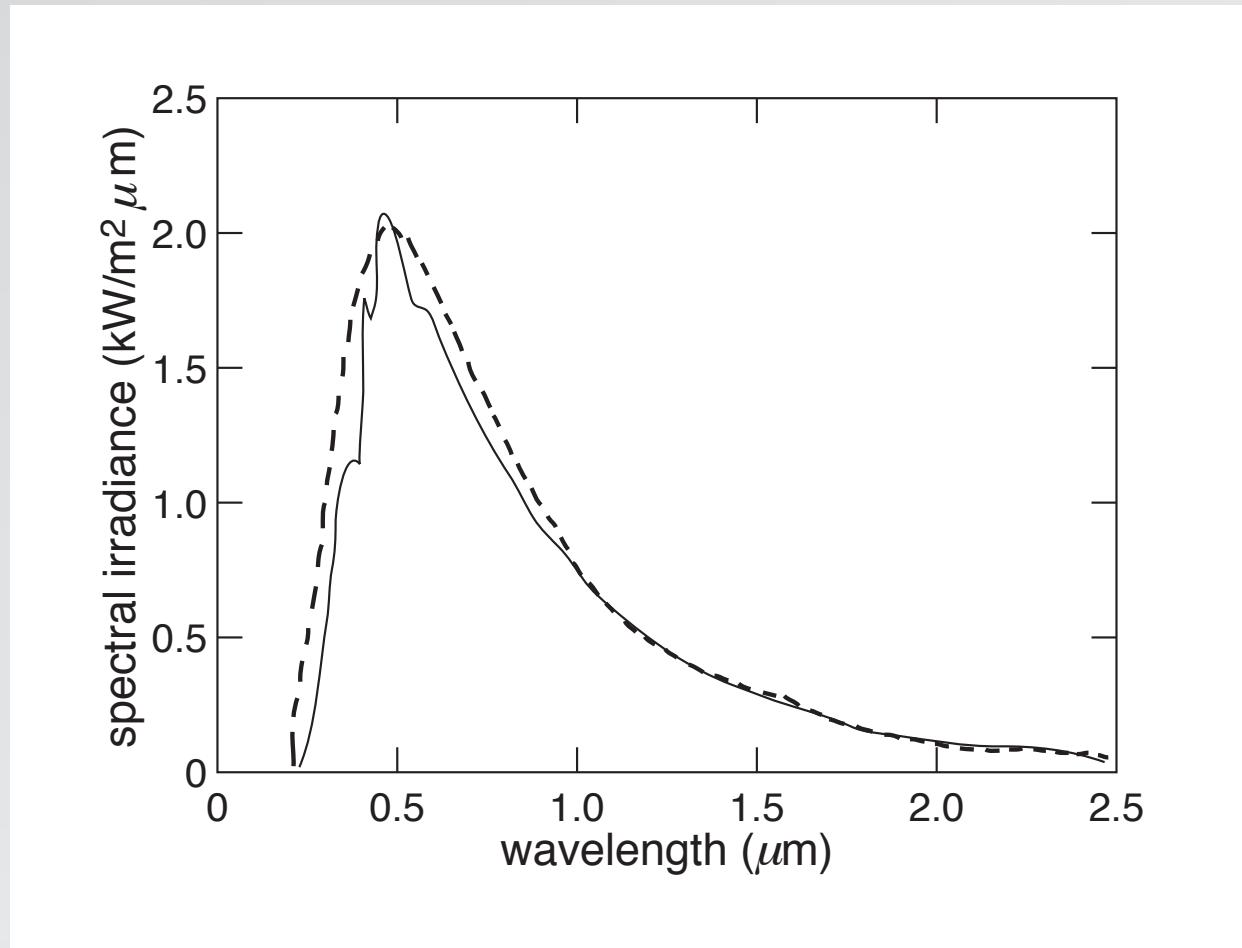


1 properties

2 intermediate band

3 devices

solar spectrum

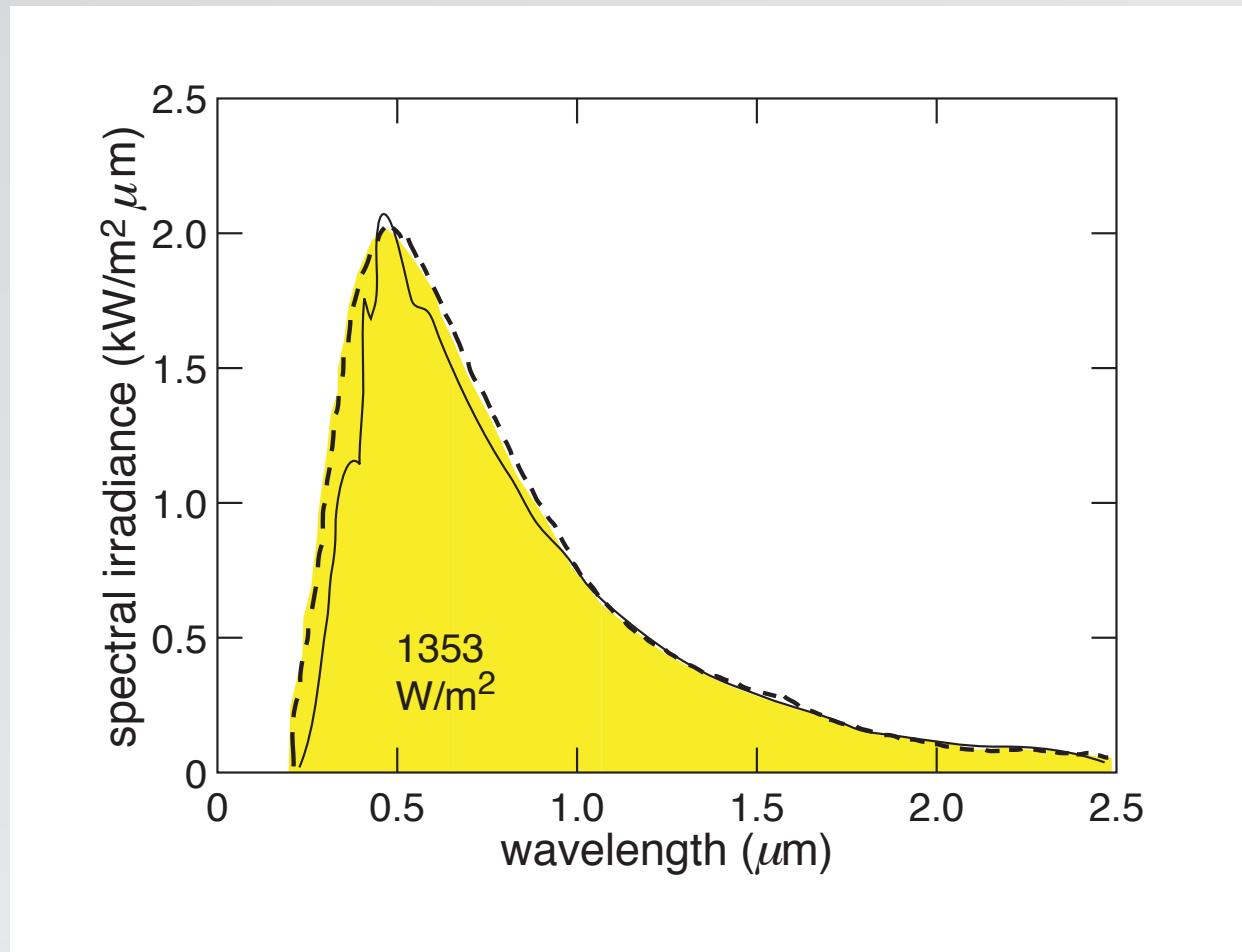


1 properties

2 intermediate band

3 devices

solar spectrum

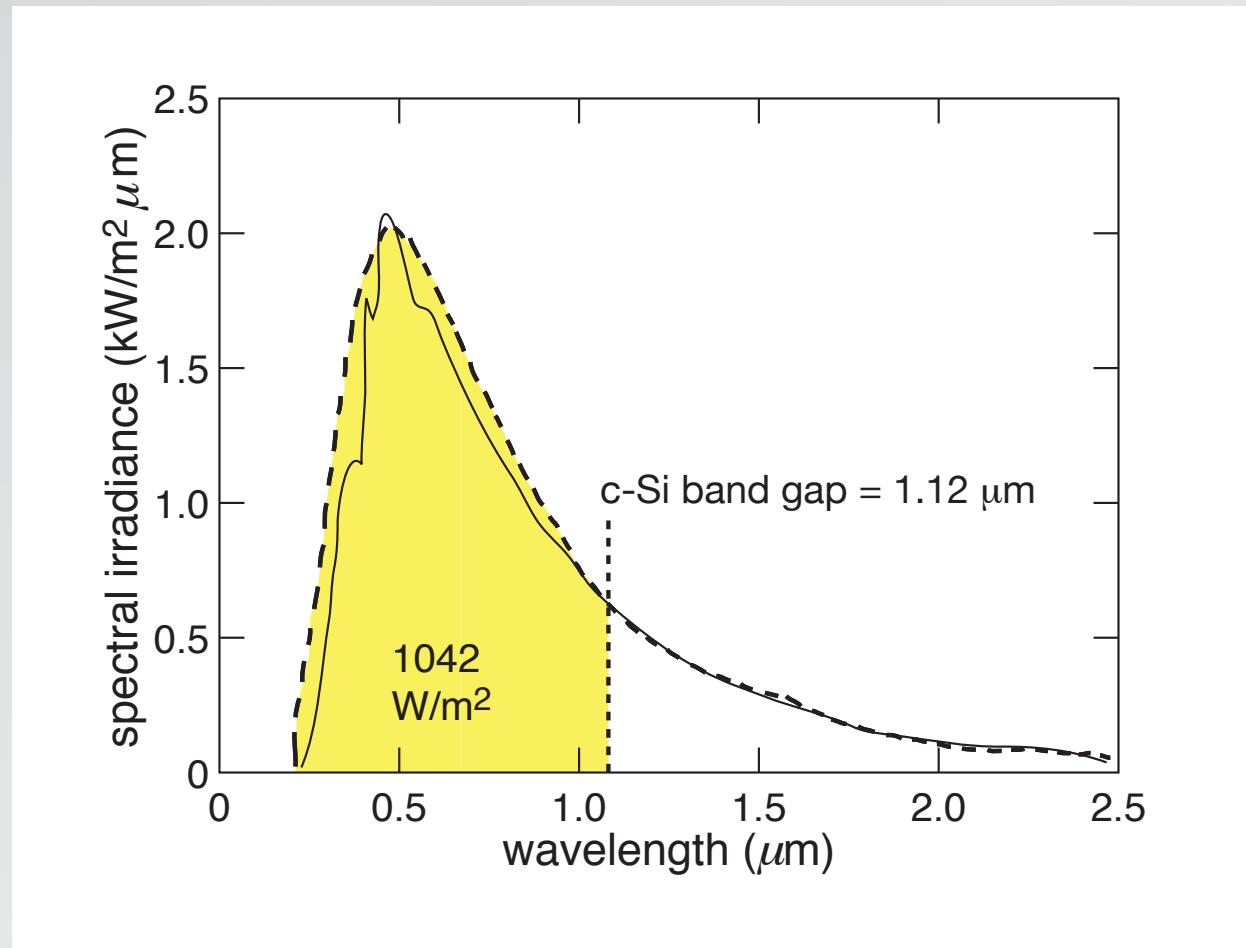


1 properties

2 intermediate band

3 devices

crystalline silicon: transparent to 23% of solar radiation

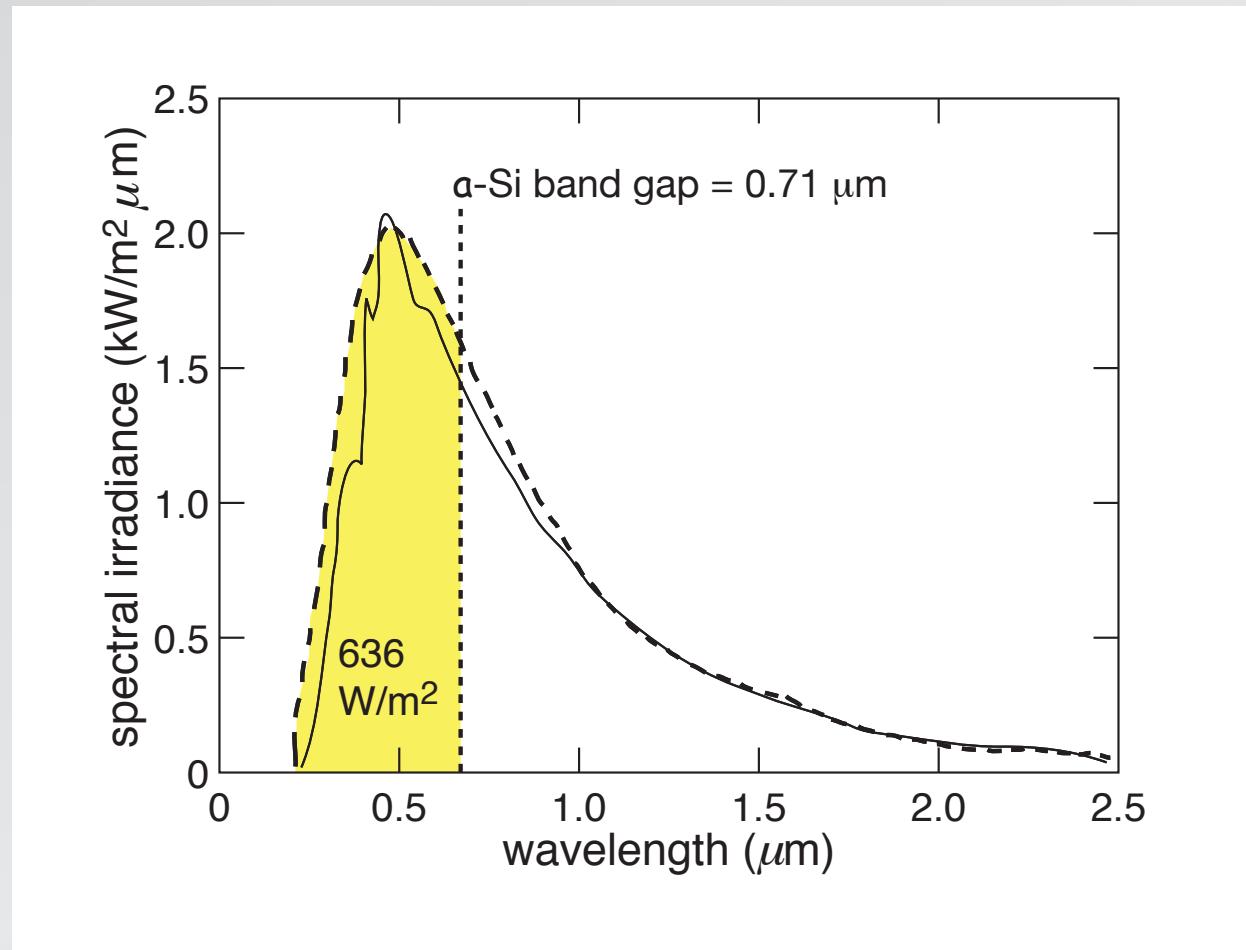


1 properties

2 intermediate band

3 devices

amorphous silicon: transparent to 53% of solar radiation



1 properties

