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ADRIATICO RESEARCH CONFERENCE on

LASERS IN SURFACE SCIENCE

11-15 September 2000

Miramare - Trieste, Italy

Lifetimes of conduction bands and surface states at semiconductor surfaces

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Lasers in Surface Science, Trieste 11. -15. 09. 2000

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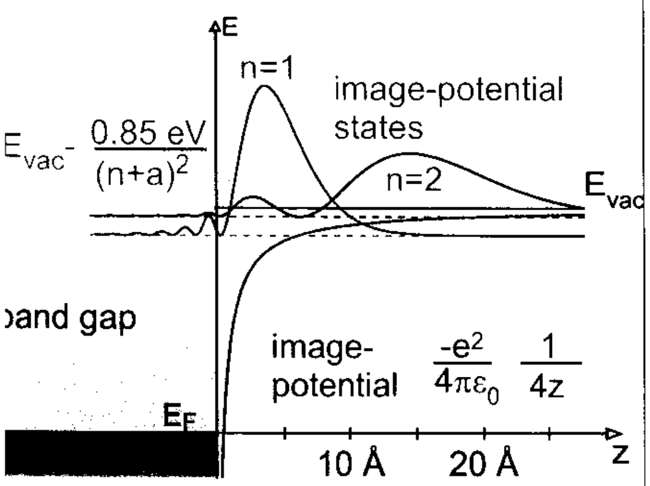
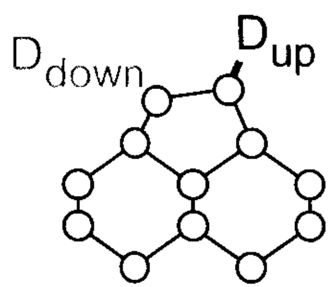
Dephasing and energy relaxation of image-potential states



- *confinement of electrons*
- *quantum-phase decay*

Lifetimes of conduction bands and surface states at semiconductor surfaces

Si(001)

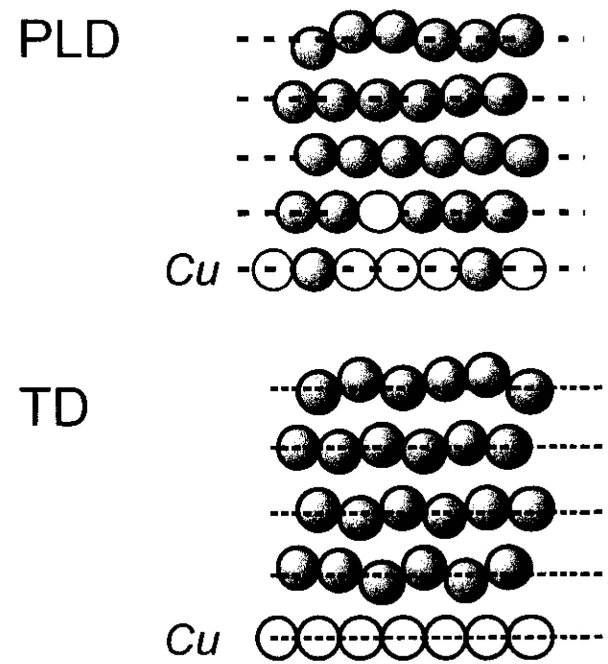


4H - SiC(0001)

- *electron & hole dynamics*
- *electronic structure*

Pulsed Laser Deposition

growth of metal layers - 4 ML Fe/Cu(001)



H. Jenniches et al. PRB ***59****(1999)1196*

Outline

- Introduction, time-resolved two-photon photoemission (2ppe)

- Linewidth analysis

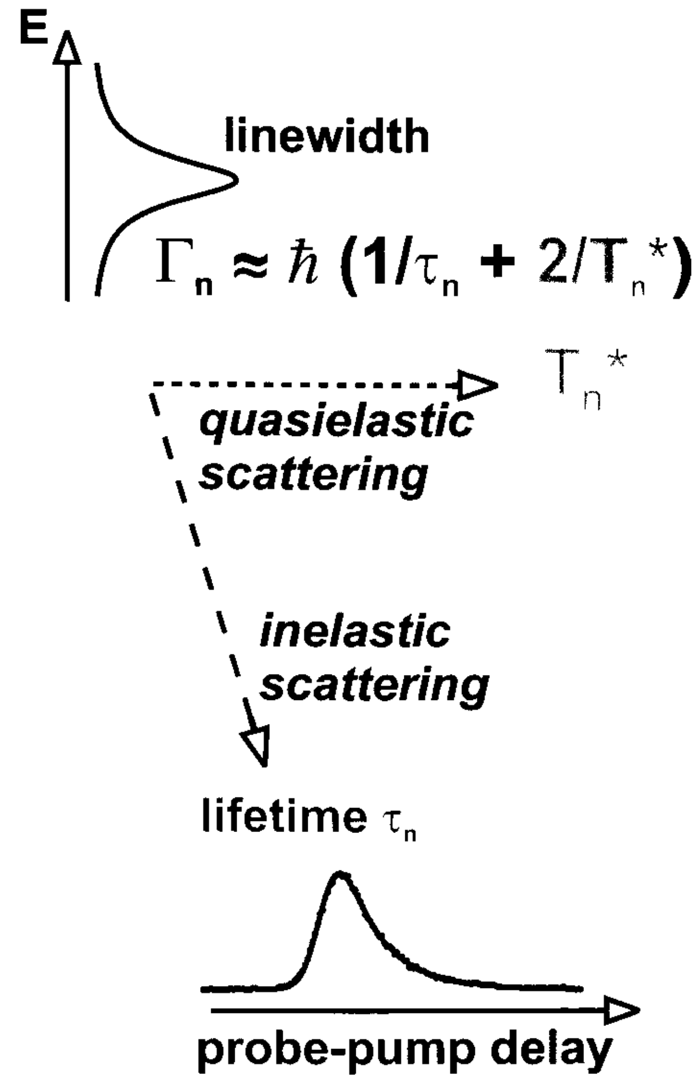
- 4H - SiC(0001)

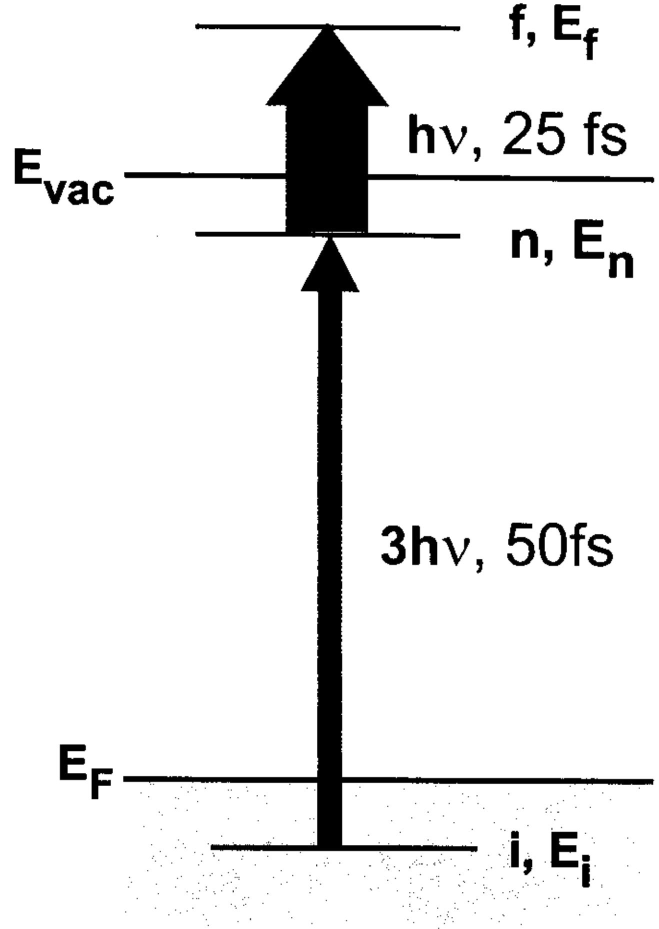
  Mott-Hubbard splitting of dangling-bond states

- Si(001)

  π and π^* - dangling bond states
  electron dynamics and recombination

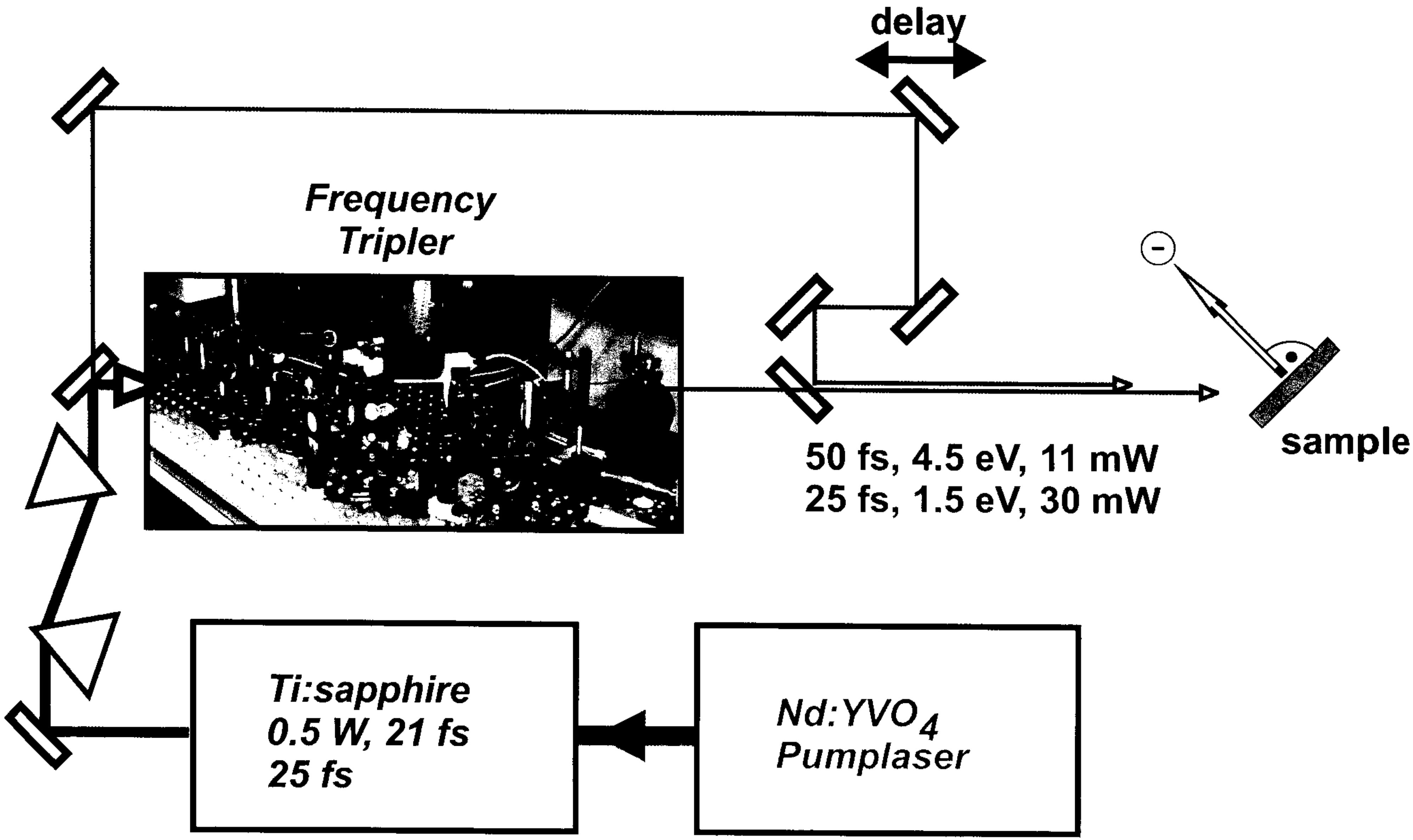
Energy and phase relaxation (decay and dephasing)





