Semiconductors in presence of gases: The relationship between charge transfer and charge transport

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Outline

- Motivation

- Classic semicondutor theory
  - Ideal „Bulk“
- Semiconductor theory at the bulk edge
  - Semiconductor surface-gas interactions

- One example of device operation
  - Electro-adsorptive effect
  - Thin film transistor
  - Drift of vacancies (dopants)

- Remarks

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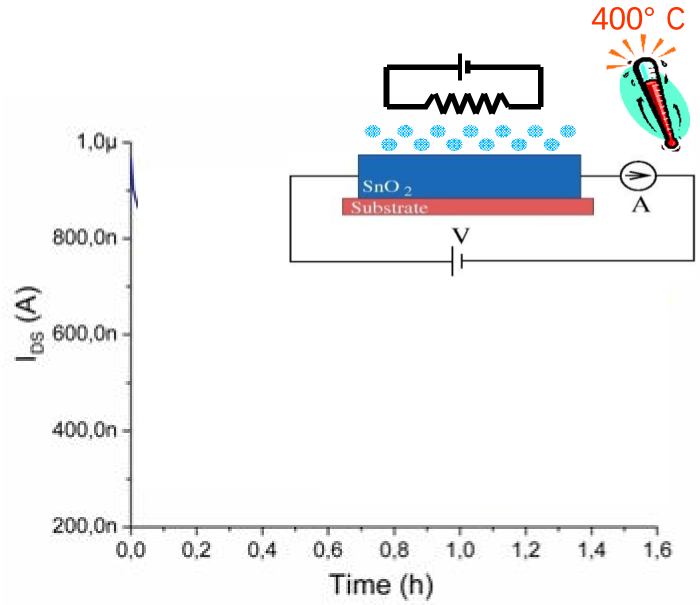
Motivation



- Change in the electron transport in semiconductors in presence of gases (Thin films)
  - ➢ Resistance is a macro effect, however it is affected by atomic scale interactions
  - ➢ The interaction at the solid-gas interface influences the bulk properties. Is the other way around also true?

Catalysis on semiconductors

- Why are important the semicondutors in catalysis?
  - In most case, metals are enclosed in a semiconductor coat.
  - Reactions take place at the surface, on the semiconductor.
- The electron Theory of Catalysis on Semiconductors*.
  - Heterogeneus catalytic process are based on electronic mechanisms.
  - Elucidate the relationship between catalytic process and electronic properties of semiconductors.
- Theory limits
  - Fully applicable to dielectrics.
  - Cannot be applied directly to metals.
    - ⌘Based on the "many-electron" aproach.

*F. F. Wolkenstein, W. B. Sandomirski, Dokl. Acad. Nauk. SSSR, 118, pp. 980-982, 1958.

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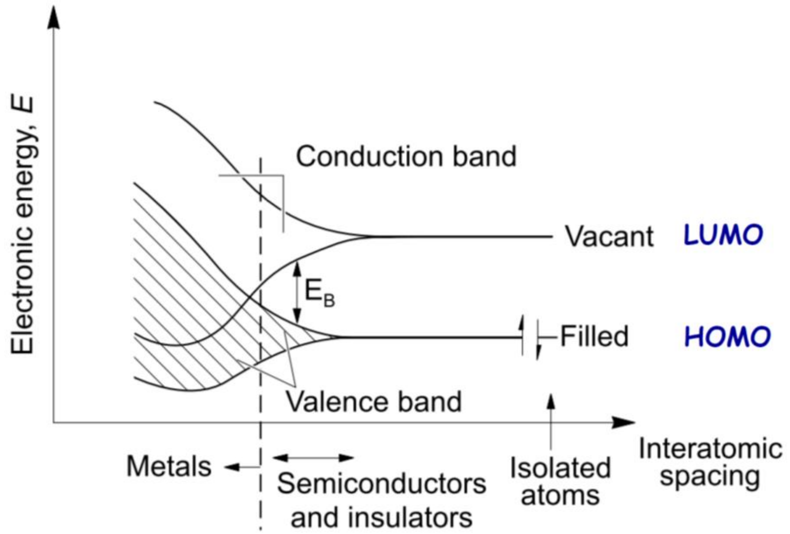
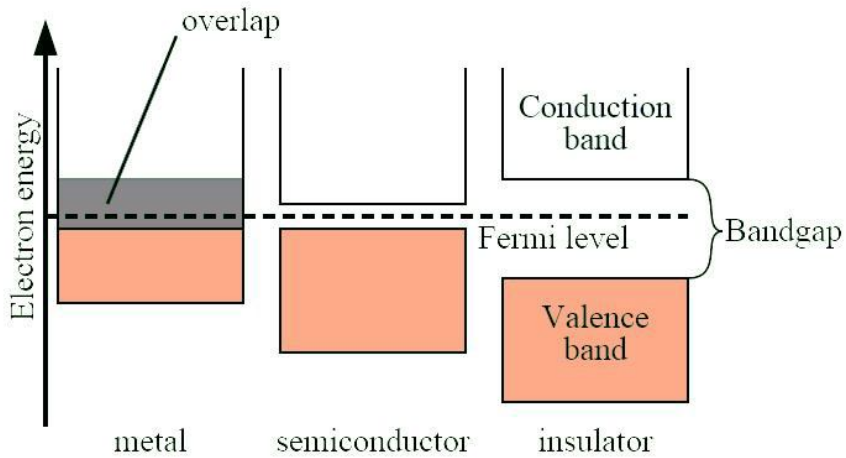
Semiconductors