2 intermediate band

3 devices

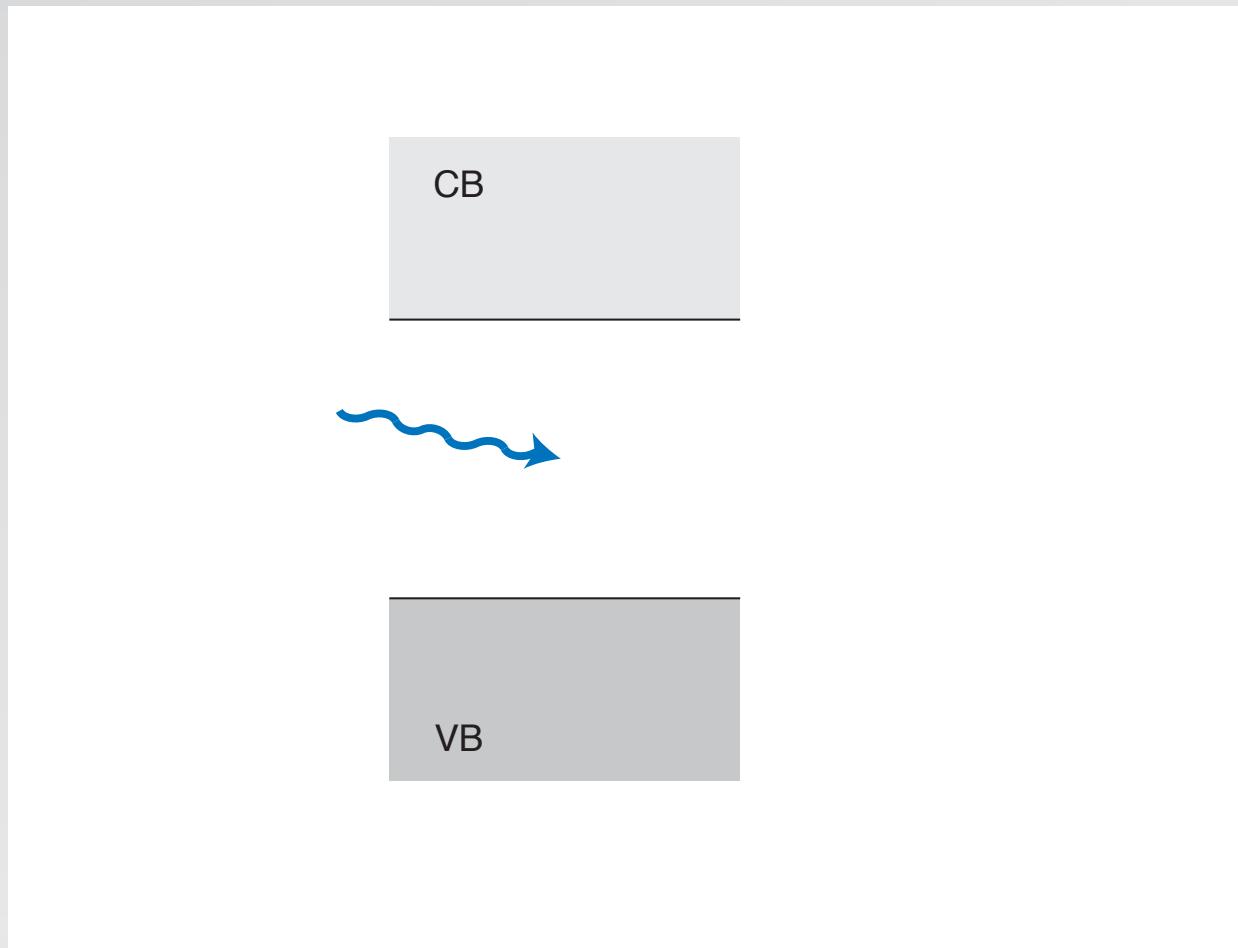


1 properties

2 intermediate band

3 devices

photon with gap energy

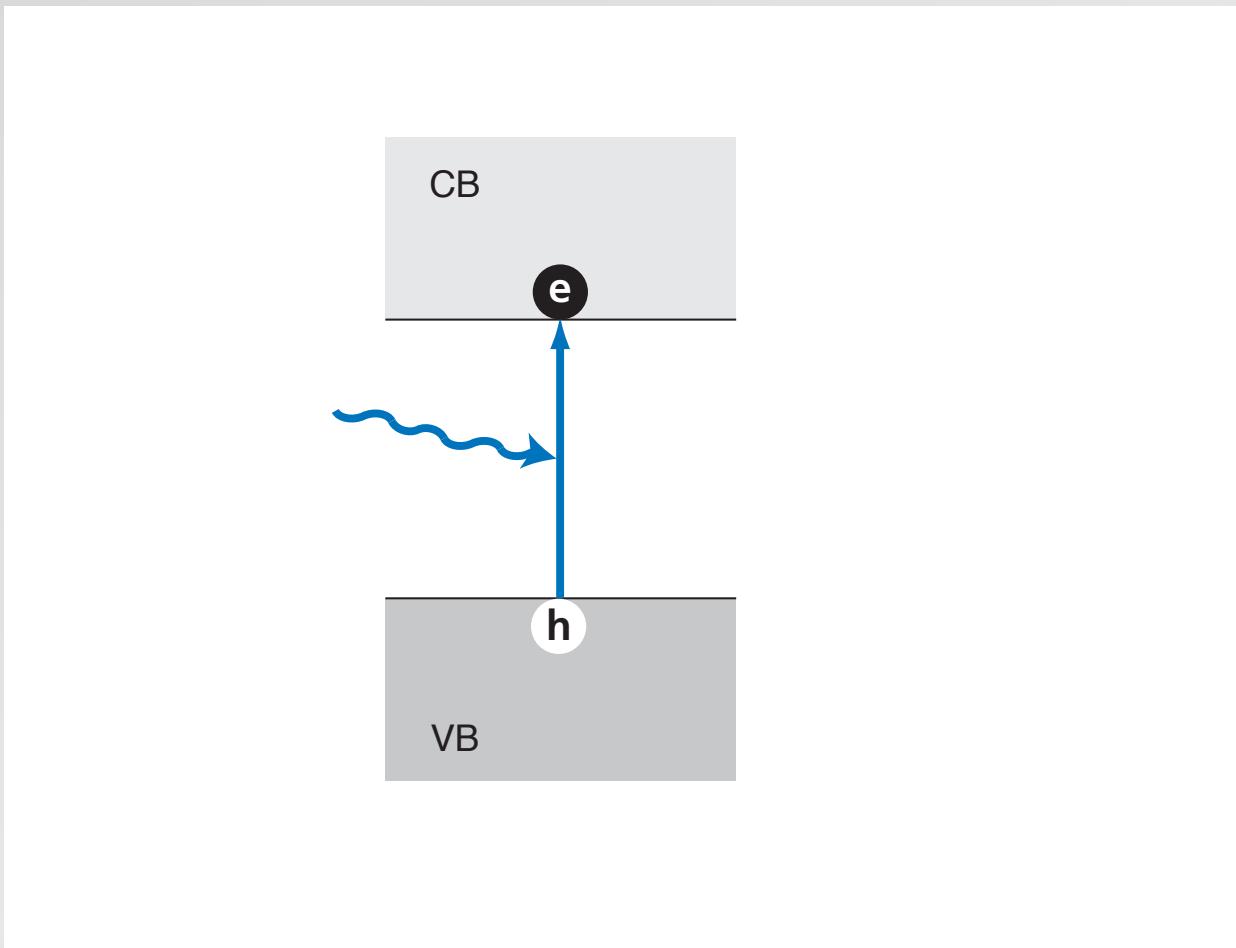


1 properties

2 intermediate band

3 devices

photon creates electron-hole pair...

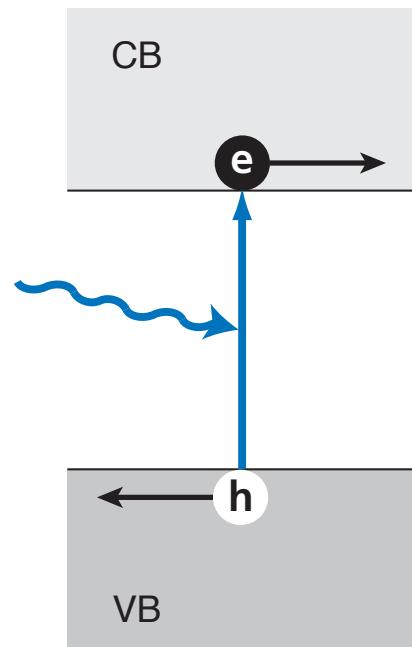


1 properties

2 intermediate band

3 devices

...whose energy can be extracted

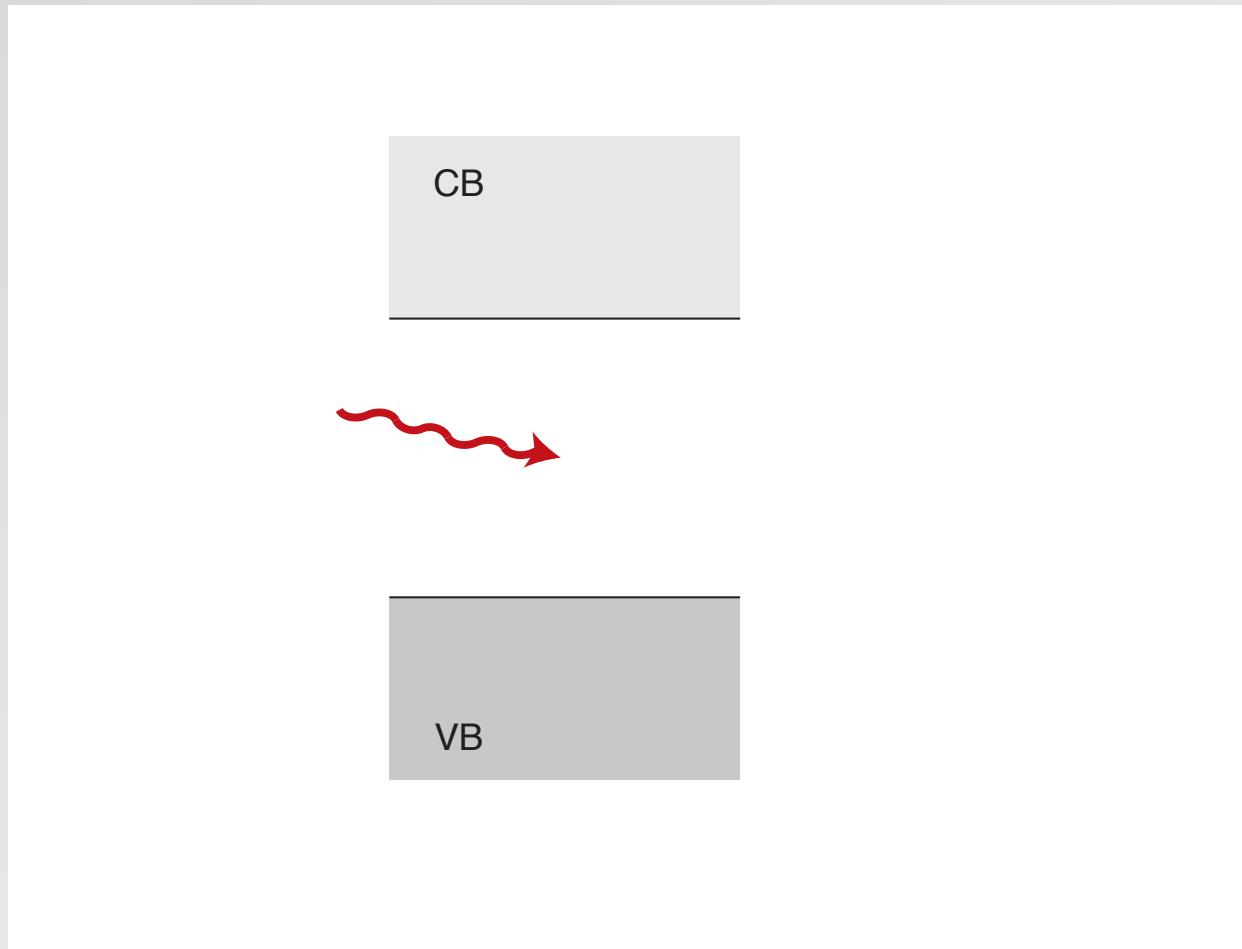


1 properties

2 intermediate band

3 devices

photons with energy smaller than gap...