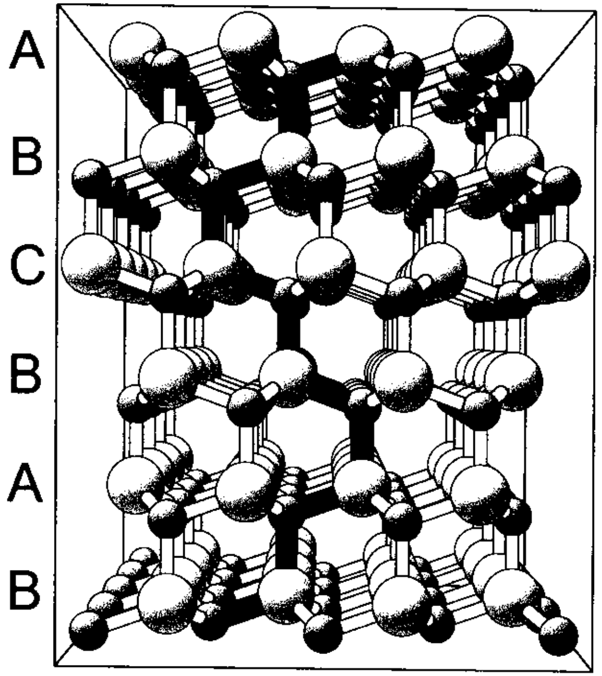
Two-photon photoemission

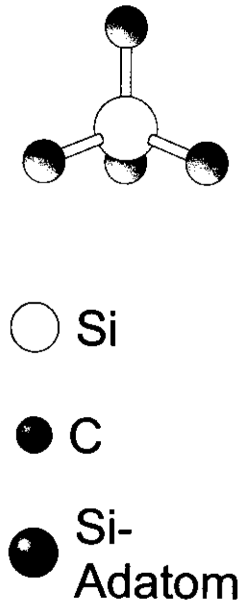
Experiment

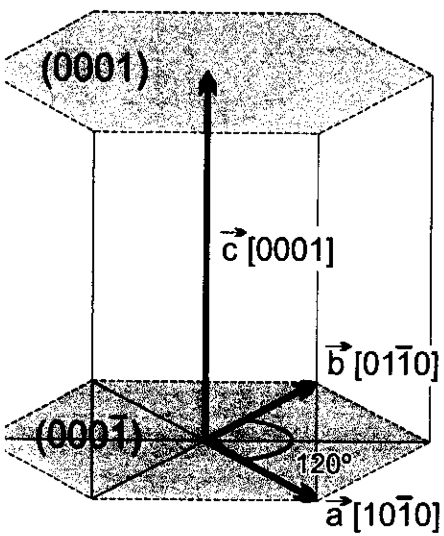


I. L. Shumay, 1997

Kristallstruktur von 4H-SiC(0001)



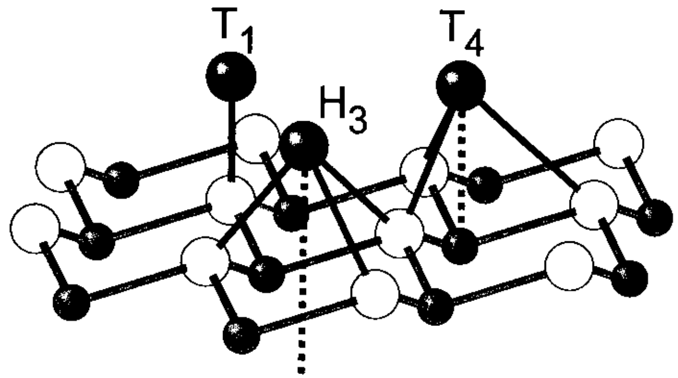


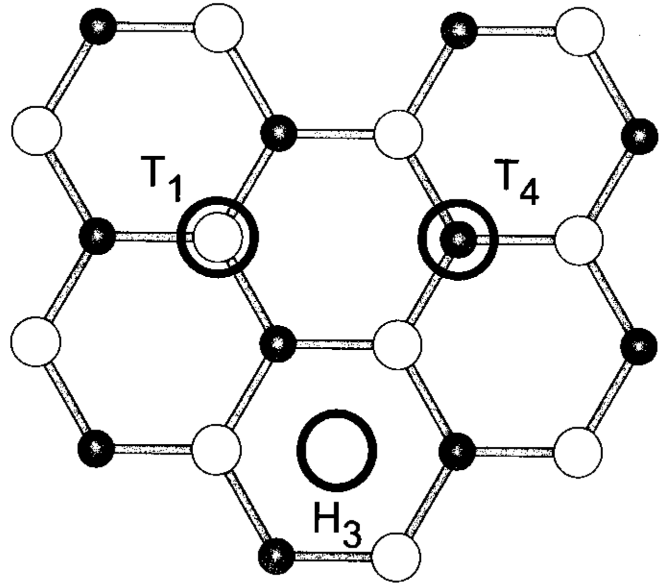


Polarität:

(0001) ↔ Si-terminiert
(000$\bar{1}$) ↔ C-terminiert

Oberfläche von Siliziumkarbid





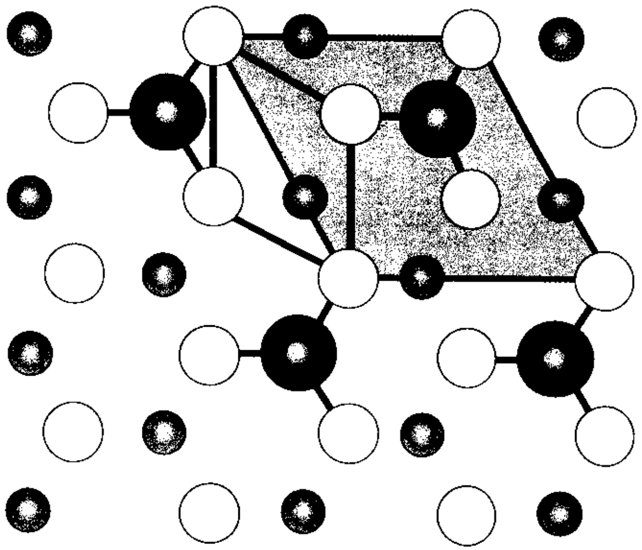
T_1 → 1-fach koord. 'Top'-Position der 1. atomaren Lage
T_4 → 4-fach koord. 'Top'-Position der 2. atomaren Lage
H_3 → 3-fach koord. 'Hollow'-Position der 1. Bilage

Warum 4H-SiC(0001)($\sqrt{3}\times\sqrt{3}$)R30° ?

Si-Adatom auf T_4

Starke et al., Phys. Rev. Lett. 82 (1999) 2107

Aufsicht

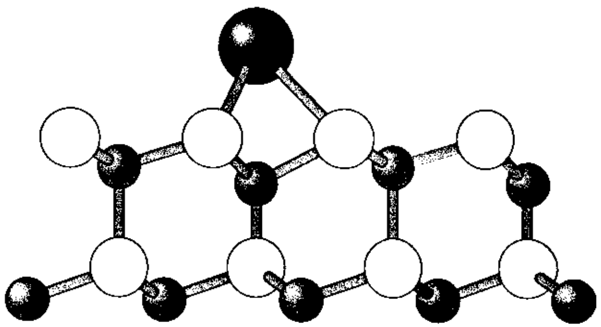


Si-Adatom

Si

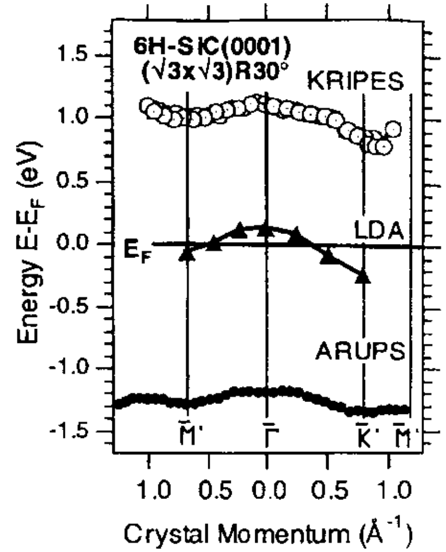
C

Seitenansicht



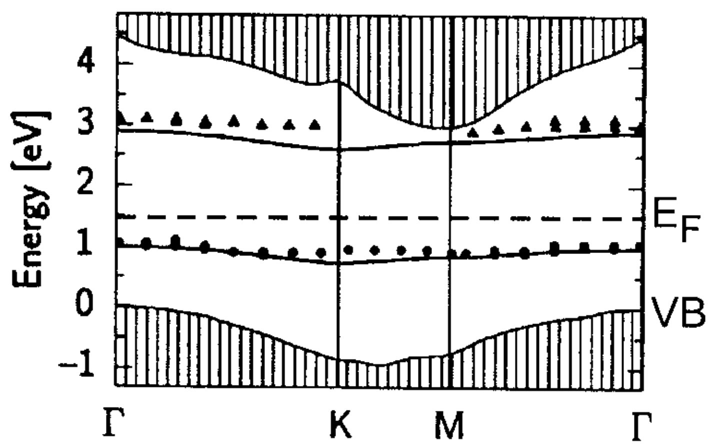
→ 'Reduktion der dangling bonds von 3 auf 1'

Diskrepanz: Theorie ↔ Experiment



Themlin et al., Europhys. Lett. 39 (1997) 61

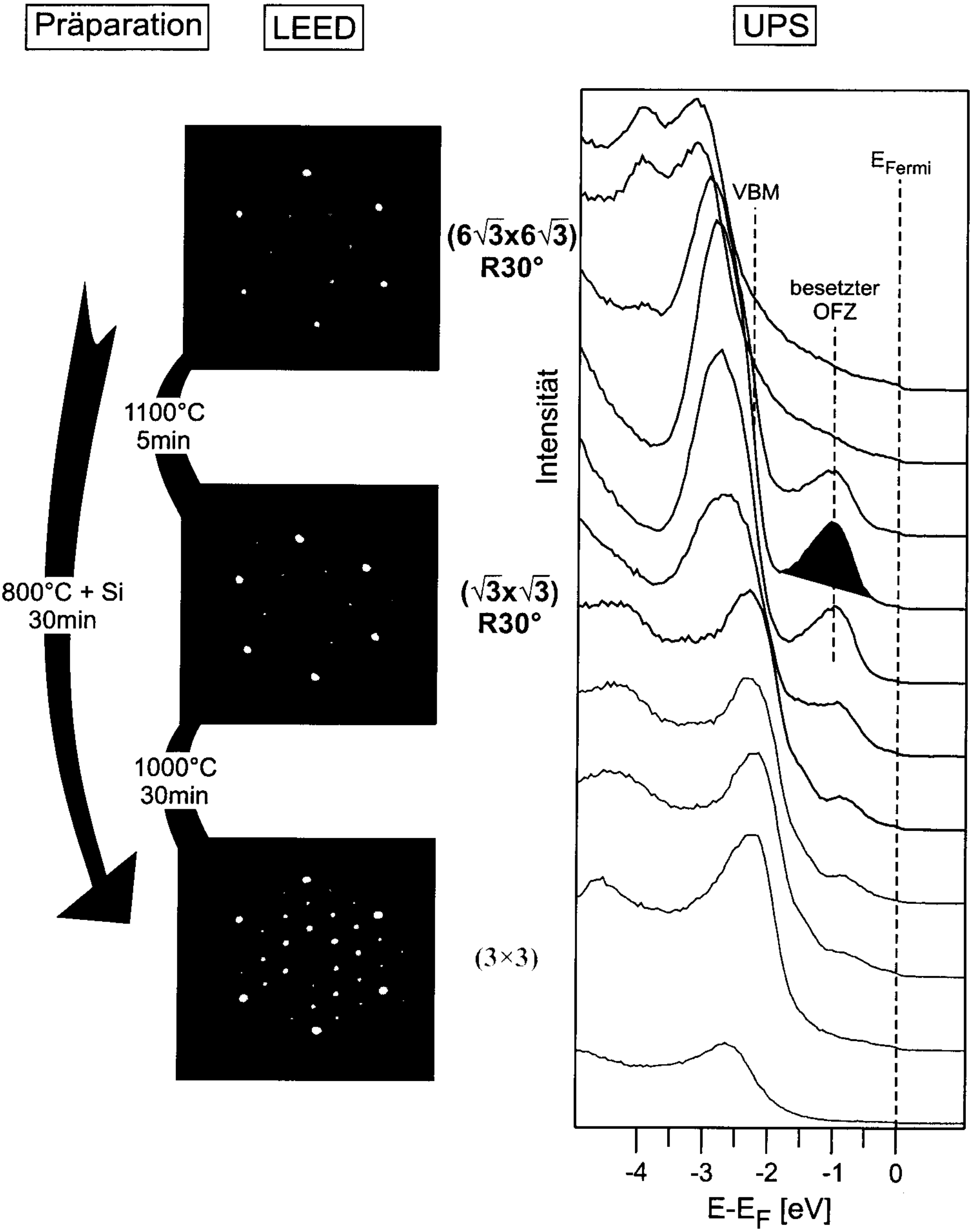
Erklärung: Mott-Hubbard-Aufspaltung

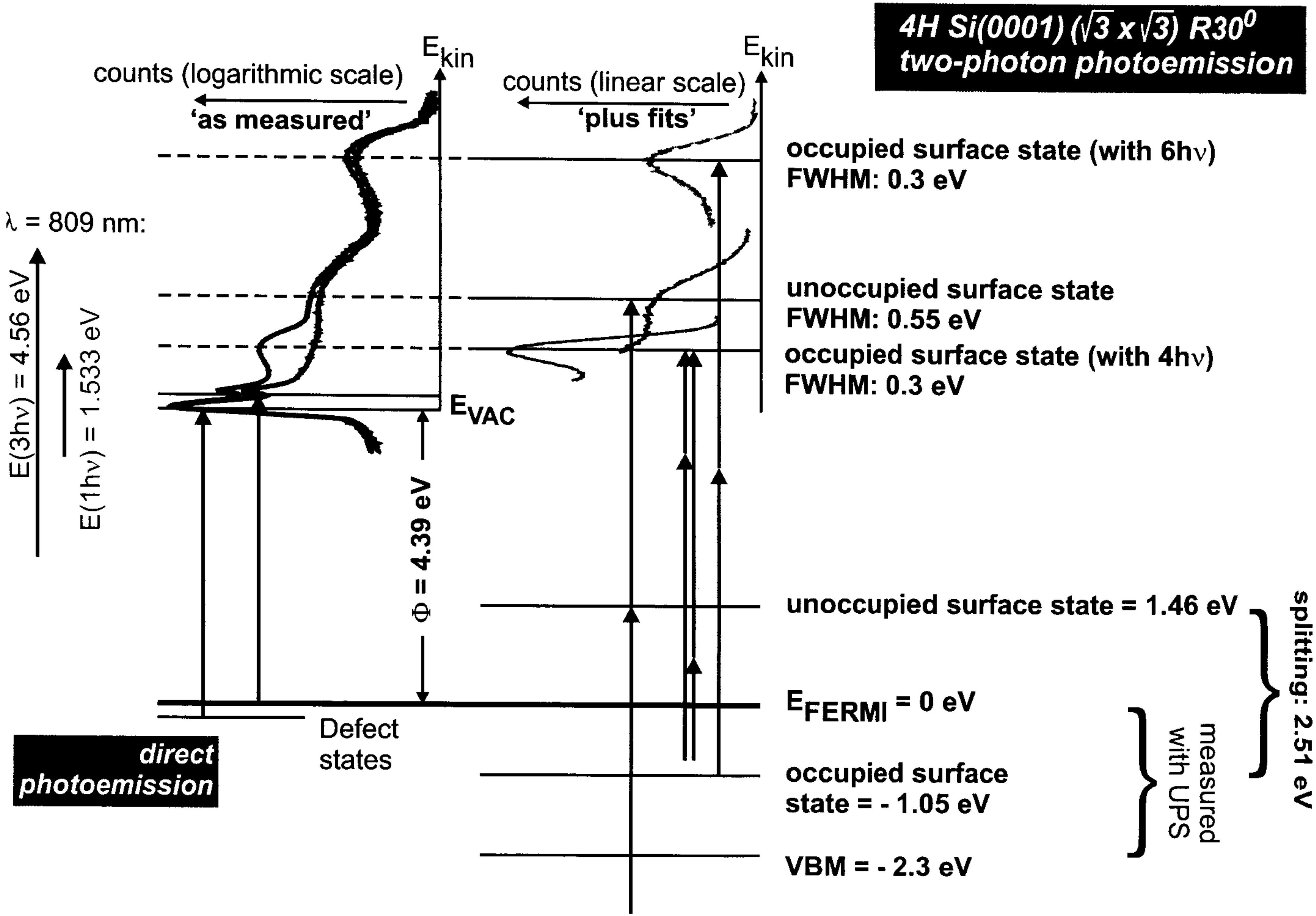


Rohlfing, Pollmann, Phys. Rev. Lett. 84 (2000) 135

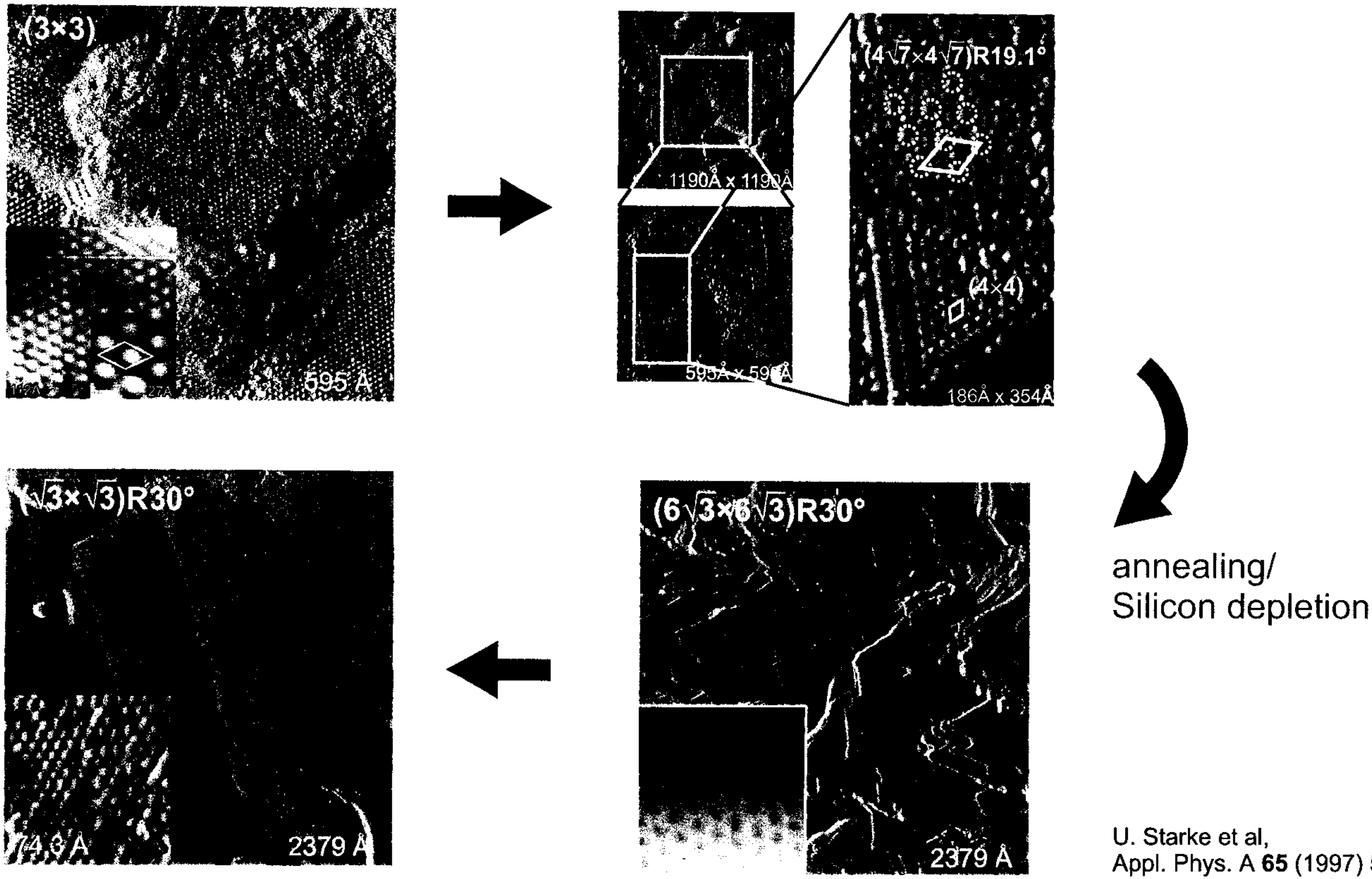
6H-SiC(0001)-($\sqrt{3}\times\sqrt{3}$)R30°