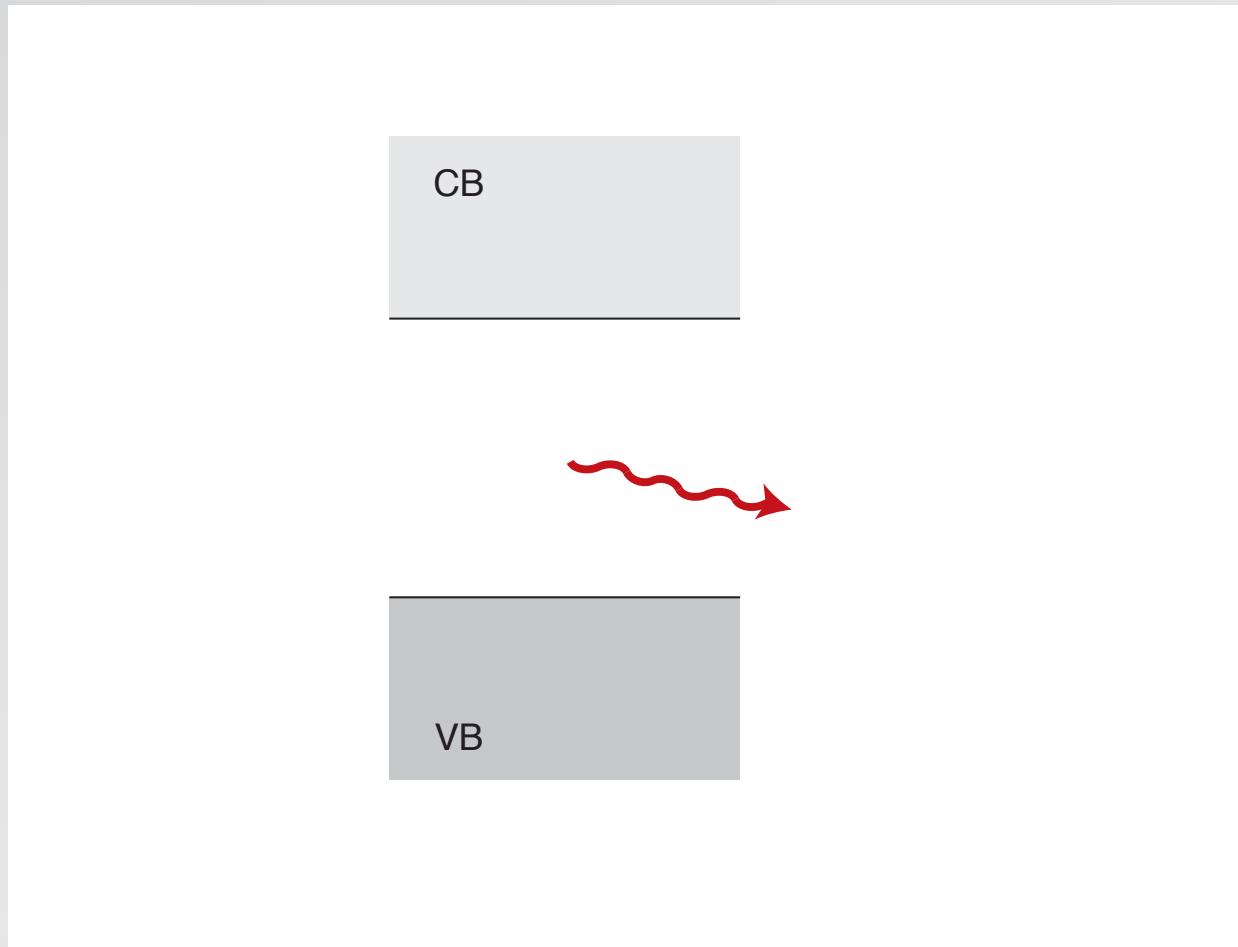


1 properties

2 intermediate band

3 devices

...do not get absorbed

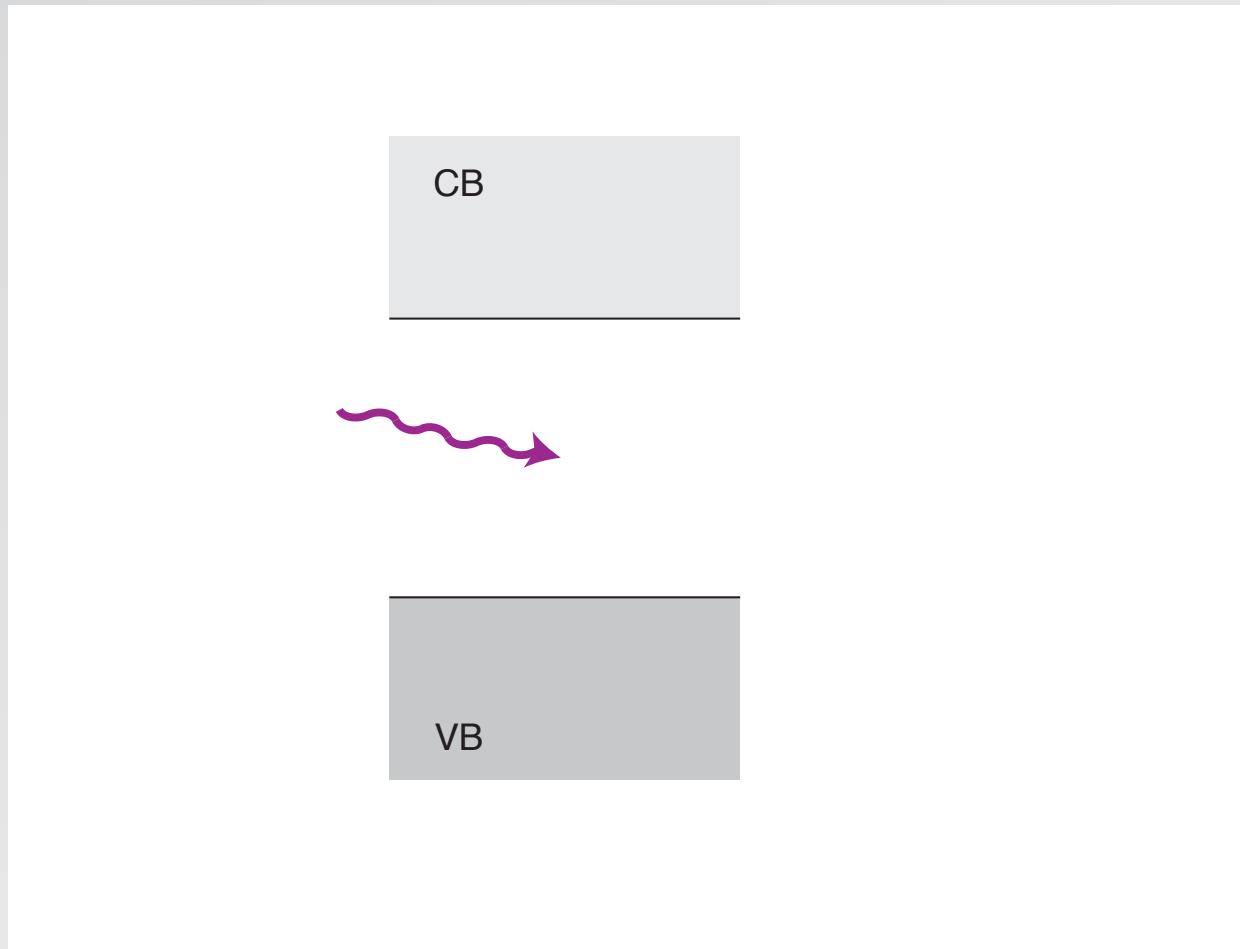


1 properties

2 intermediate band

3 devices

photons with energy larger than the gap...

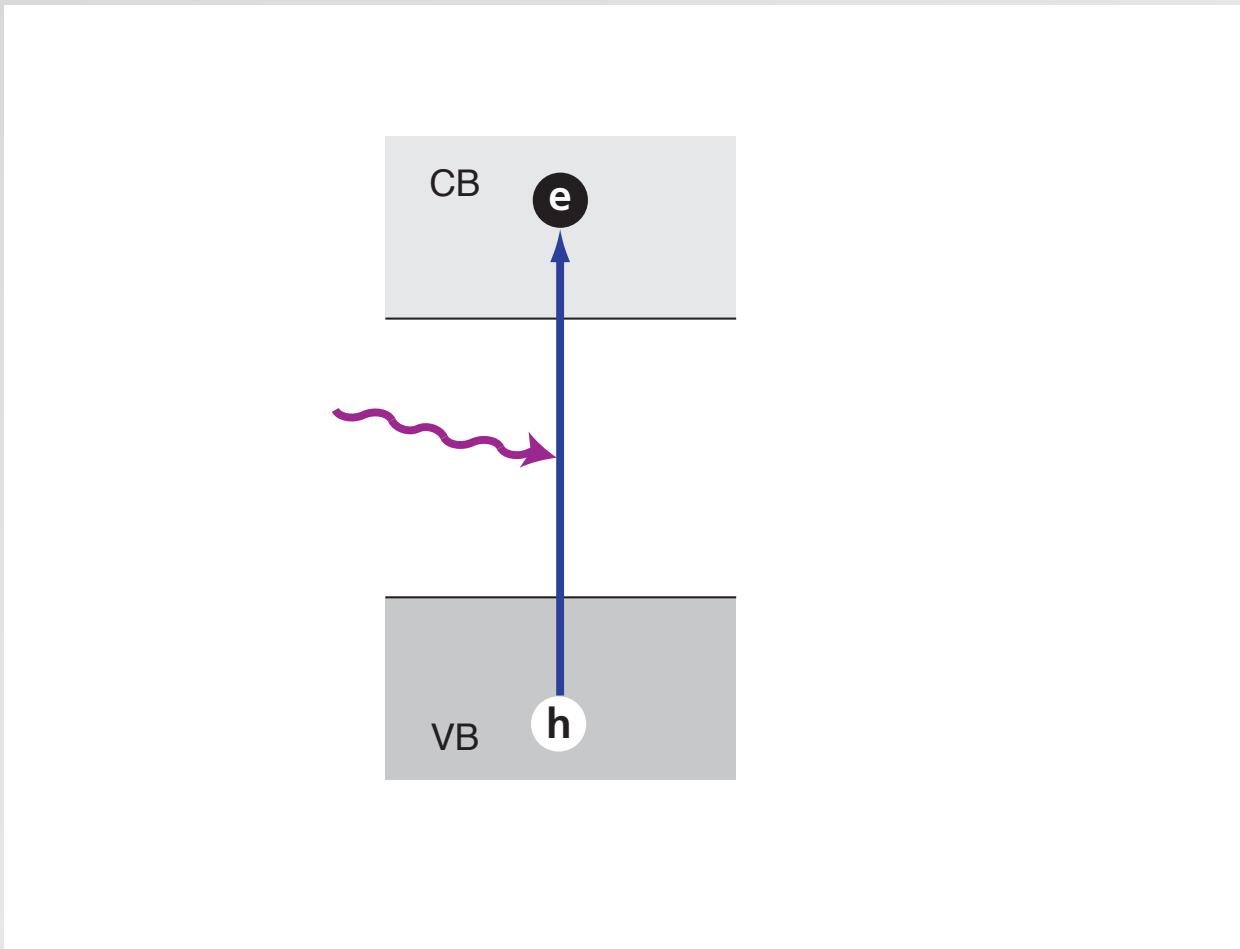


1 properties

2 intermediate band

3 devices

...create electron-hole pairs with excess energy...

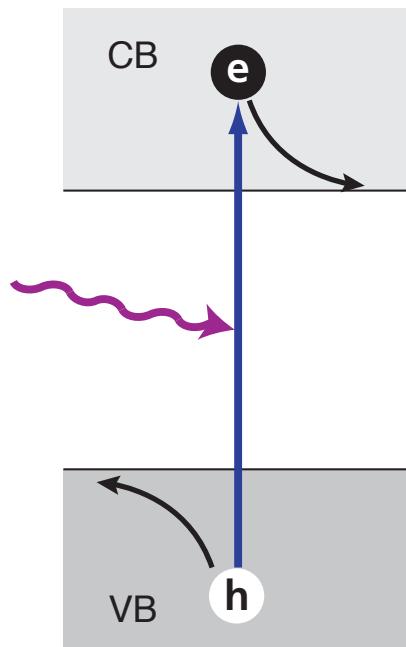


1 properties

2 intermediate band

3 devices

...which is lost rapidly

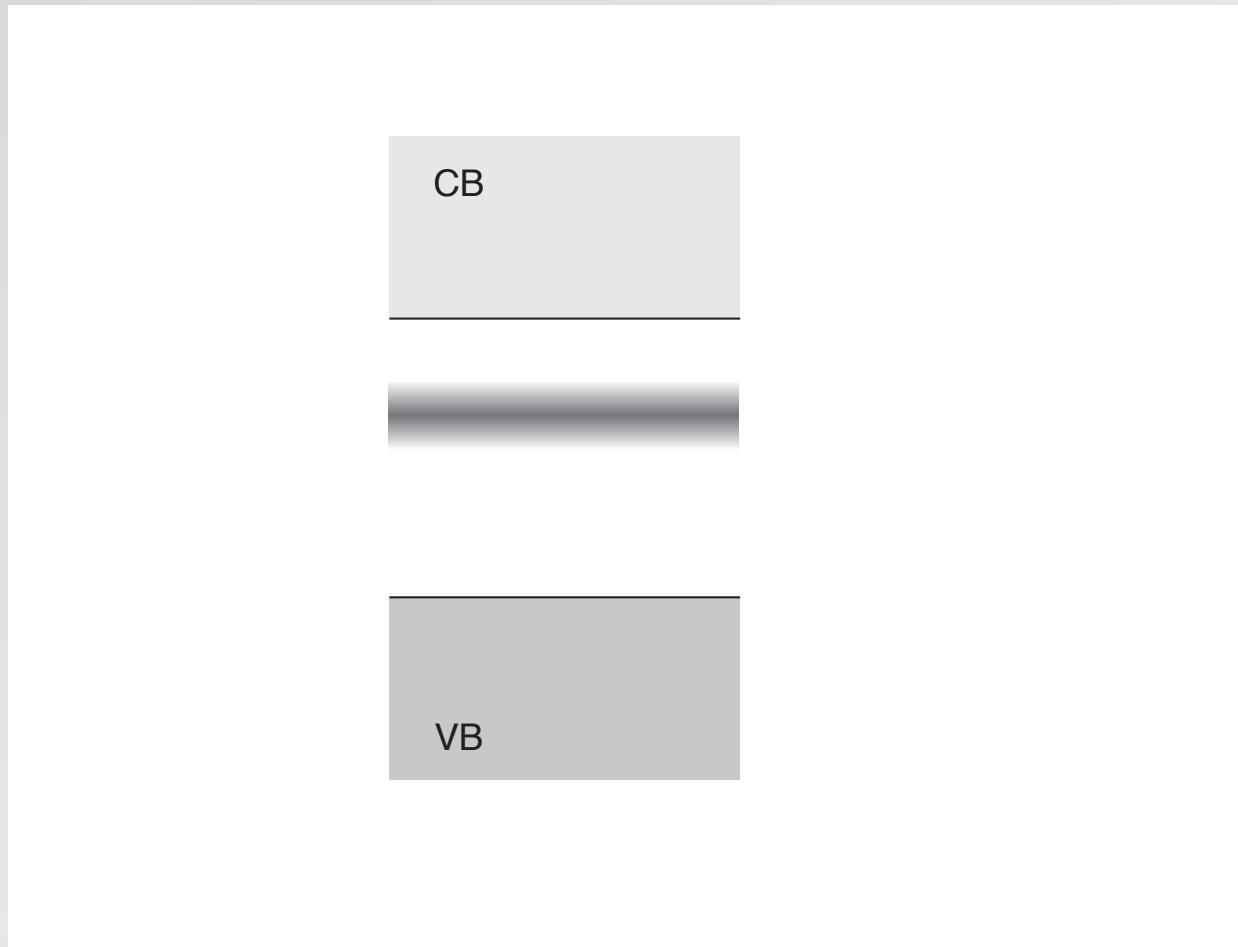


1 properties

2 intermediate band

3 devices

black silicon has an intermediate band

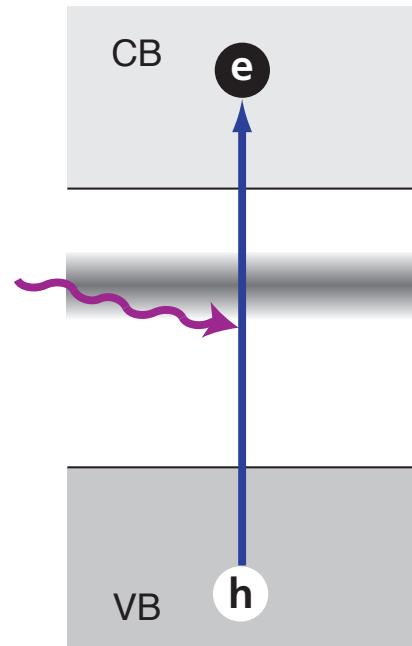


1 properties

2 intermediate band

3 devices

absorbs same photons as ordinary silicon...