Rekonstruktionsphasen auf 4H-SiC(0001)



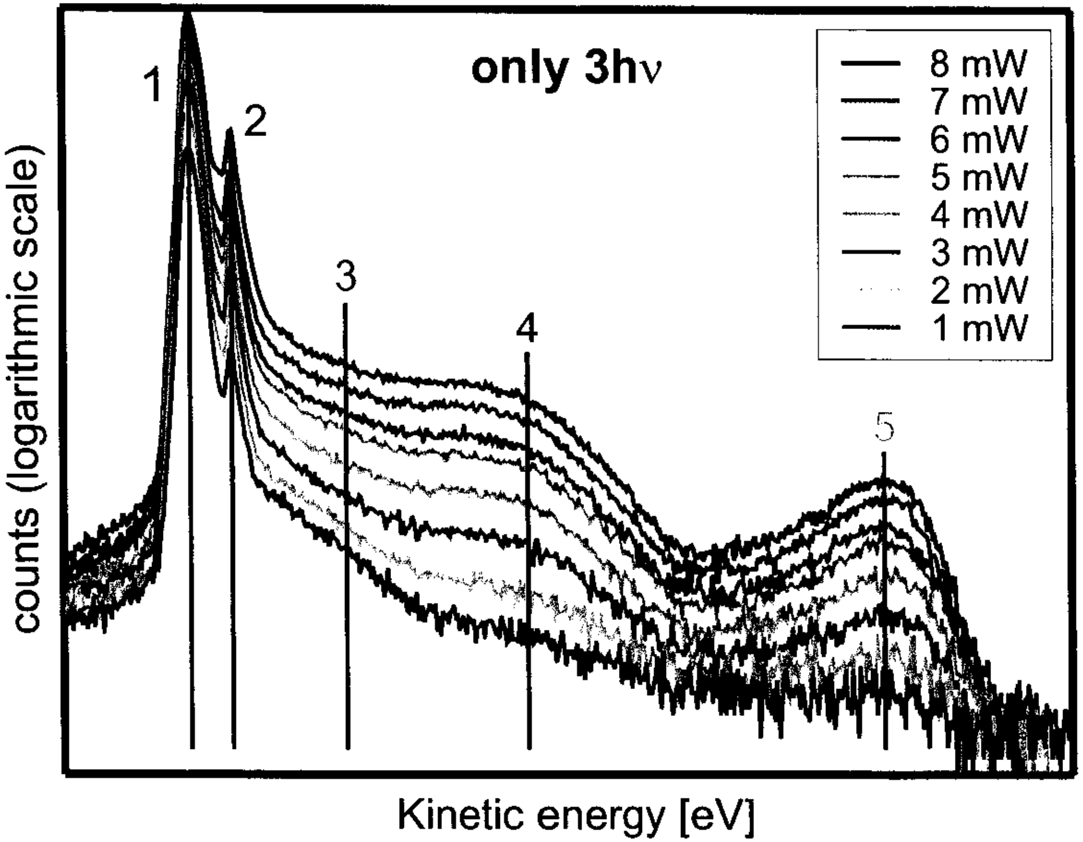

4H Si(0001) (√3 x √3) R30⁰
two-photon photoemission
counts (logarithmic scale)
'as measured'
Ekin
counts (linear scale)
'plus fits'
Ekin
occupied surface state (with 6hν)
FWHM: 0.3 eV
unoccupied surface state
FWHM: 0.55 eV
occupied surface state (with 4hν)
FWHM: 0.3 eV
λ = 809 nm:
E(3hν) = 4.56 eV
E(1hν) = 1.533 eV
EVAC
Φ = 4.39 eV
unoccupied surface state = 1.46 eV
splitting: 2.51 eV
EFERMI = 0 eV
measured
with UPS
Defect
states
direct
photoemission
occupied surface
state = - 1.05 eV
VBM = - 2.3 eV

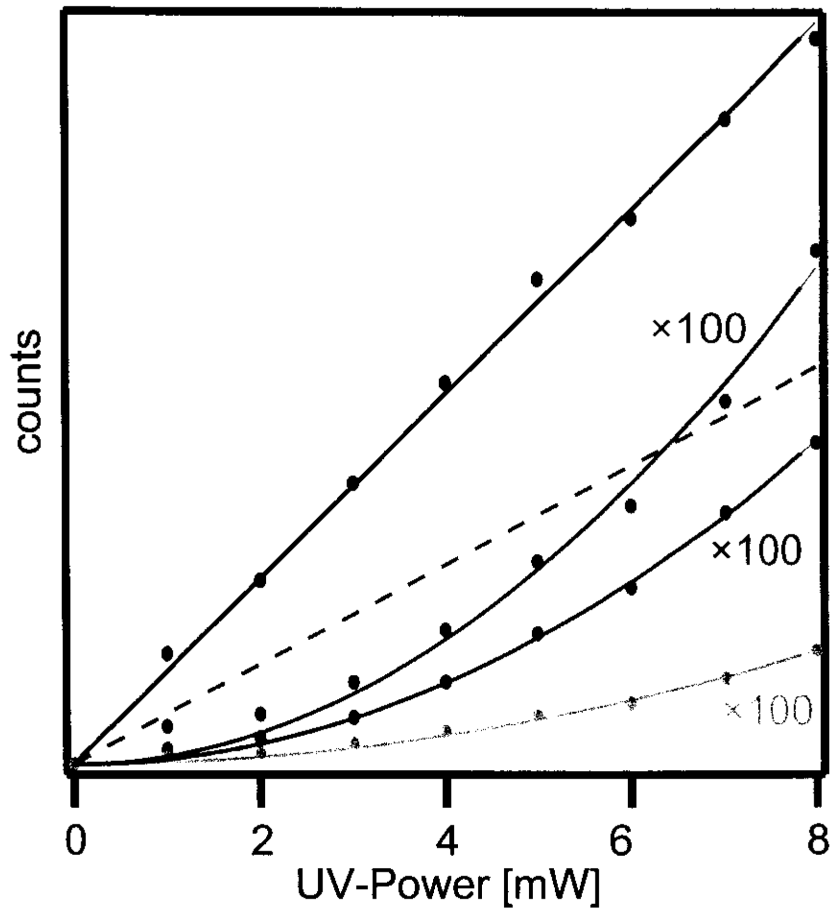
STM snapshots of SiC(0001) surface phases



U. Starke et al,
Appl. Phys. A **65** (1997) 587

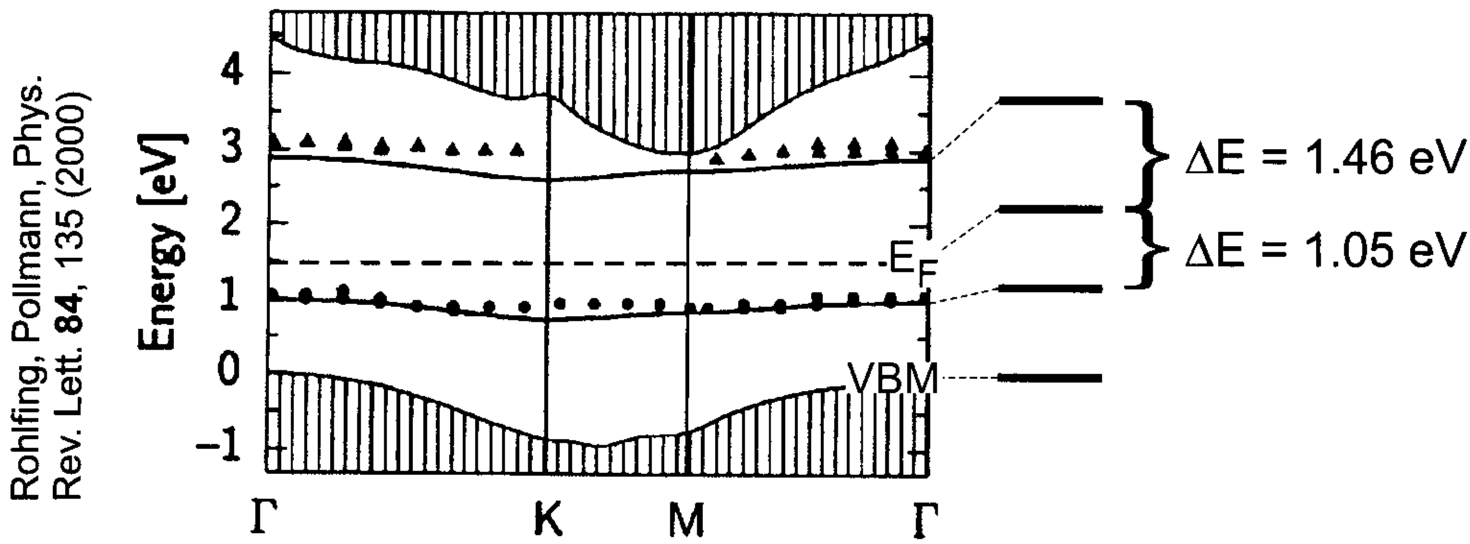
Dependence on UV power





Peak 1: linear with UV power	⇒	1PPE
Peak 3: quadratic with UV power	⇒	36 % 2PPE
Peak 3: linear with UV power Contribution of Peak 2?		64 % 1PPE
Peak 4: quadratic with UV power	⇒	2PPE
Peak 5: quadratic with UV power	⇒	2PPE

⇒ asymmetric splitting



calculation done for 6H-SiC(0001)-(√3×√3)R30° !
4H-SiC(0001): larger splitting (due to a larger bulk band gap)

Conclusions

4H-SiC(0001)-(√3×√3)R30°:

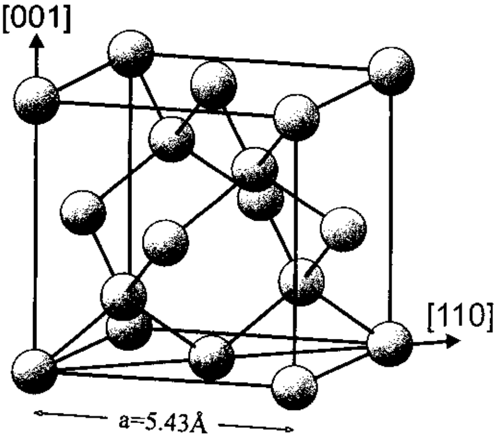
The **occupied and** the **unoccupied** surface state can be observed with ***2PPE*** **simultaneously** in **one** experiment.