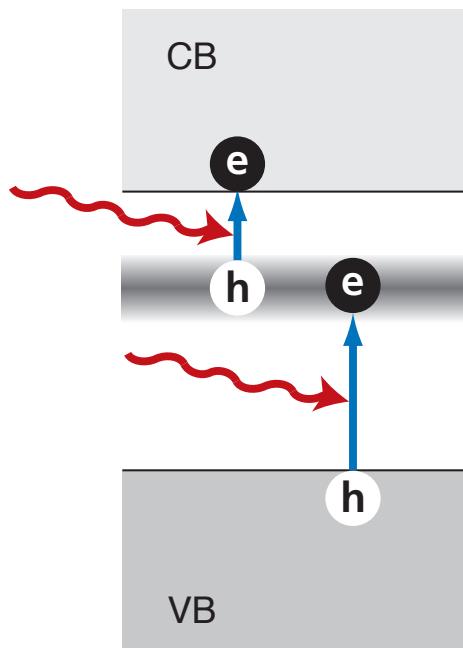


1 properties

2 intermediate band

3 devices

...but extends absorption to longer wavelengths

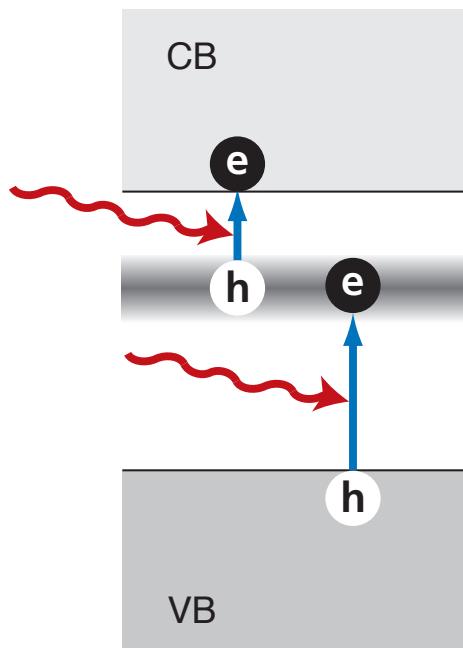


1 properties

2 intermediate band

3 devices

could theoretically get efficiencies over 50%

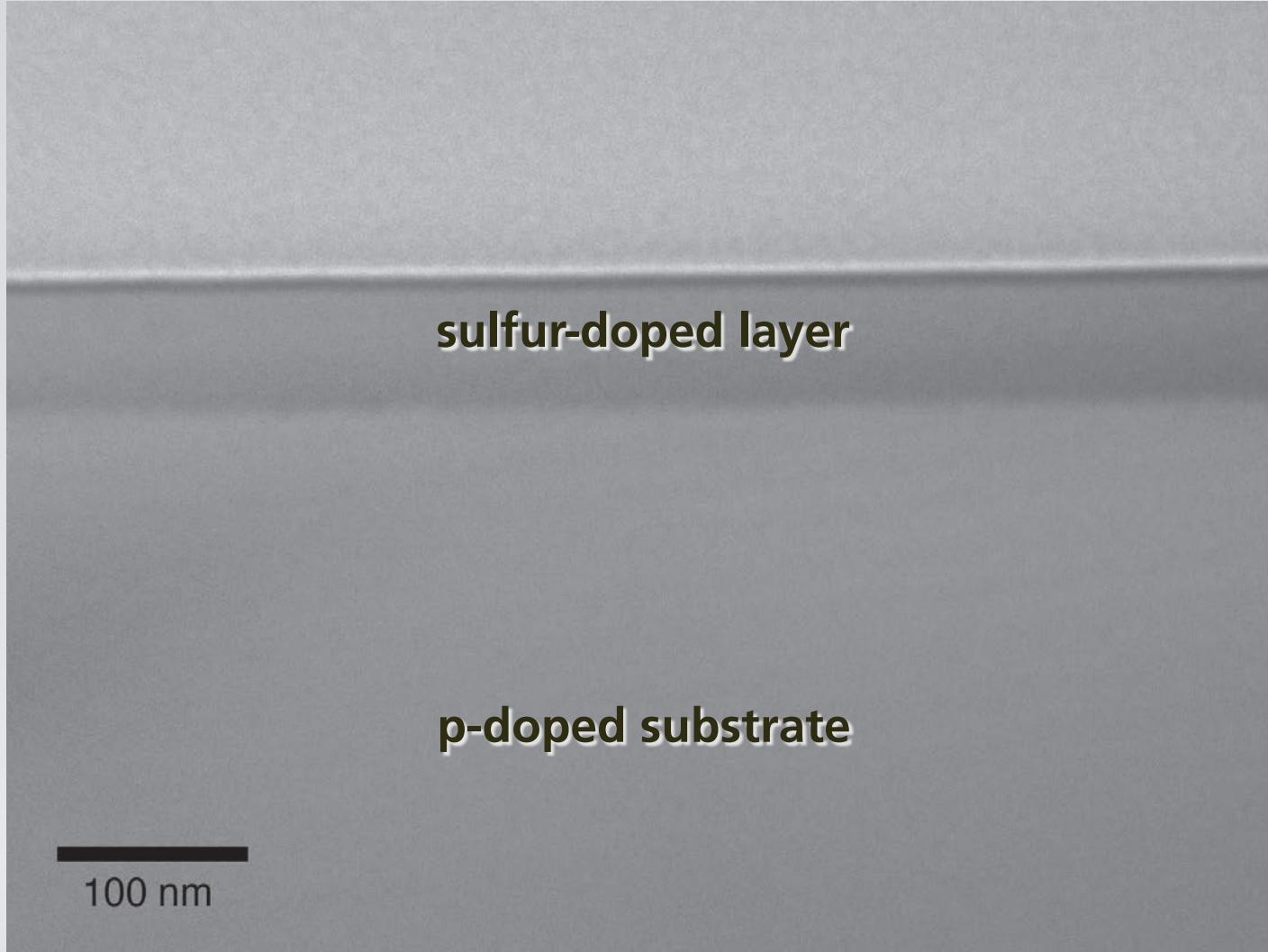


1 properties

2 intermediate band

3 devices

should have shallow junction below surface

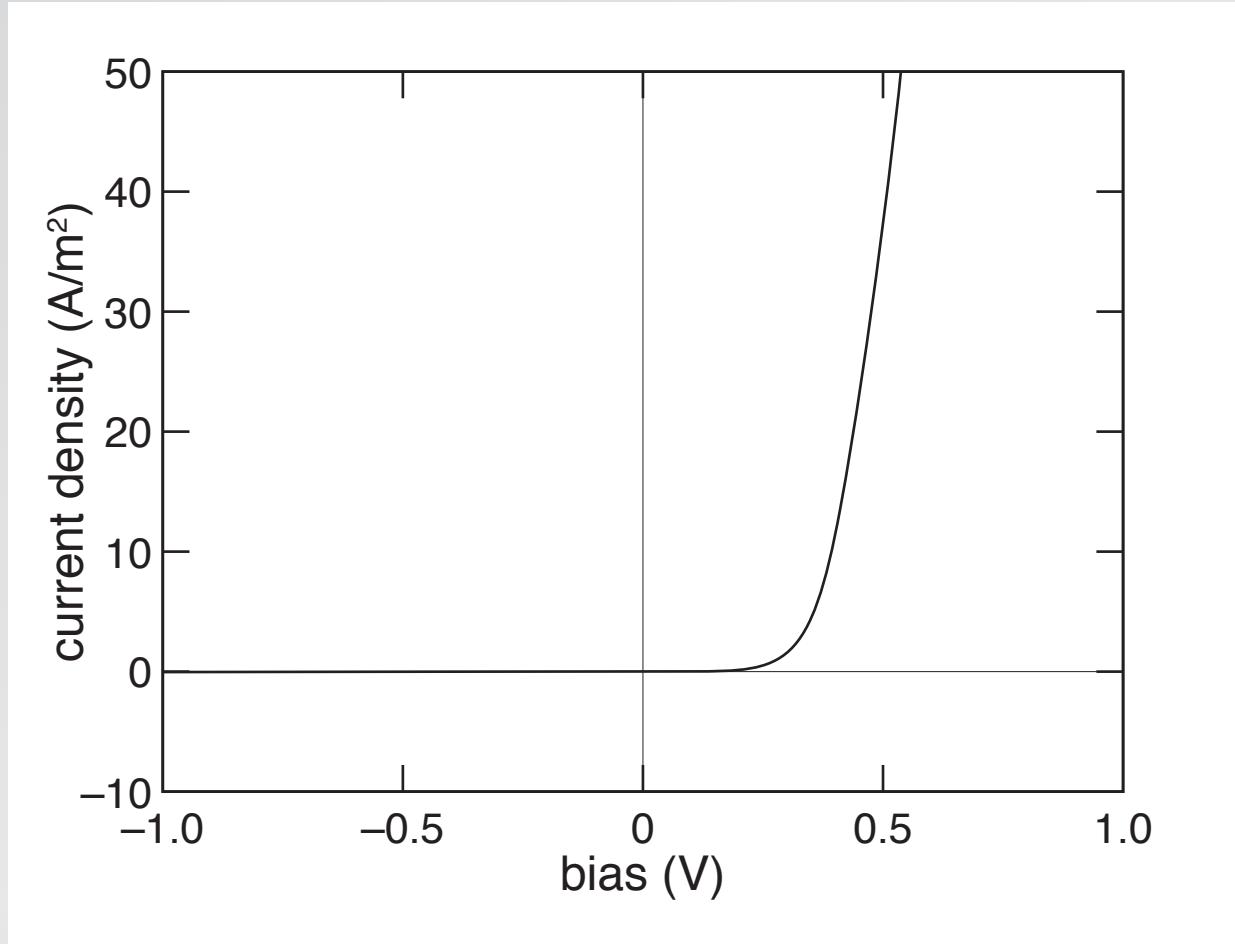


1 properties

2 intermediate band

3 devices

excellent rectification (after annealing)

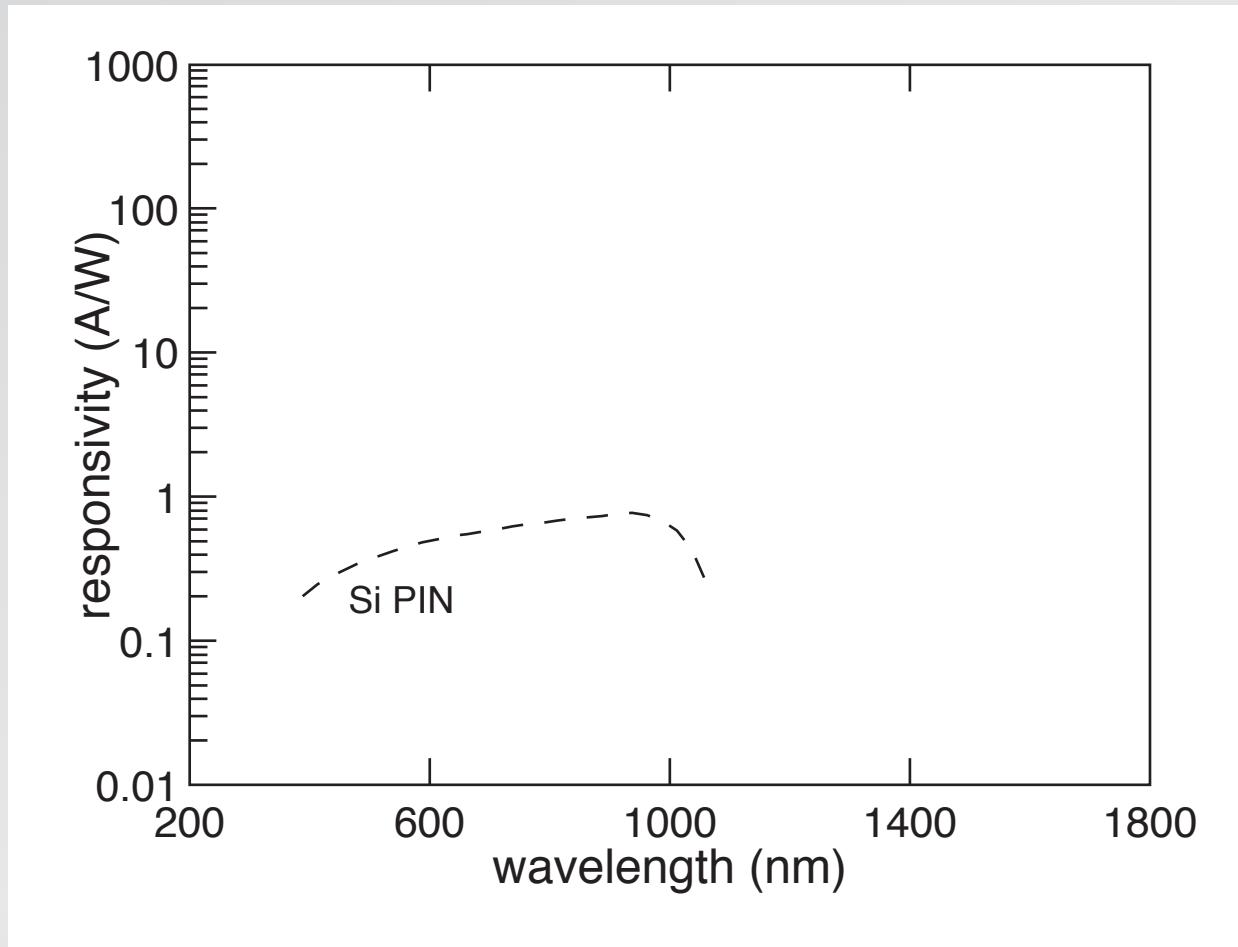


1 properties

2 intermediate band

3 devices

responsivity

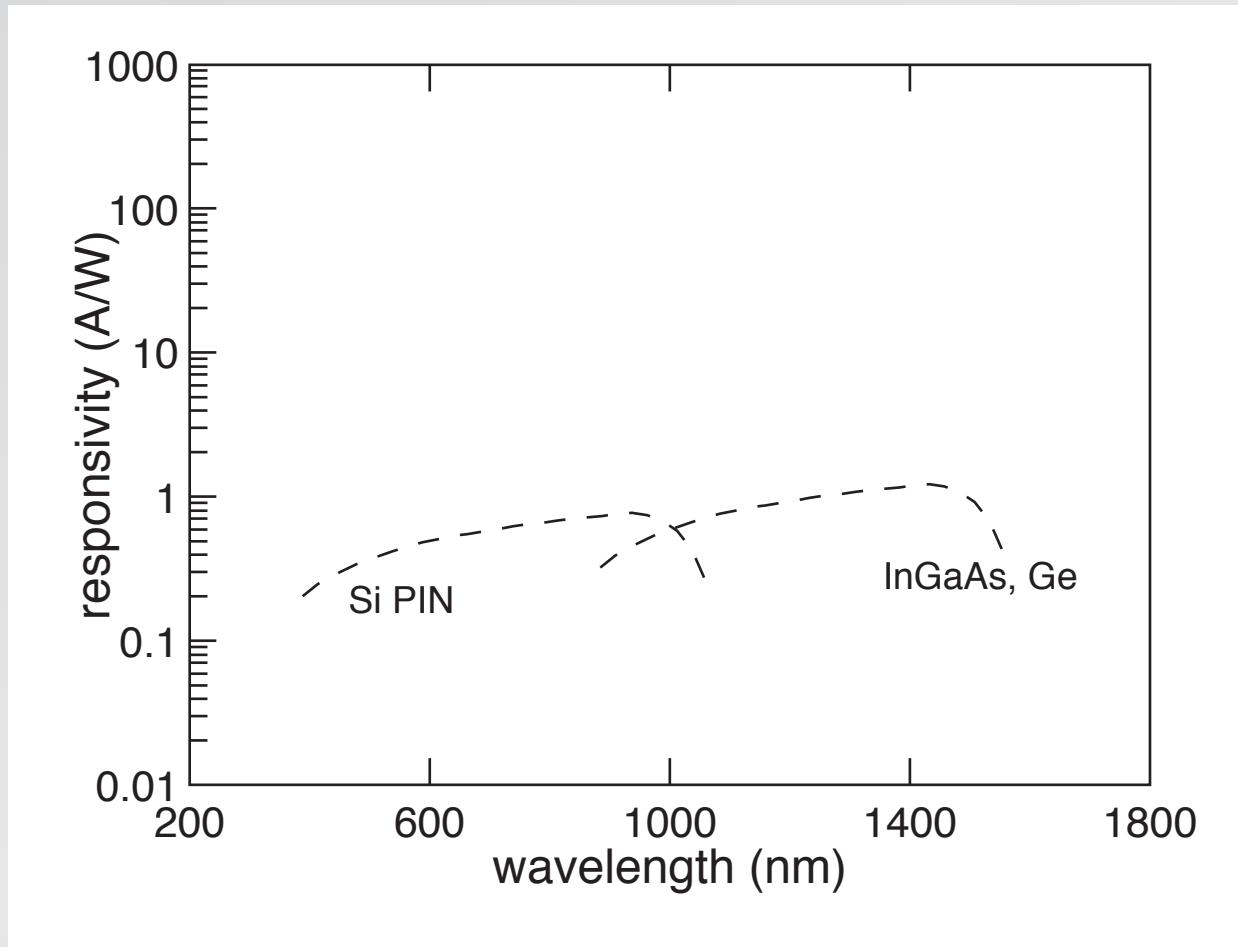


1 properties

2 intermediate band

3 devices

responsivity

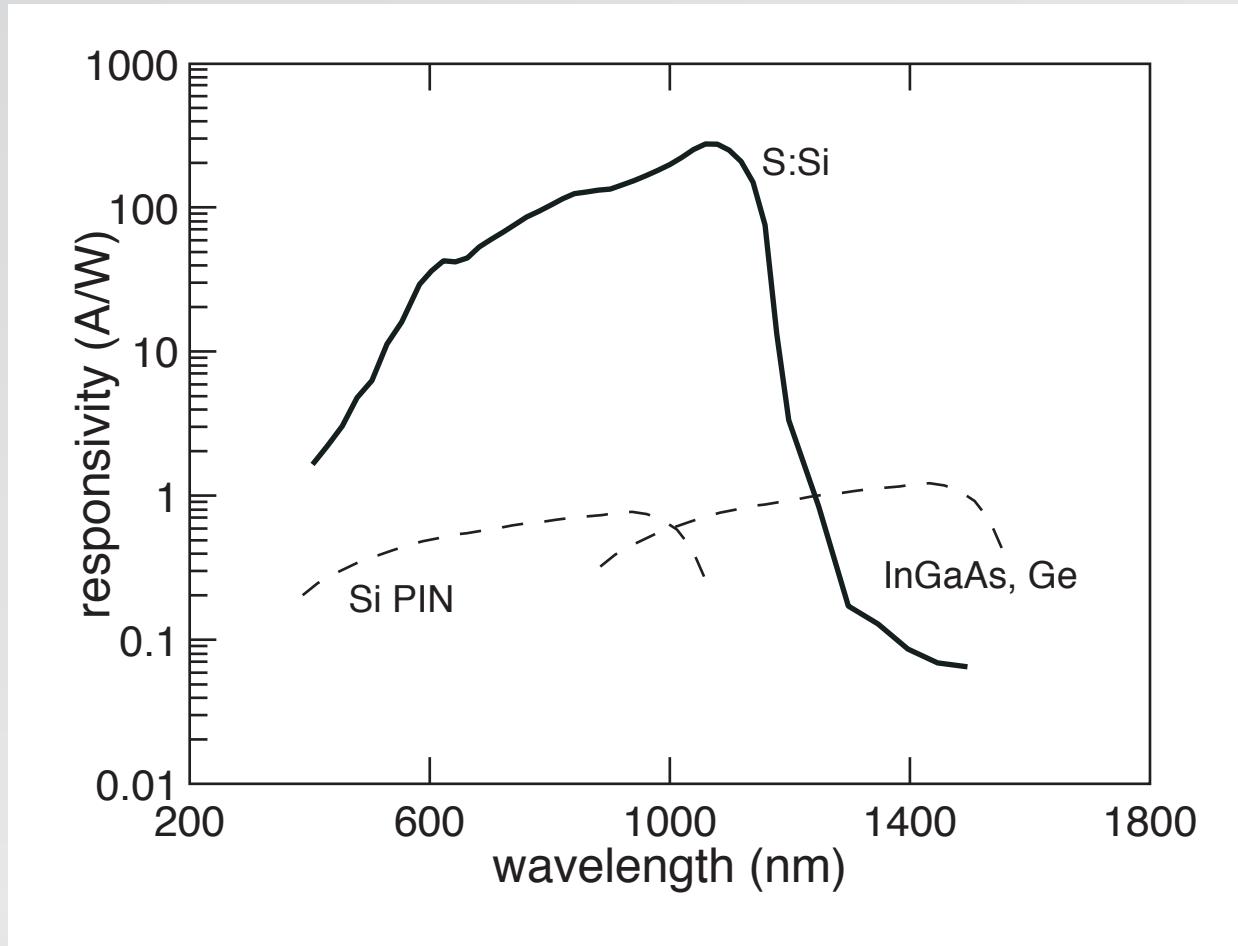


1 properties

2 intermediate band

3 devices

responsivity

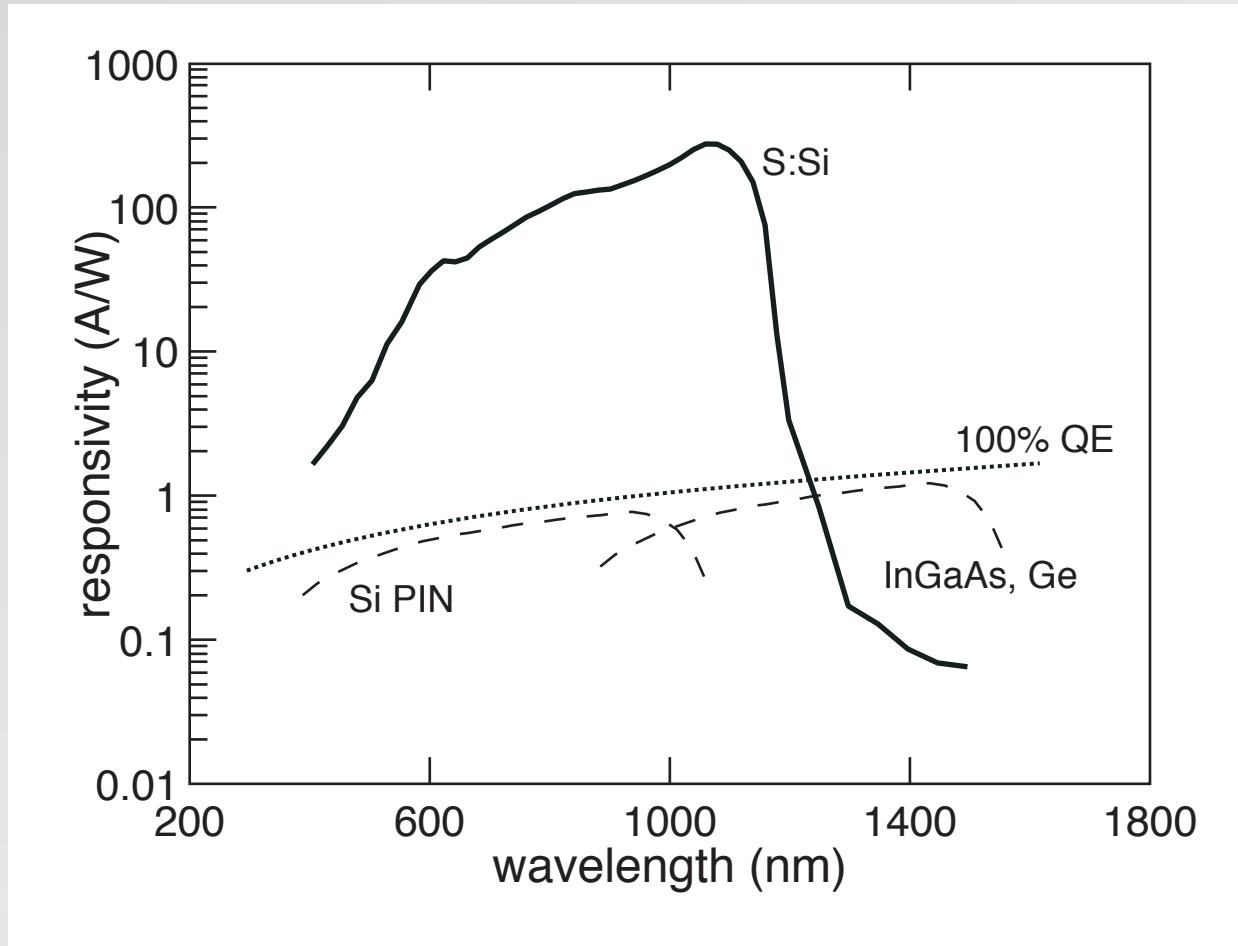


1 properties

2 intermediate band

3 devices

responsivity

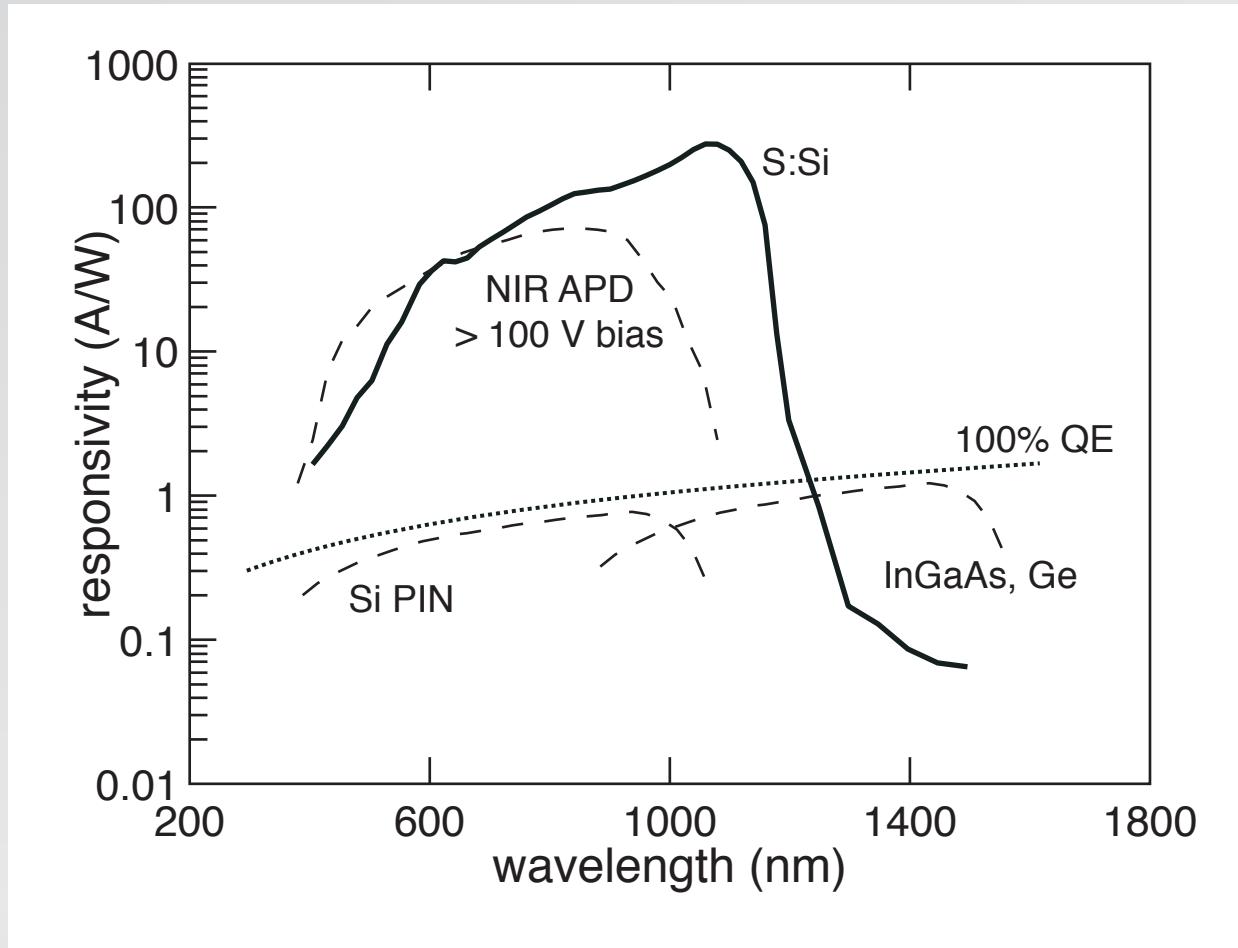


1 properties

2 intermediate band

3 devices

responsivity



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?