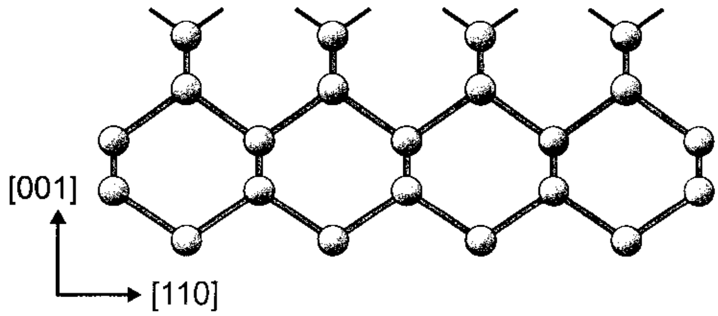
The proposed **asymmetric splitting** can be confirmed: ΔE = 2.51 eV.
→ Occupied surface state ↔ E_{FERMI}: 1.05 eV ± 0.02 eV
→ Unoccupied surface state ↔ E_{FERMI}: 1.46 eV ± 0.02 eV

6H-SiC(0001)-(√3×√3)R30°:

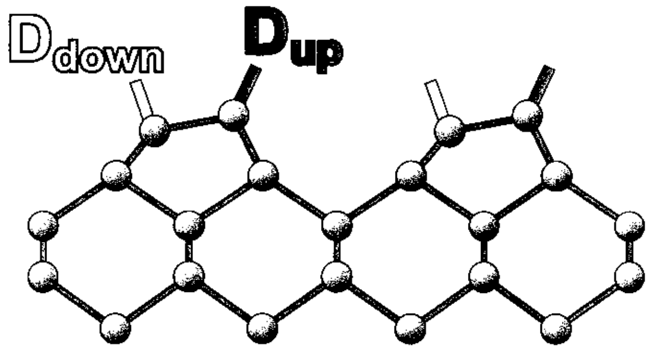
A **smaller splitting** can be observed: ΔE = 2.34 eV.
This can occur due to the smaller bulk band gap.

Si(001): Geometric structure

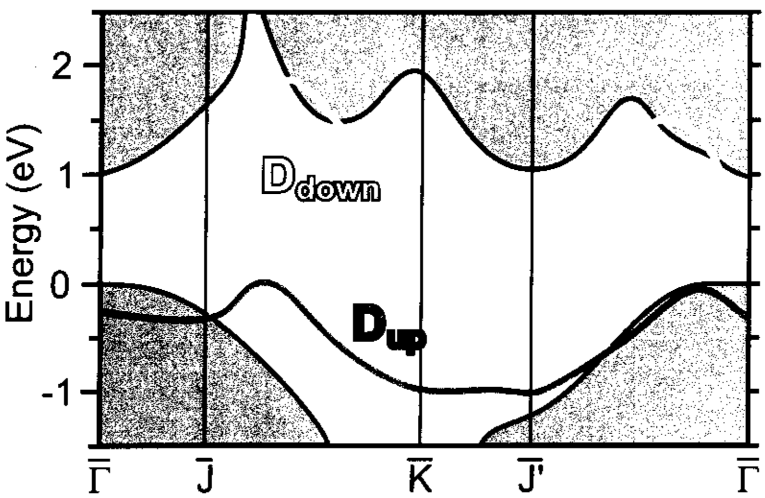




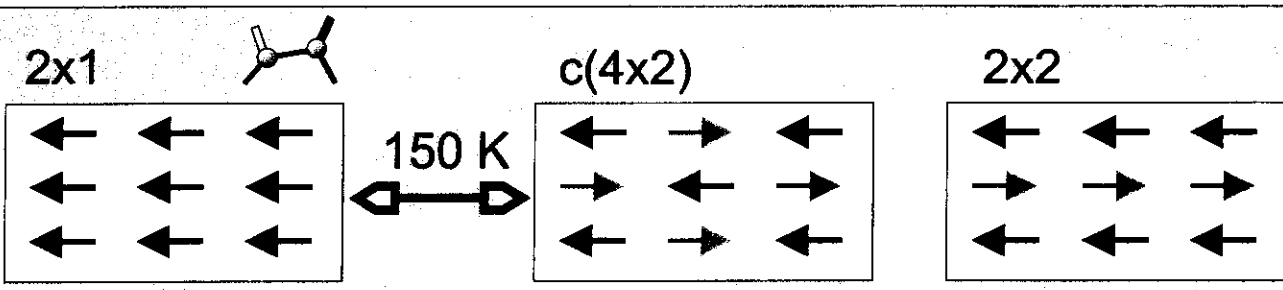
Surface reconstruction
Si(001) 2×1 :



Asymmetric Dimers ⇒ occupied D_{up} & unoccupied D_{down} surface band

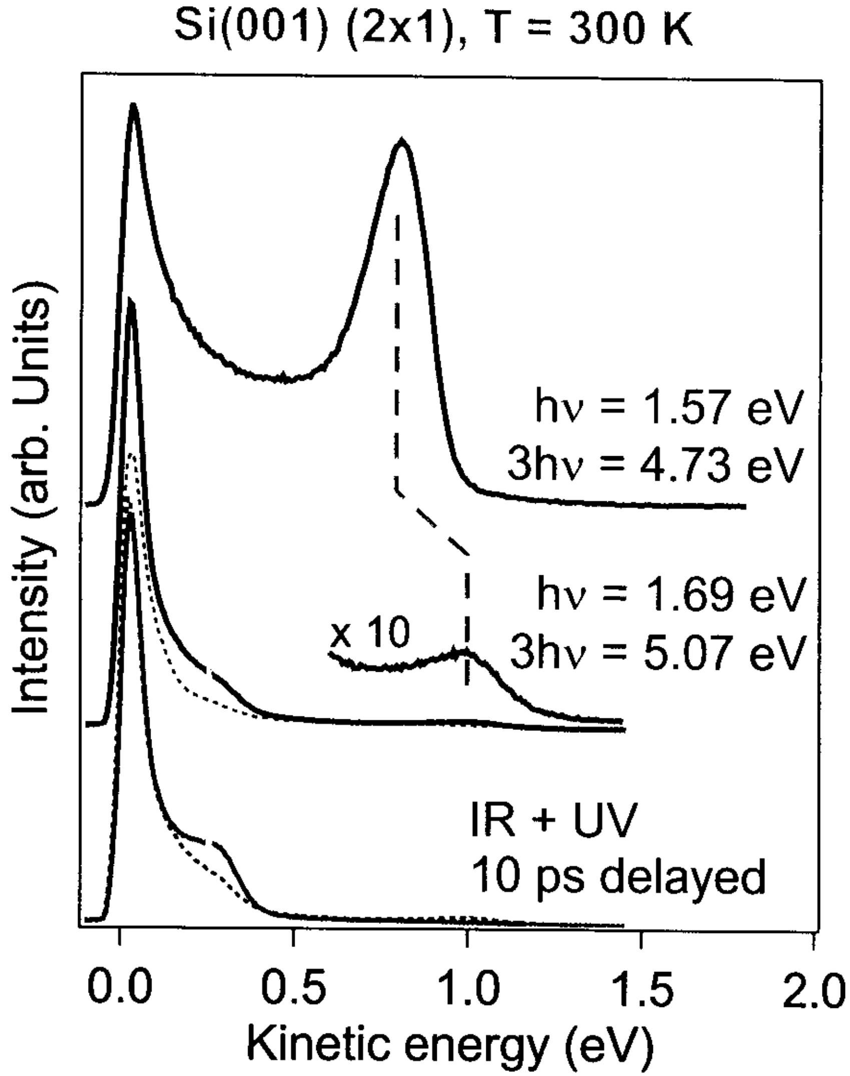


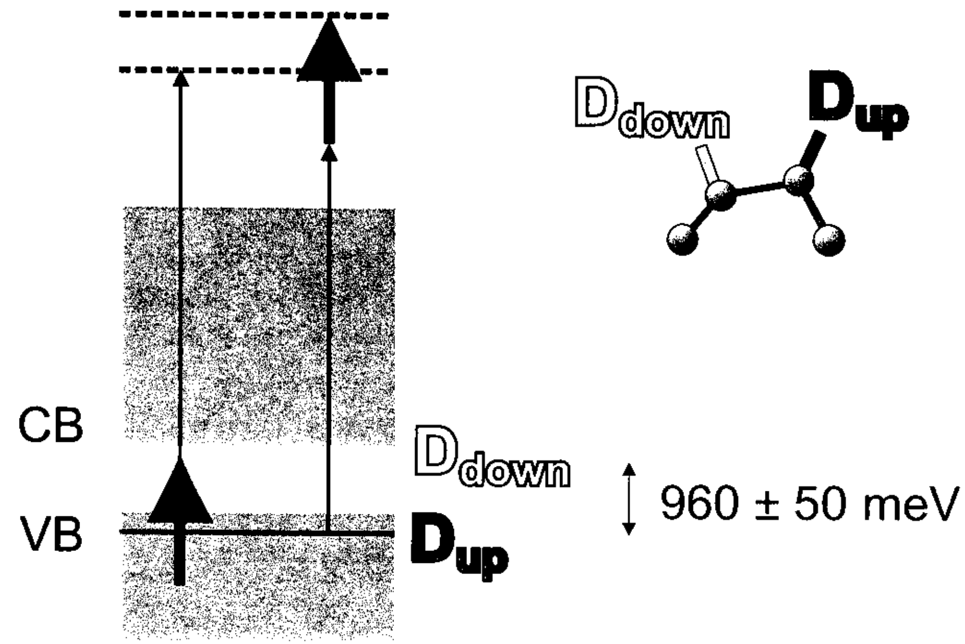
J. Pollmann et al., Appl. Surf. Sci. **104/105**, 1 (1996)



↳ *W. Mönch, Semiconductor Surfaces and Interfaces (1993)*

Si(001) Two-photon photoemission





⇨ IR + UV excitation of occupied D_{up}

energy relative to valence-band maximum:

D_{up} = -260 meV

D_{down} = 700 meV

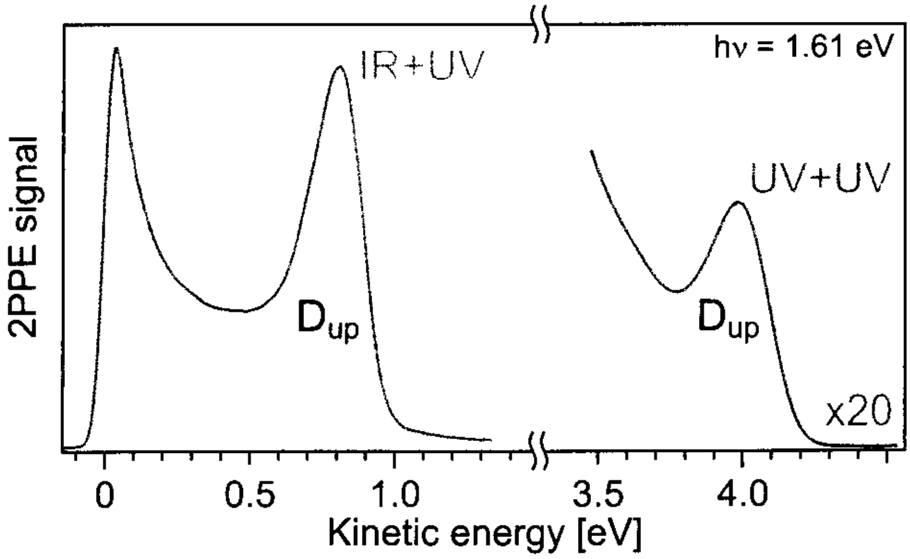
good agreement with UPS (1), IPS (2) and GWA calculations (3)

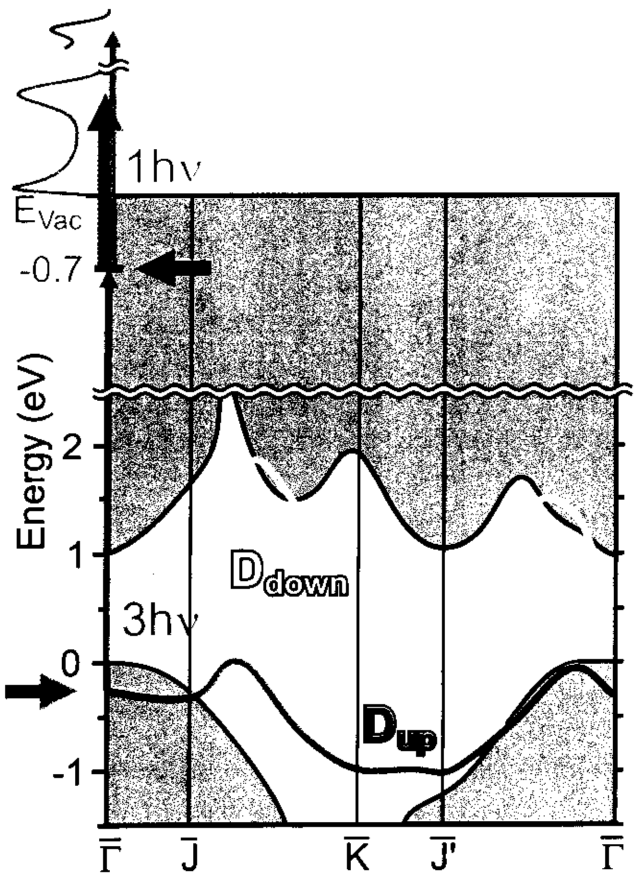
(1) R. I. G. Uhrberg, PRB 24 (1981) 4684.
L. S. O. Johansson PRB 42 (1990) 1305.

(2) J. E. Ortega and F. J. Himpsel, PRB 47 (1992) 2130
L. S. O. Johansson and B. Reihl,
Surf. Sci. 269/270 (1992) 810

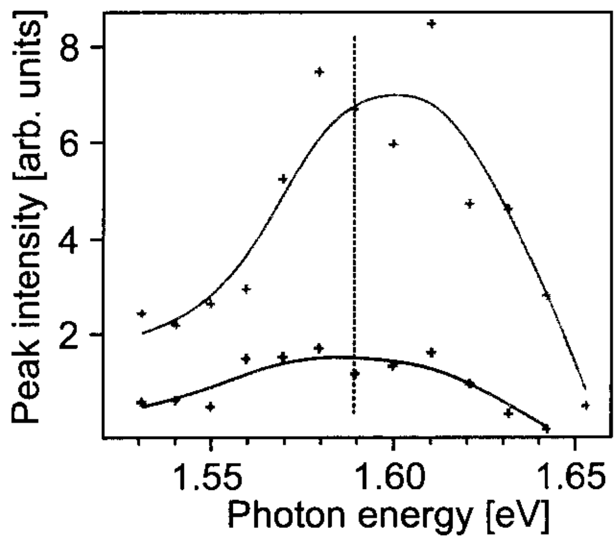
(3) J. Pollmann et al.,
Apl. Surf. Sci. 104/105(1996)1.

Si(001): Occupied D_{up} state





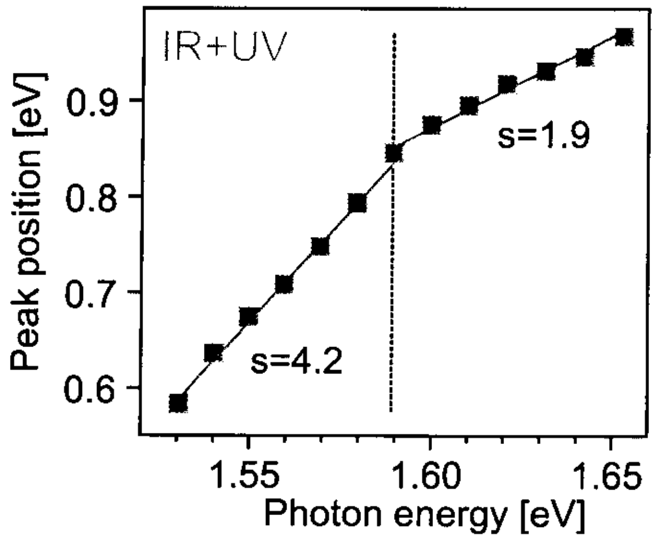
Resonance:

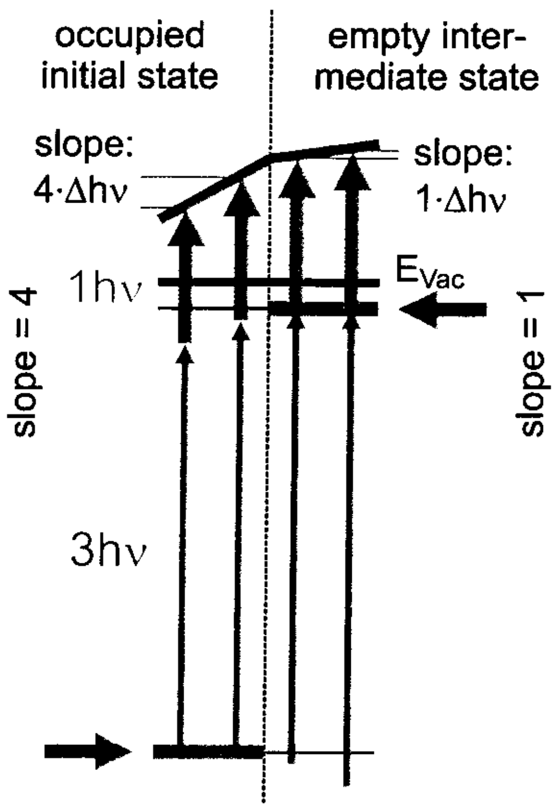


⇒ Occupied D_{up} SS

⇒ Resonance with unoccupied SS approx. 0.7 eV below E_{Vac}

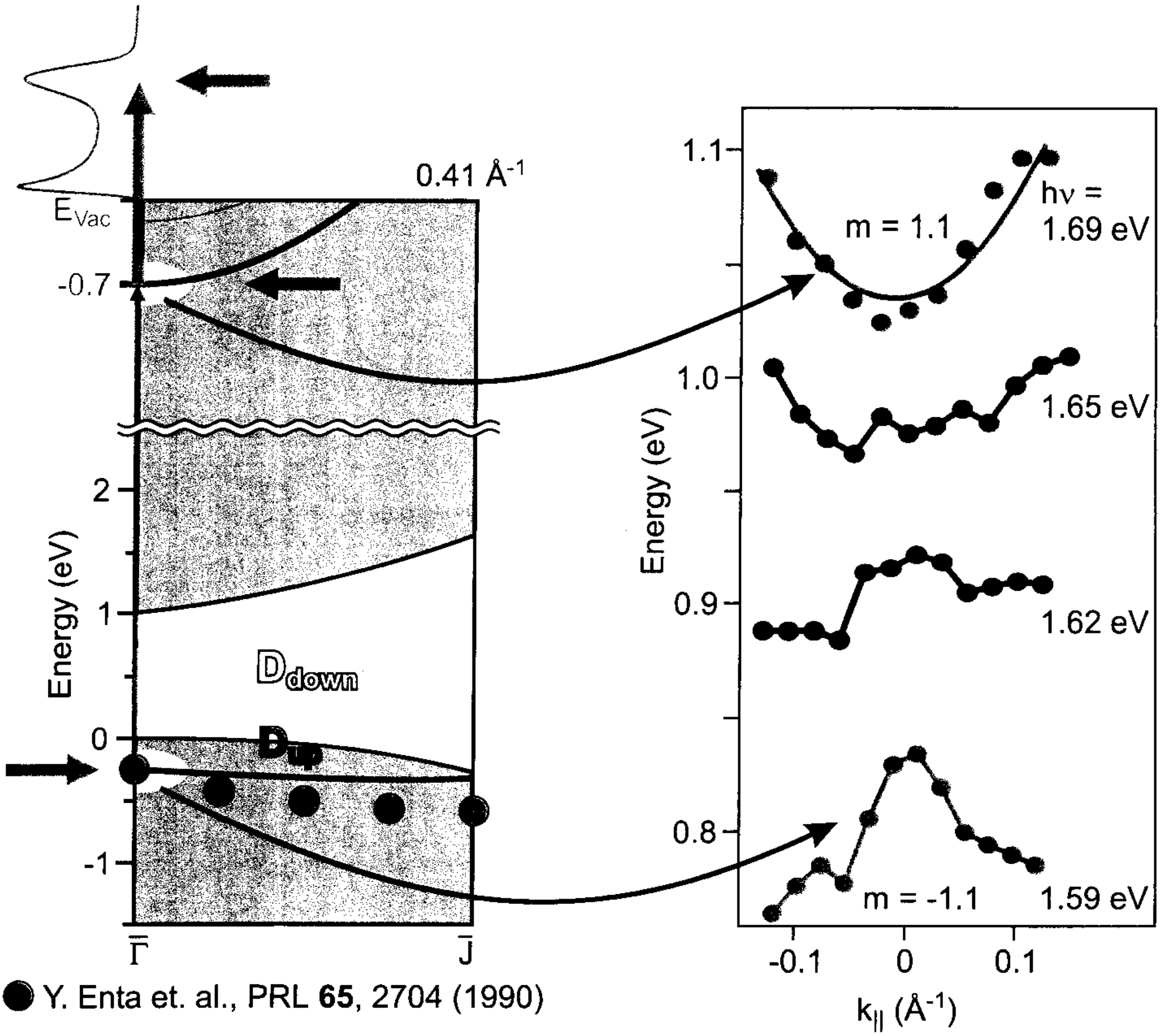
Si(001): Unoccupied state





⇒ Resonance between occupied (D_{up}) and unoccupied (0.7 eV below E_{Vac}) surface state

Si(001): Dispersion & lifetime of the resonance



● Y. Enta et. al., PRL **65**, 2704 (1990)

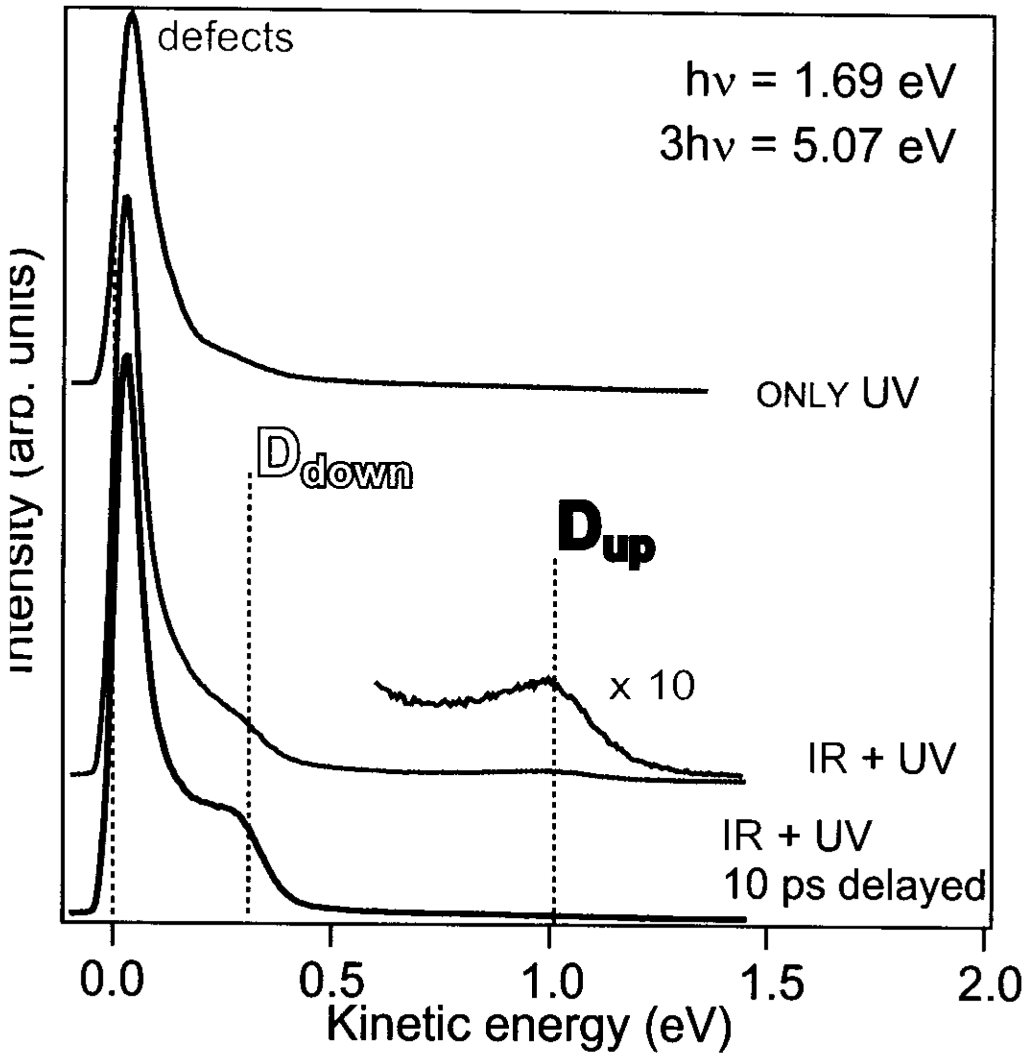
⇒ Is unoccupied SS an image-potential state?
$\varepsilon_{surface}$(Si 001) = 12...13

$$\rightarrow \quad E_{bind} = \frac{0.85\,eV}{n^2} \cdot \left(\frac{\varepsilon - 1}{\varepsilon + 1}\right)^2 = 0.6\,eV$$