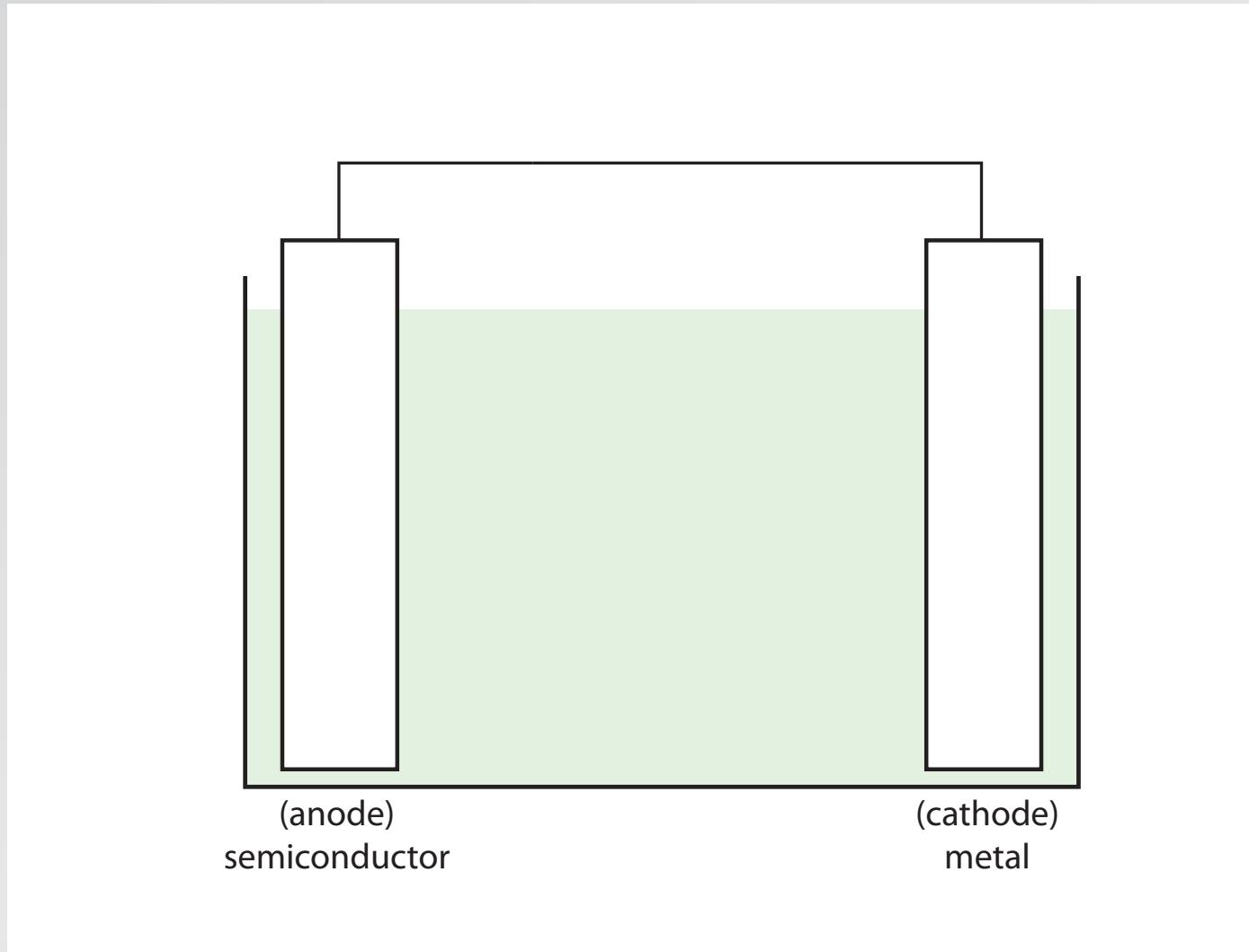
Potential benefits for photovoltaics

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

1 intermediate band

2 Si devices

water splitting

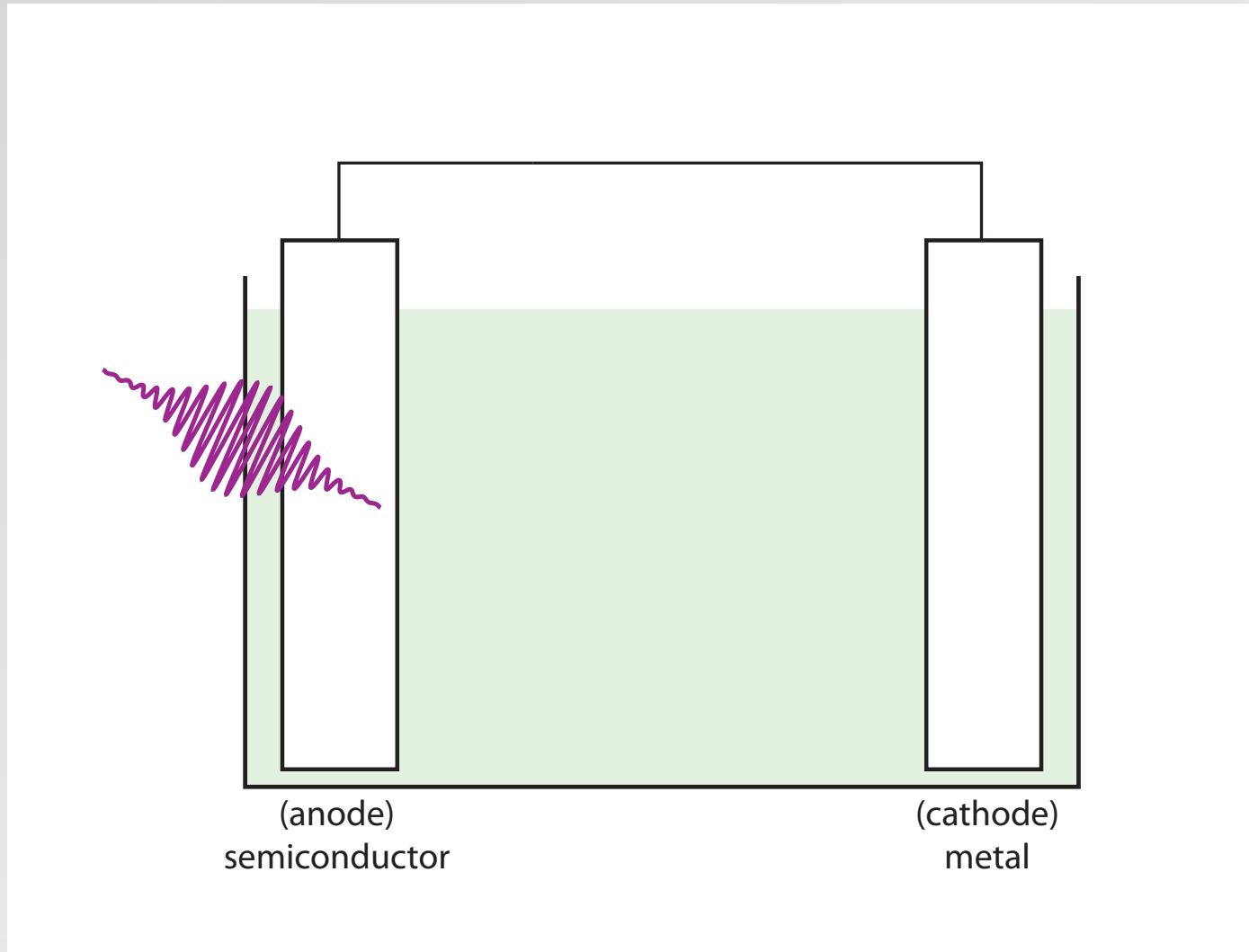


1 properties

2 intermediate band

3 devices

water splitting

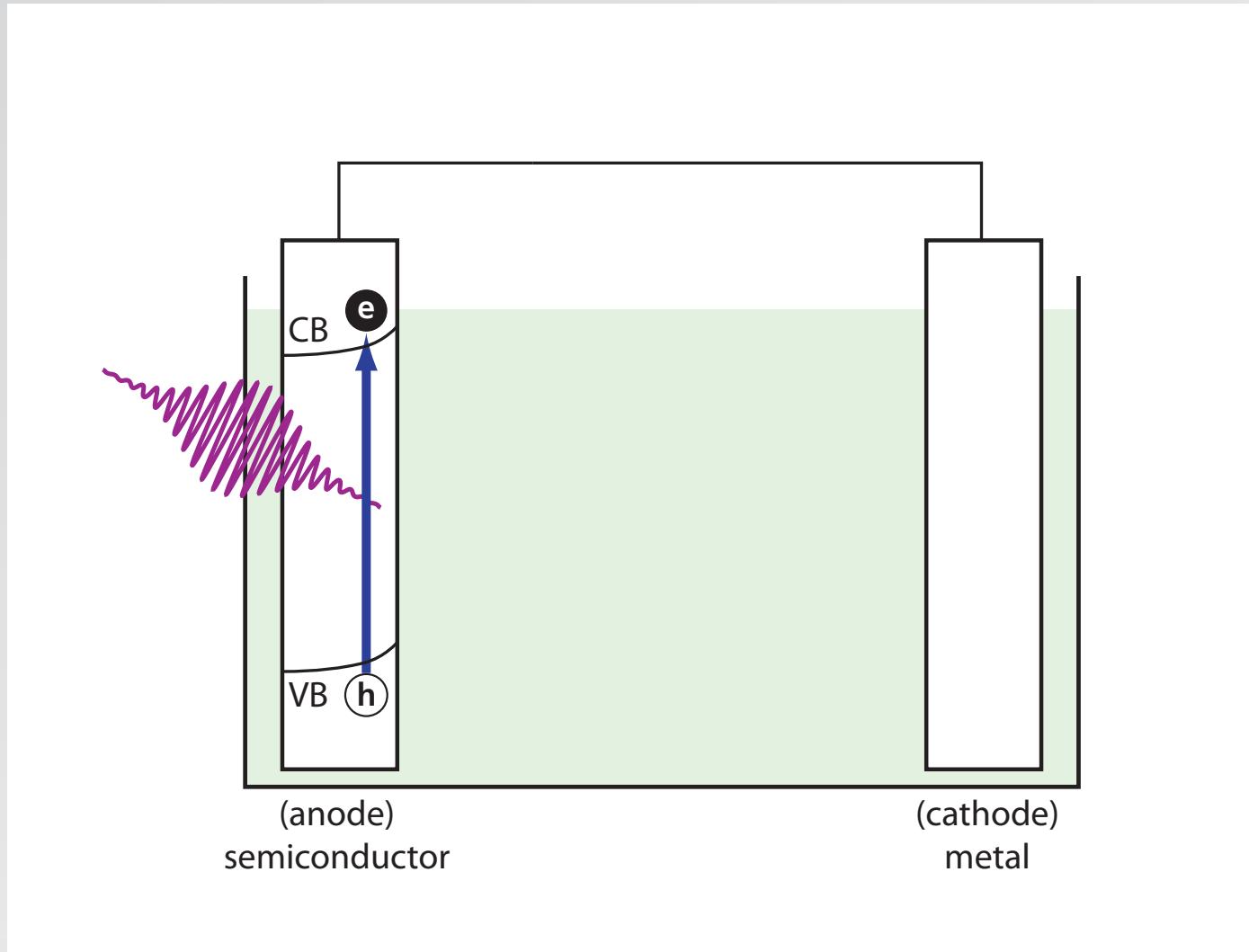


1 properties

2 intermediate band

3 devices

water splitting

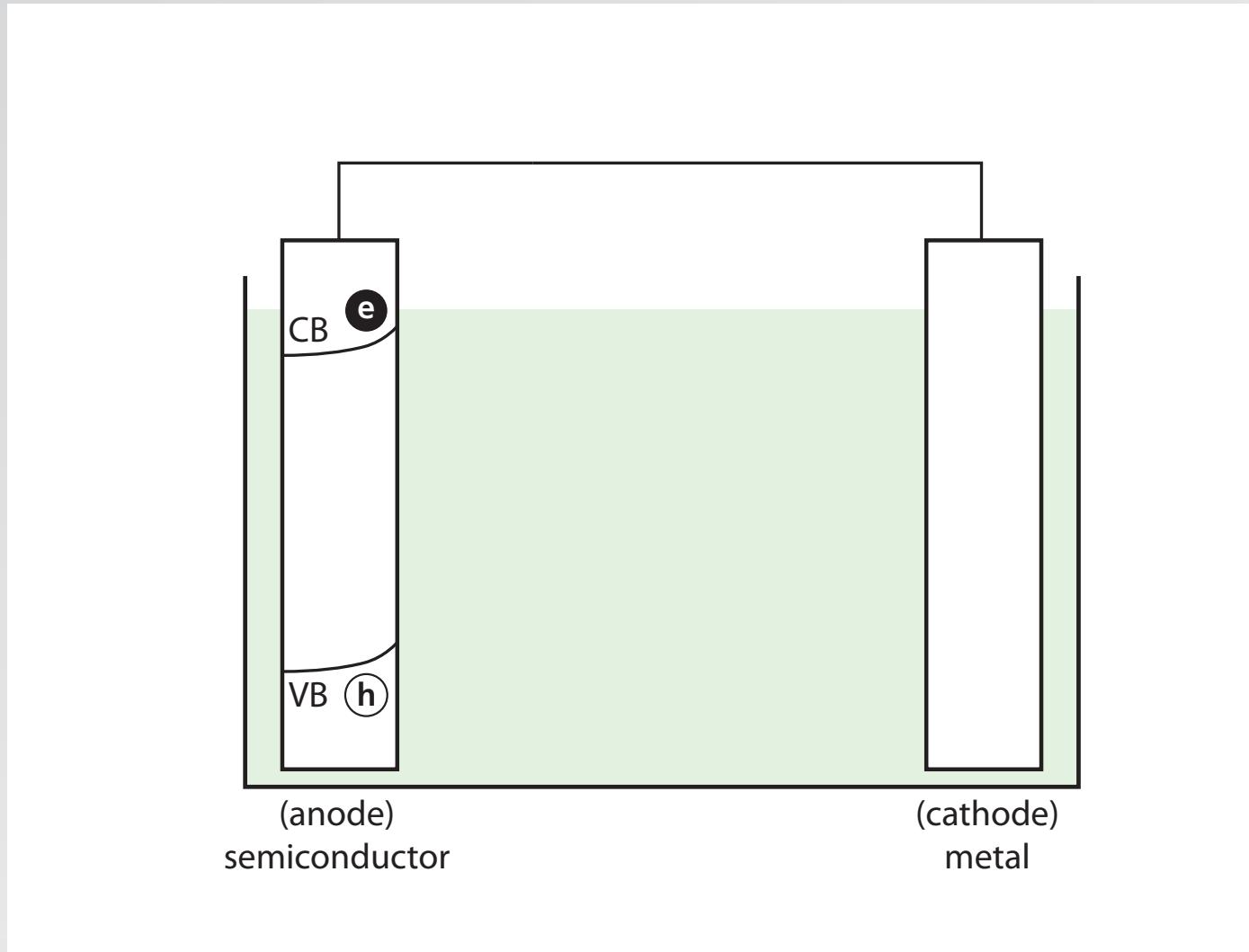


1 properties

2 intermediate band

3 devices

water splitting

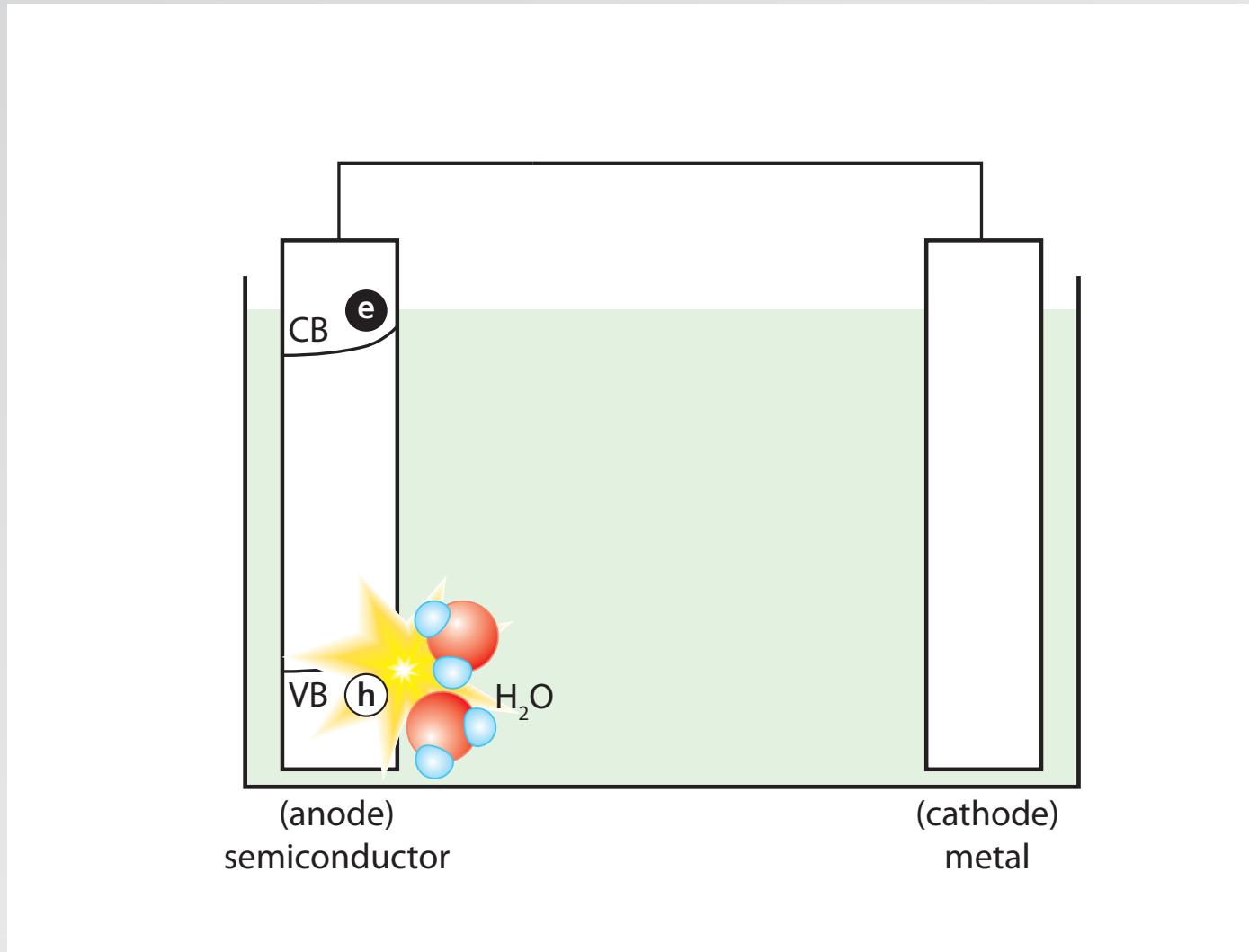


1 properties

2 intermediate band

3 devices

water splitting

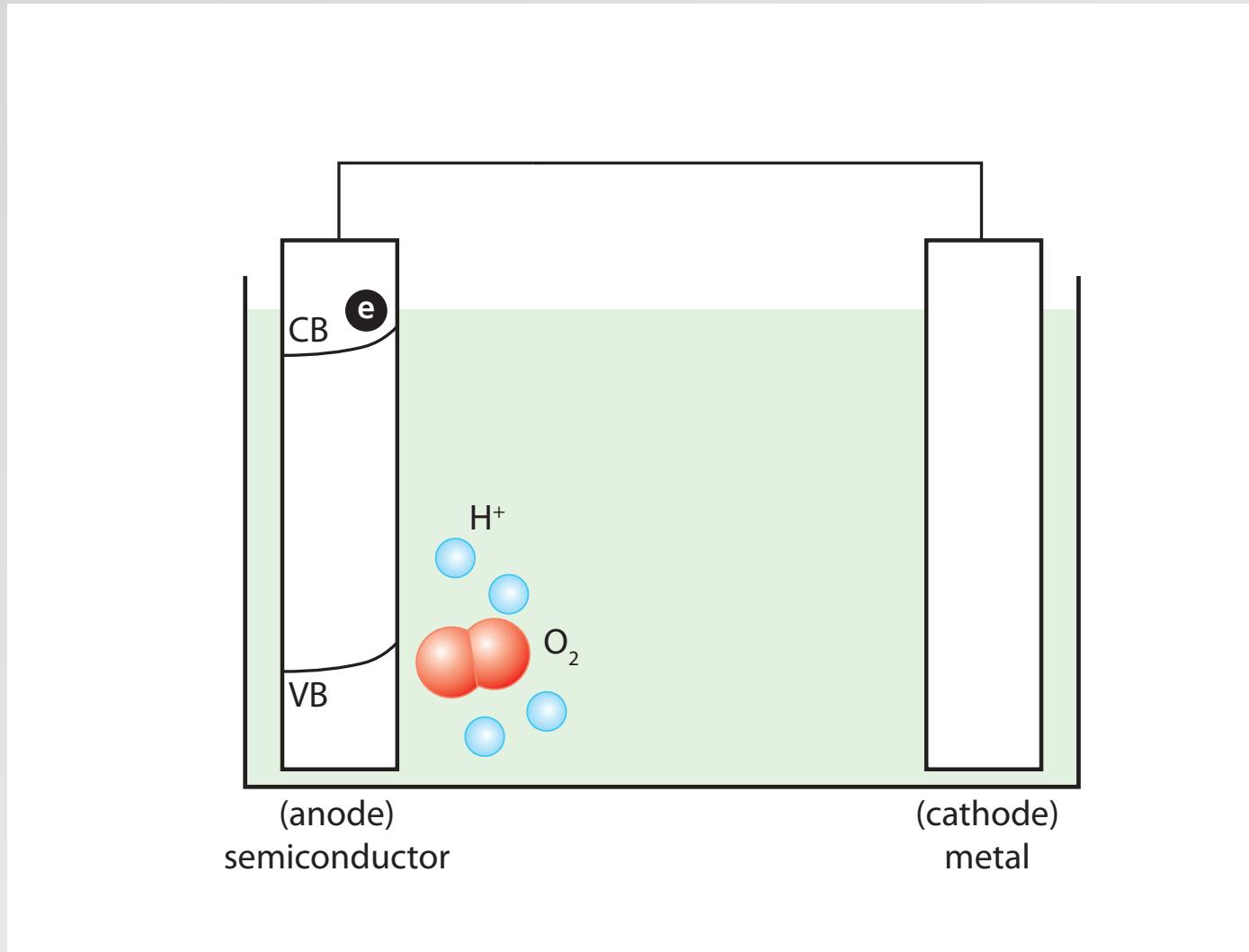


1 properties

2 intermediate band

3 devices

water splitting

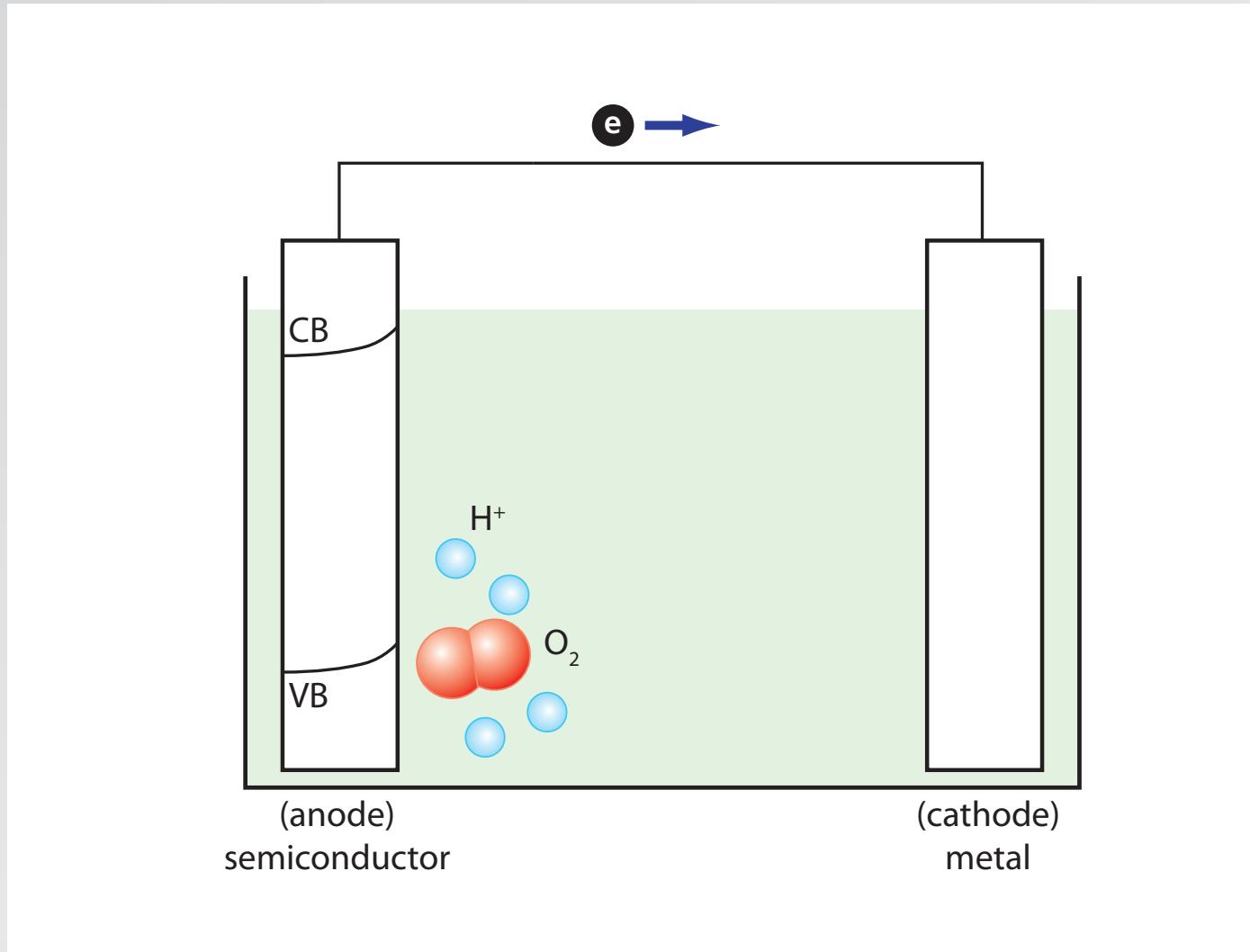


1 properties

2 intermediate band

3 devices

water splitting

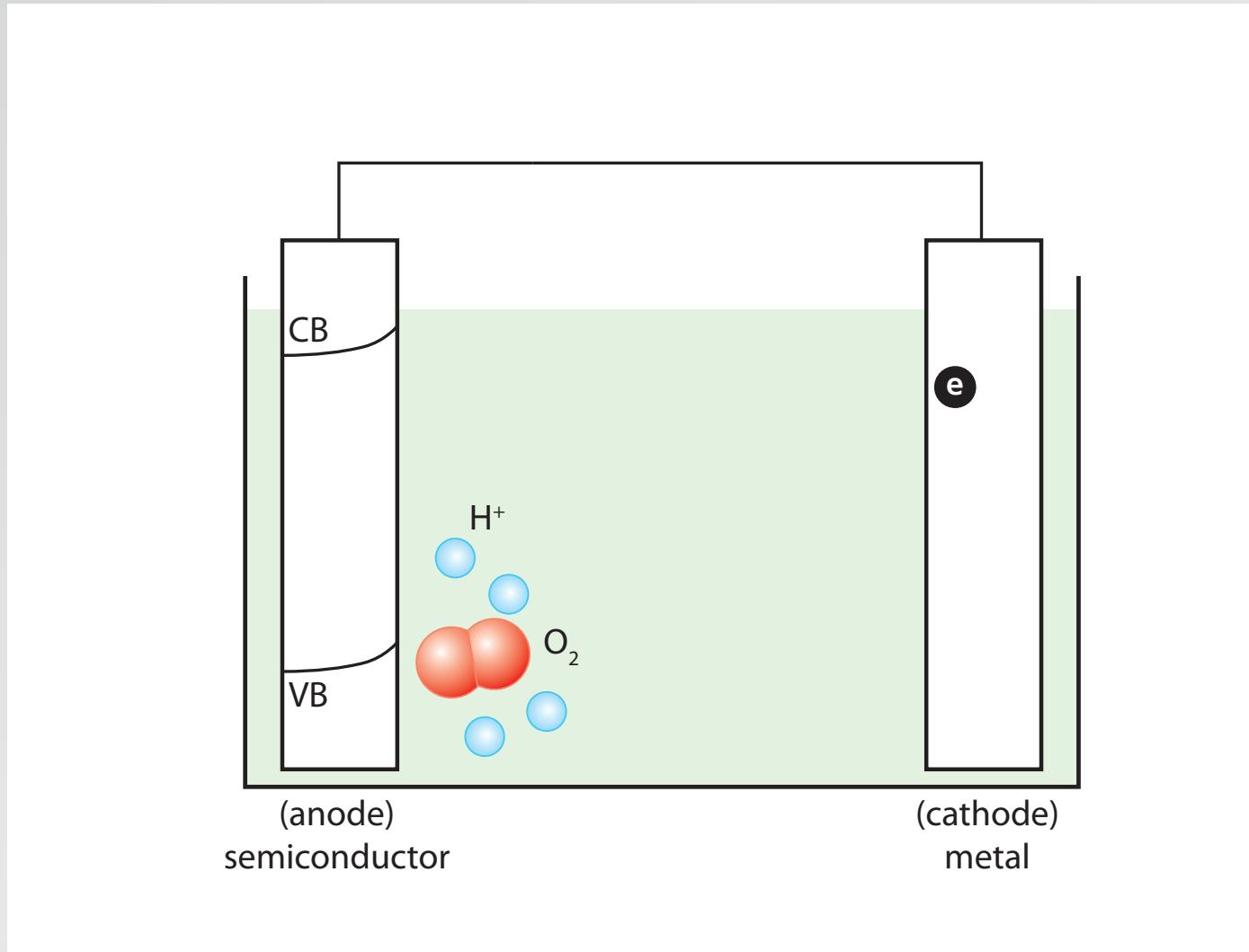


1 properties

2 intermediate band

3 devices

water splitting

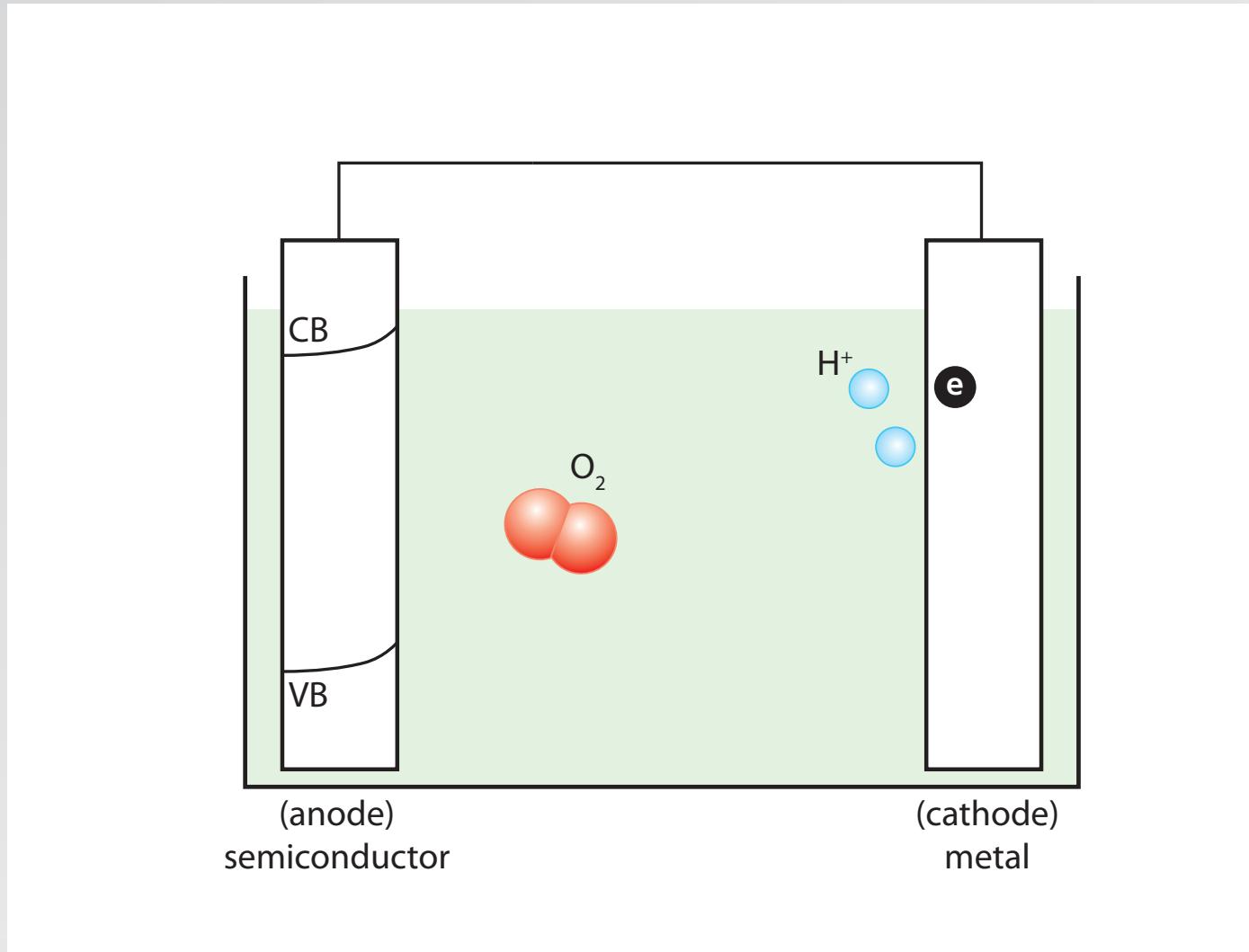


1 properties

2 intermediate band

3 devices

water splitting

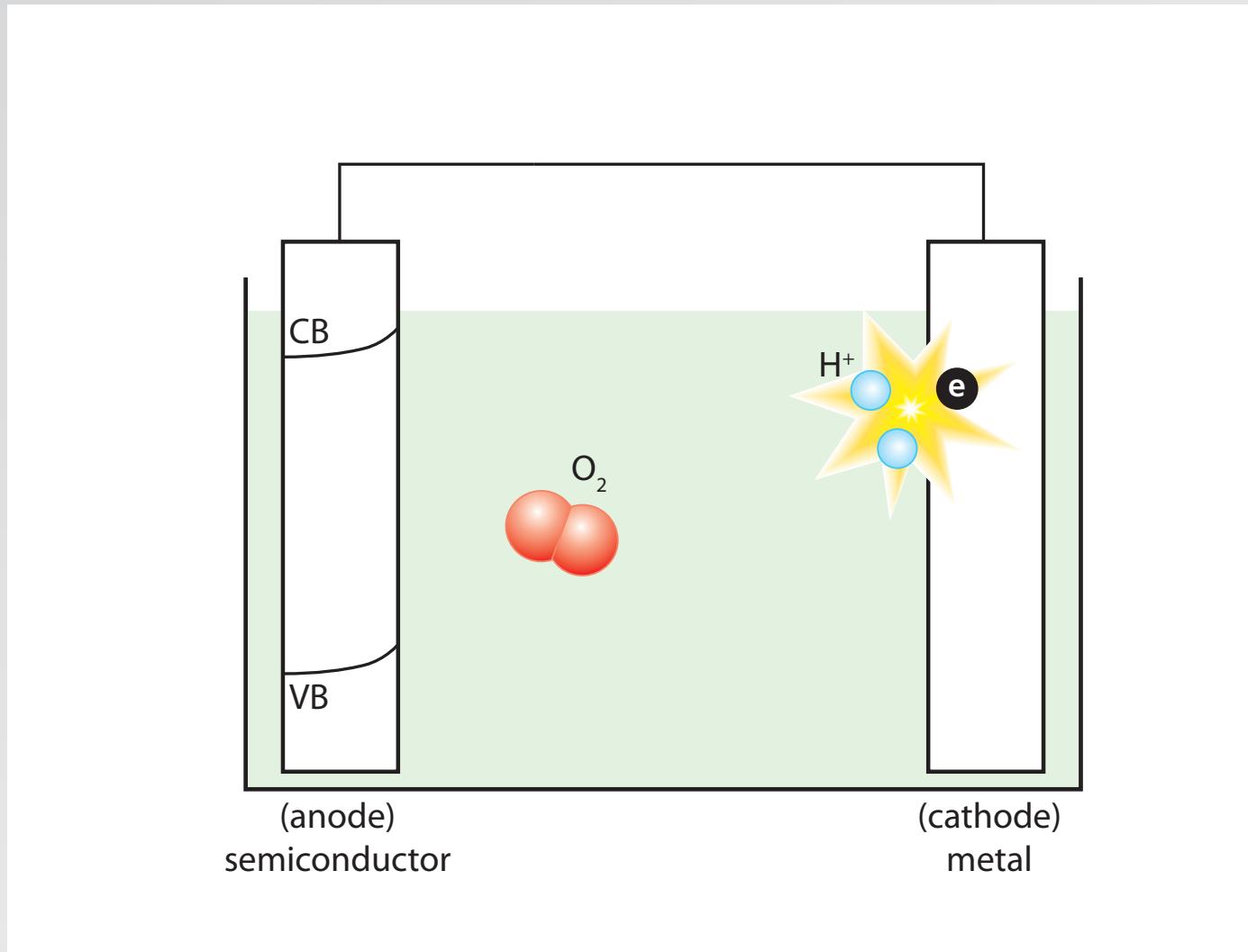


1 properties

2 intermediate band

3 devices

water splitting

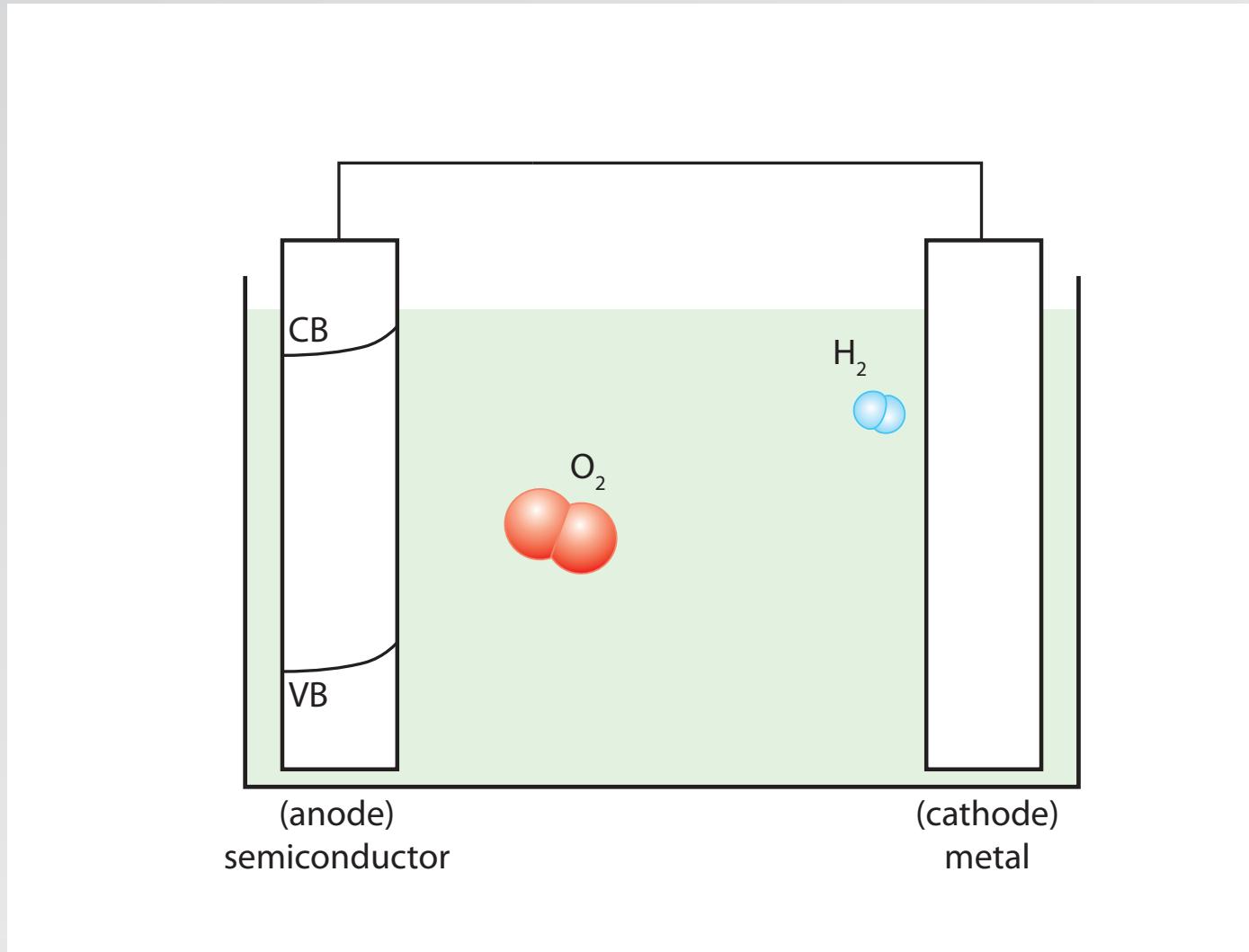


1 properties

2 intermediate band

3 devices

water splitting