⇒ Lifetime of unoccupied state: 10 ± 5 fs

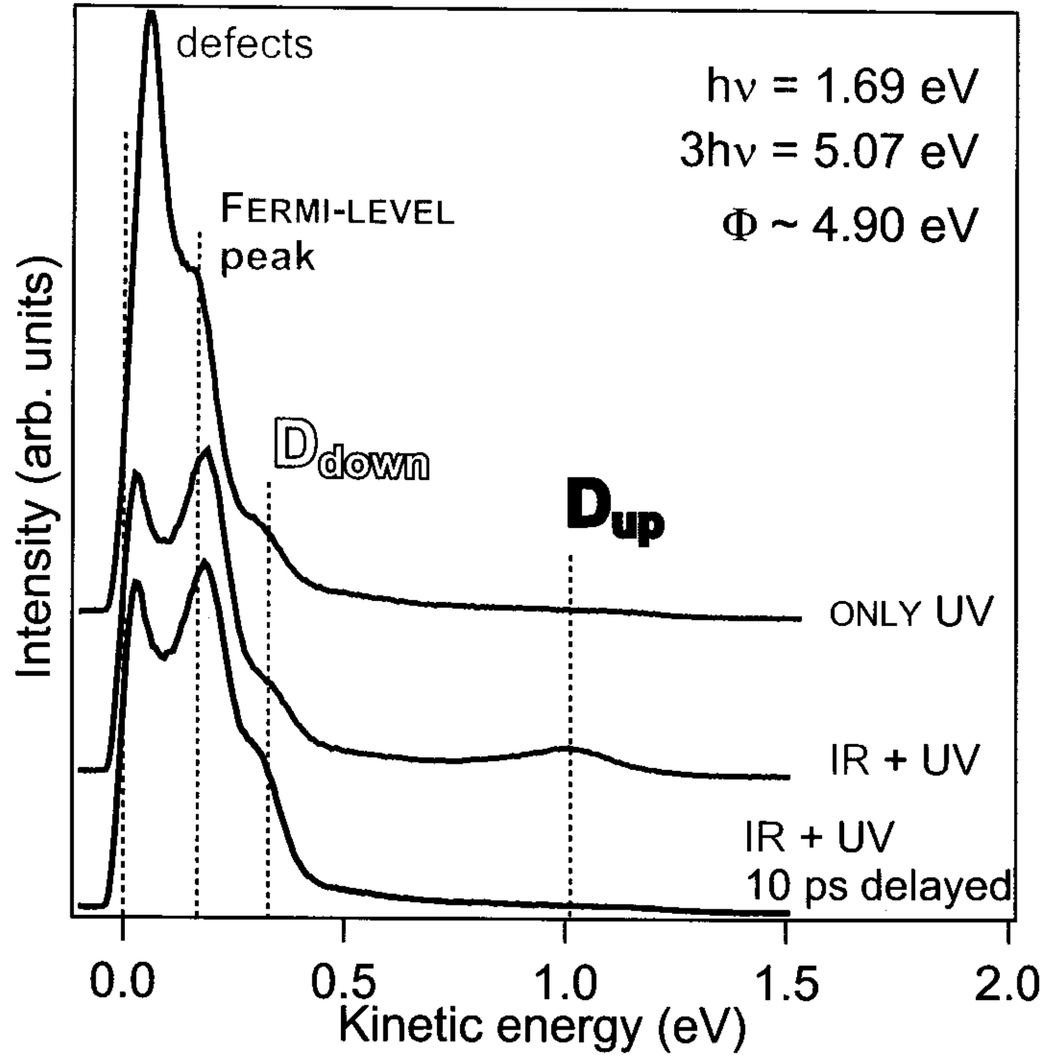
Si(001) Two-photon photoemission

Si(001) (2x1), T = 300 K



- IR heating of direct photoemission
- "no" Fermi-level peak

Si(001) c(4x2), T = 150 K

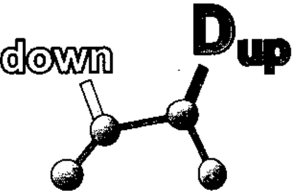


- IR depletion of defect states
- strong Fermi-level peak

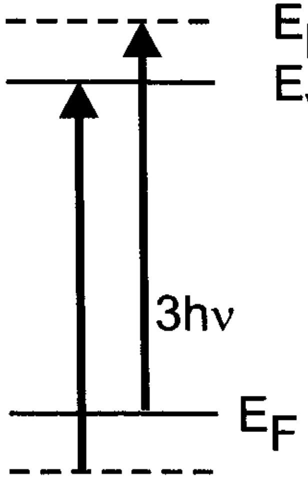
- IR + UV excitation of occupied D_{up}

Direct vs. two-photon photoemission UV-UV

Si(001) D_{down} D_{up}



direct photoemission

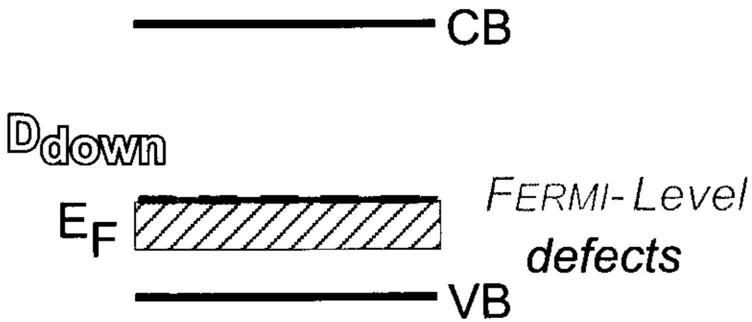


$E_{kin} = 3h\nu - \Phi$

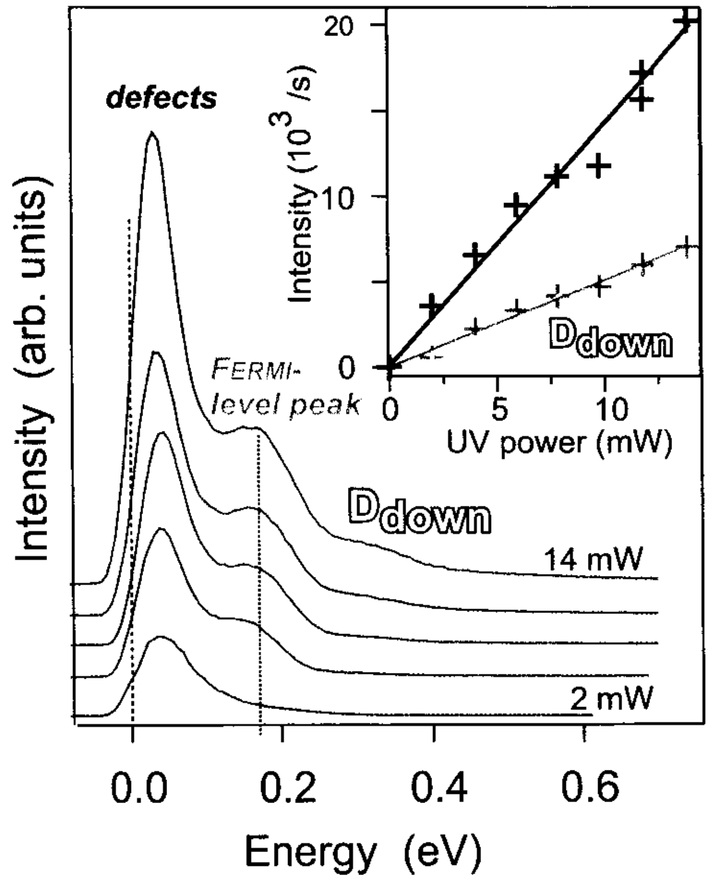
E_{vac}, $E_{kin} = 0$ eV

1ppe ~ I

2ppe



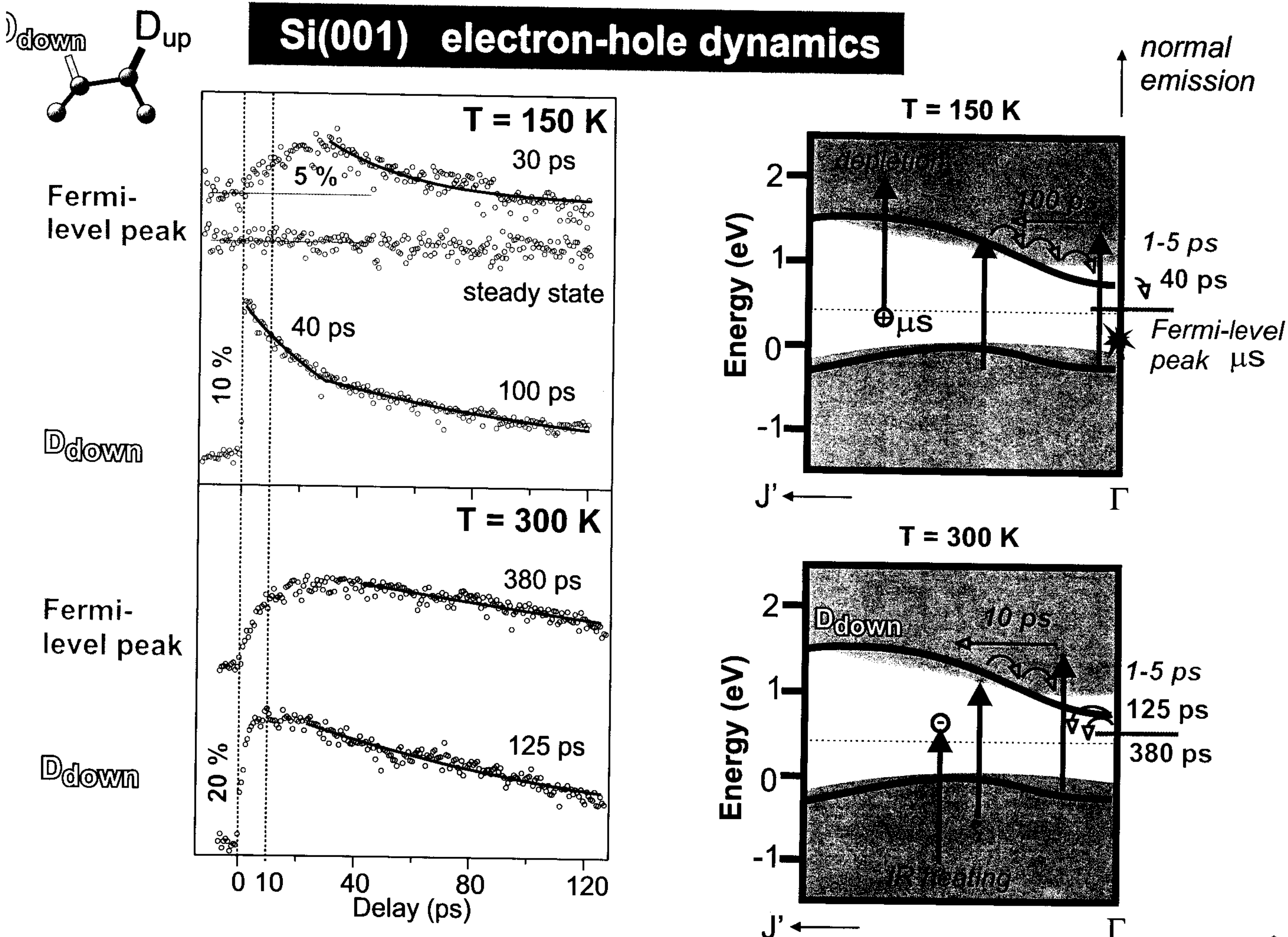
T = 120 K, c(4x2)



Fermi-level peak: direct photoemission

surface defects in band gap ~$10^{11}/cm^2$

Si(001) electron-hole dynamics

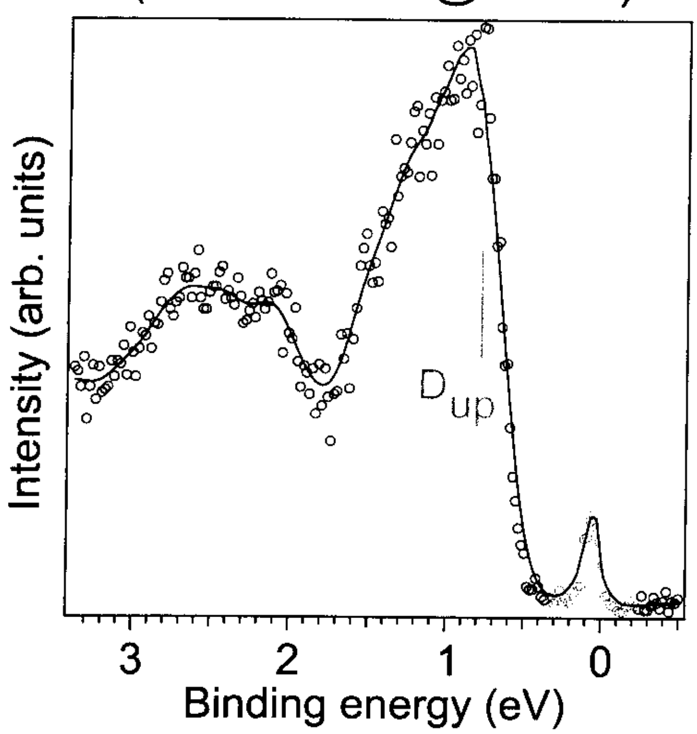


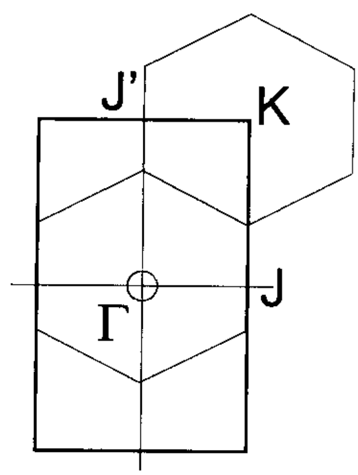
Si(001)

- D_{up}, D_{down} identified in two-photon photoemission
- Surface-state gap at $\bar{\Gamma}$: 960 ± 50 meV
- Electron & hole dynamics: ps for T = 300K
  ~ μs T = 150K

What is the Fermi-level peak?

(seen in UPS @ 120 K)





2ppe: energy 0.15 eV below D_{down}

Theory: D_{down} (2x1) vs. c(4x2): 0.35 eV (GWA) [1]
0..56 eV (LDA) [2]

[1] J. E. Northrup, PRB 47 (1993) 10032.

[2] P. Krüger, priv. Commun.

Conclusions

- Linewidth analysis (large delay)

  ~ Im { Lorentzian(pure dephasing $2\Gamma^*$) $\otimes$

  | **F**[pulse-envelope x exp($-\Gamma/2$ t)] $|^2$}

- 4H - SiC(0001)

  Mott-Hubbard splitting
  of dangling-bond states is asymmetric
  and dependent on band gap (6H)

  lifetime measurements failed - defects ?

- Si(001)

  π and π^* - dangling bond states
  electron dynamics and recombination
  Fermi-level peak ?

  T = 300 K
  ps - time scale, phonon assisted recombination
  T = 150 K
  μs - time scale, radiative recombination