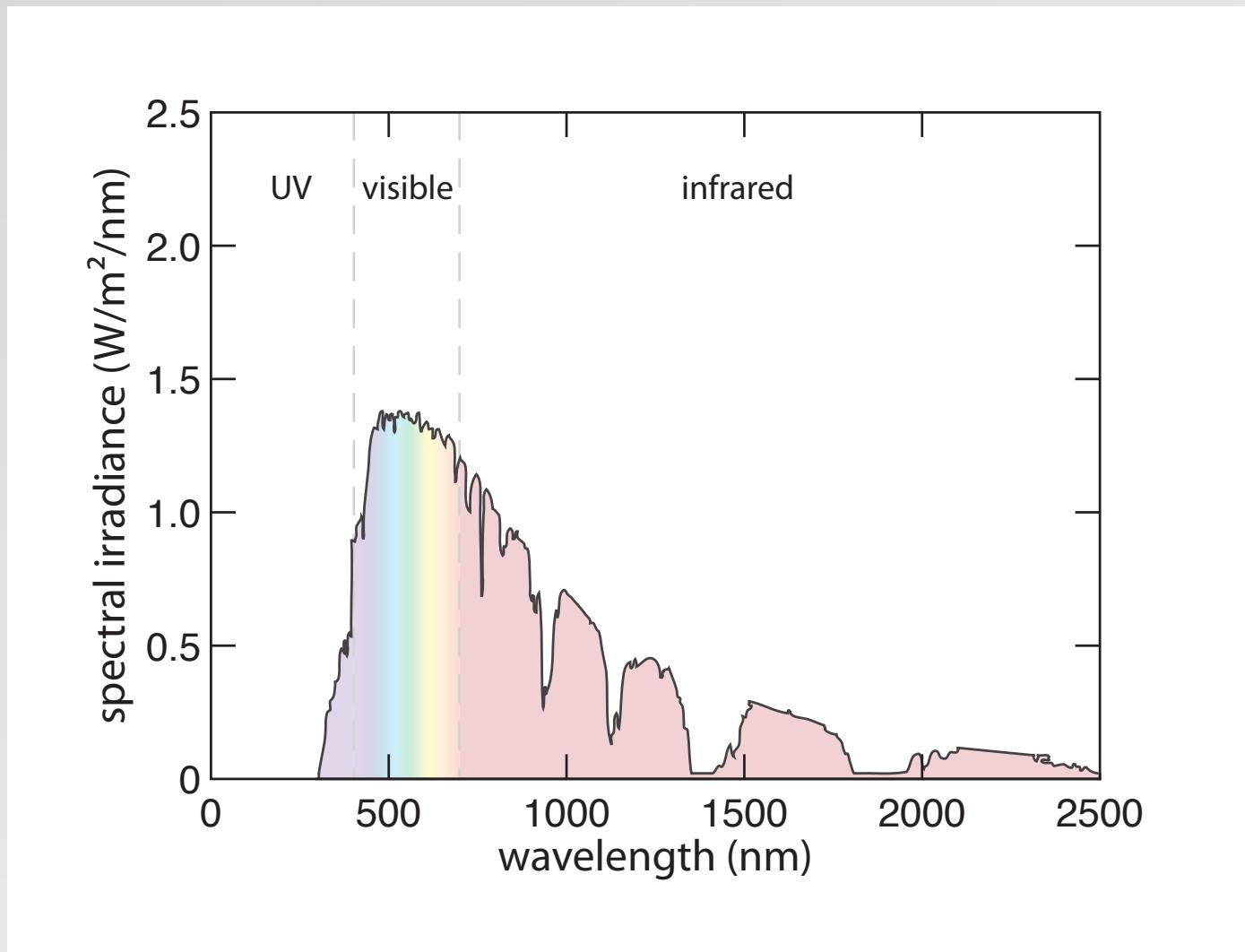


1 properties

2 intermediate band

3 devices

solar radiation spectrum

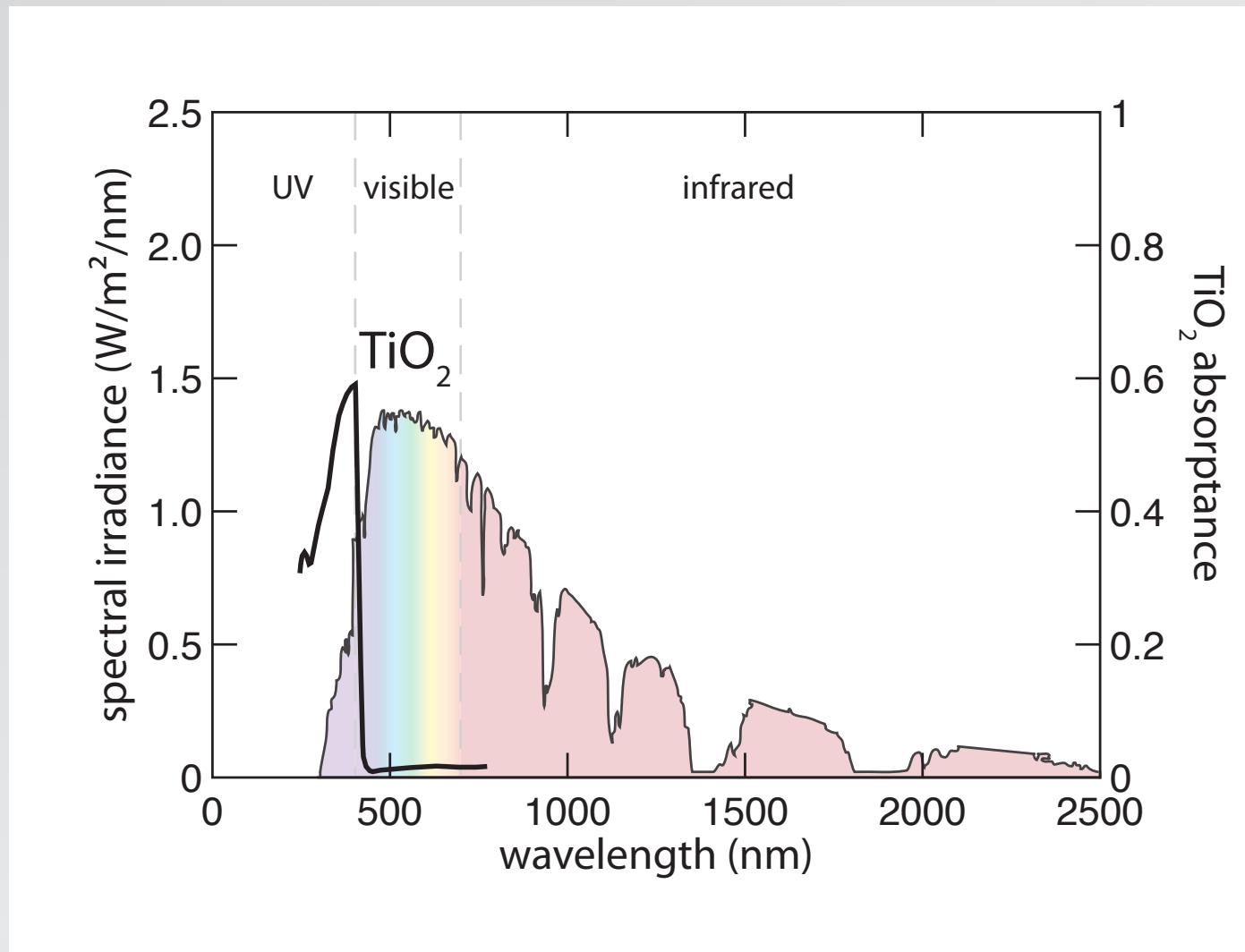


1 properties

2 intermediate band

3 devices

solar radiation spectrum

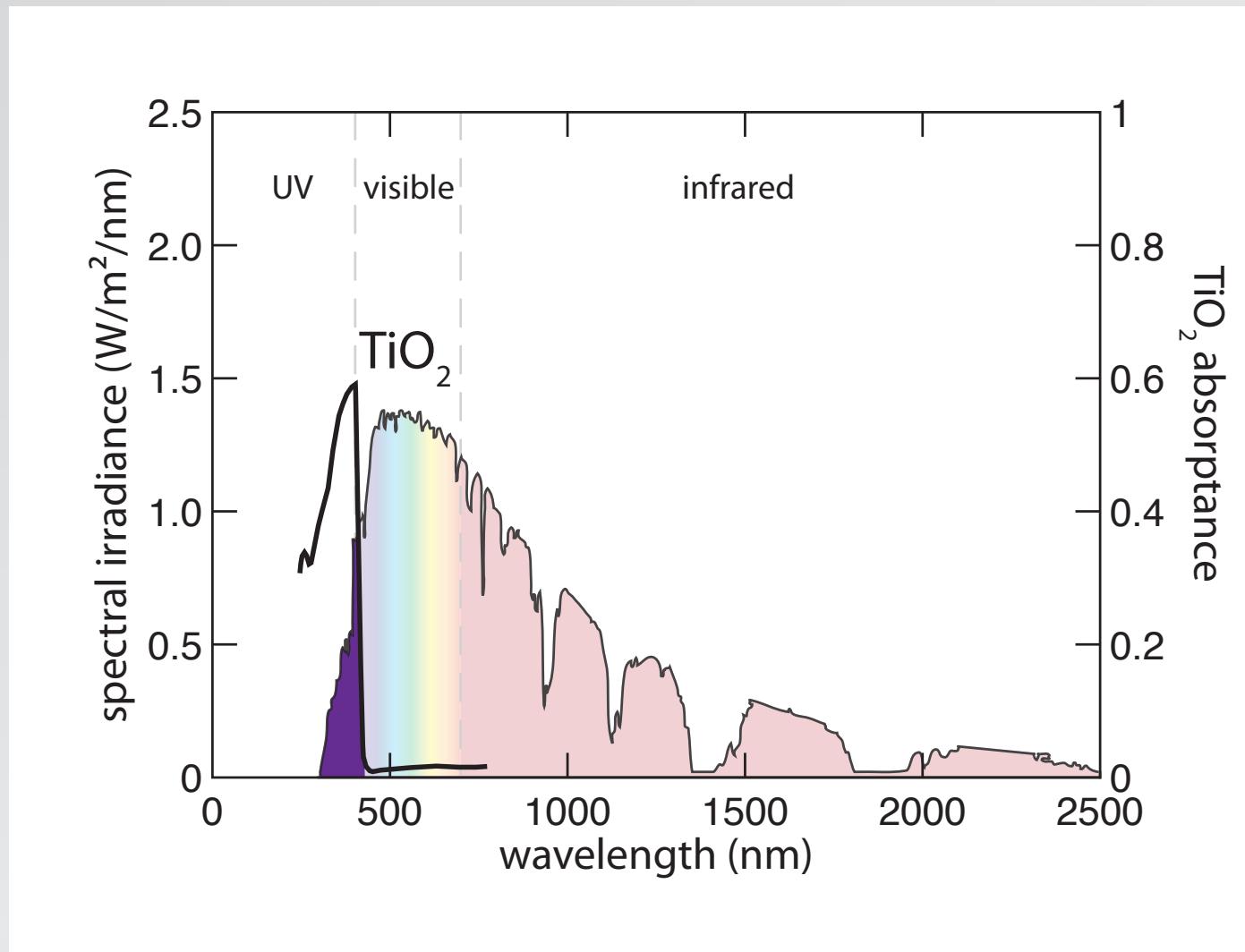


1 properties

2 intermediate band

3 devices

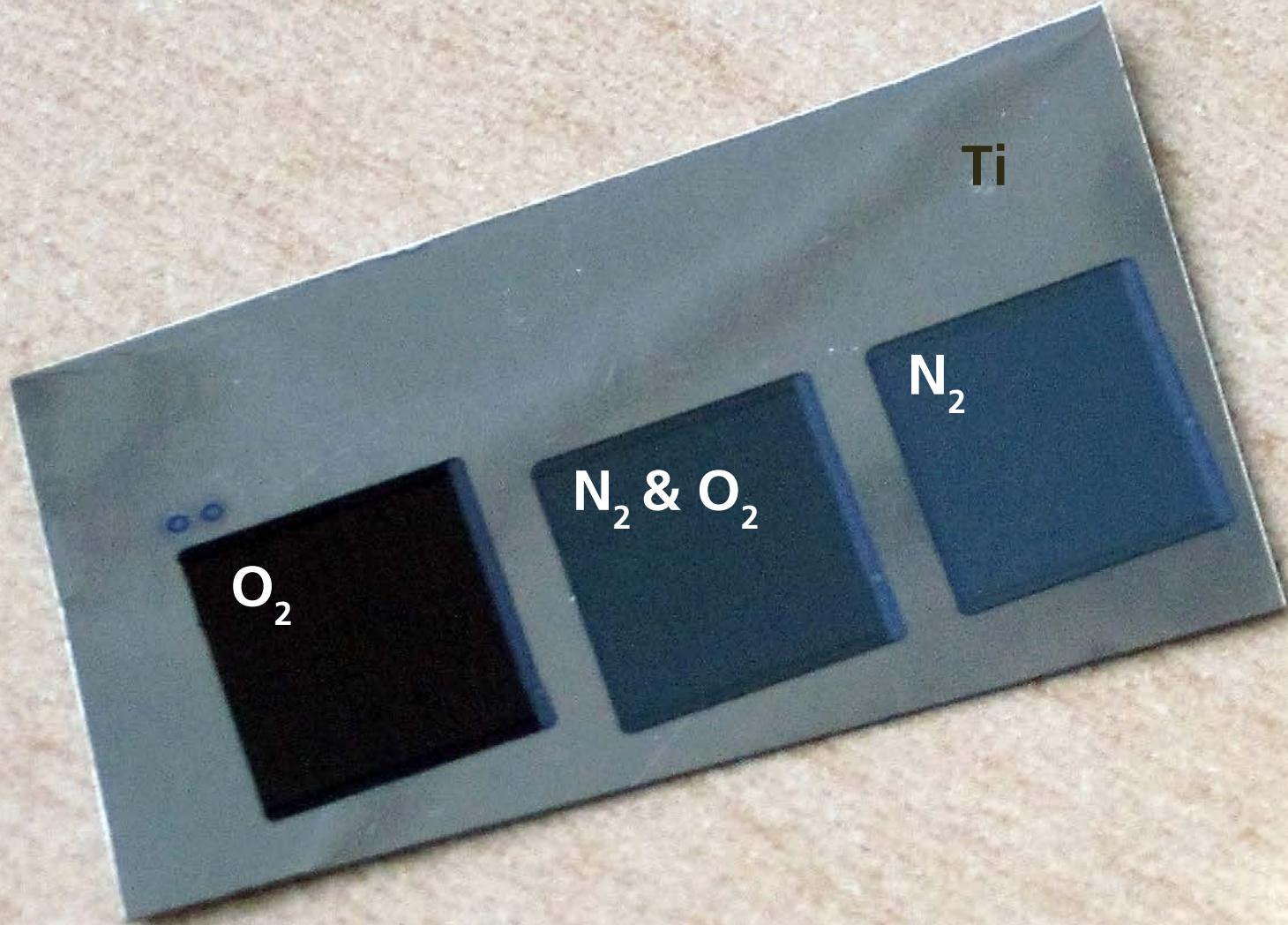
solar radiation spectrum



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





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DARPA

Department of Energy

NDSEG

National Science Foundation

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