- Is a material with an electrical conductivity between metal and insulator
  - Conductivity can be modified by different parameters:
    - ⌘Temperature
    - ⌘Electric field
  - Two types of carriers: electrons and holes





Physics of solids

- Weakly bound valence electrons interact with positively charged atomic cores
  - Schödinger equations rules the motion of electrons in solids
    - ⌘Charge doesn't change with time

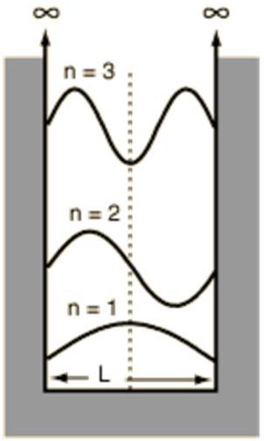
$$E\,\psi(\mathbf{r}) = (-\hbar^2/2m)\,\nabla^2\,\psi(\mathbf{r}) + U(\mathbf{r})\psi(\mathbf{r})$$

    - ⌘Equipotential U(**r**)=0

⌘ $E\psi(x) + \frac{\hbar^2}{2m}\frac{d^2\psi}{dx^2} = 0$ solution Ψ(0)=Ψ(L)=0 $\psi_n(x) = A\sin\left(\frac{\pi n}{L}x\right)$

Then we obtain as Eigenvalue:

$$E_n = \frac{\hbar^2}{2m}\left(\frac{\pi n}{L}\right)^2$$



x = 0 at left wall of box.

Fermi distribution (U(r)=0)

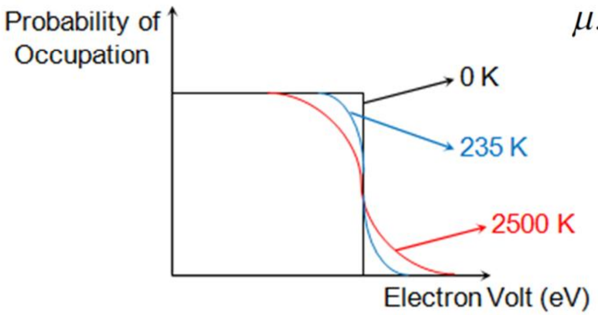
- For N valence electrons, highest occupied level energy

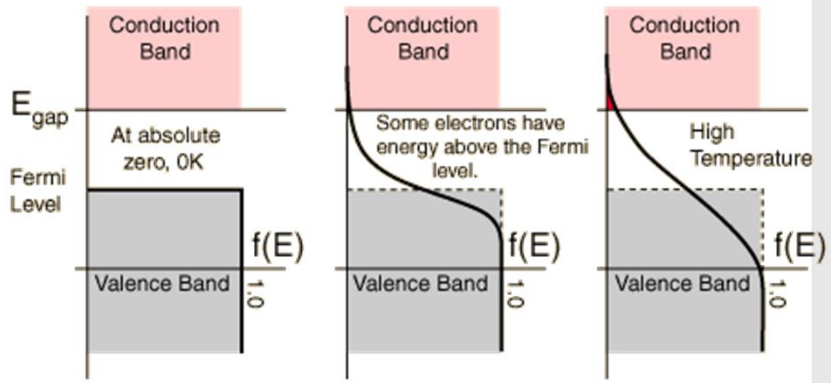
Fermi energy $$E_F = \frac{\hbar^2}{2m}\left(\frac{\pi N}{2L}\right)^2$$

- KE of electrons increases with temperature: distribution function

$$f(E) = \frac{1}{e^{(E-\mu)/k_B T} + 1}$$

E...Energy
k_B...Boltzmann constant
T...Temperature
μ...(Electro-)Chemical potential

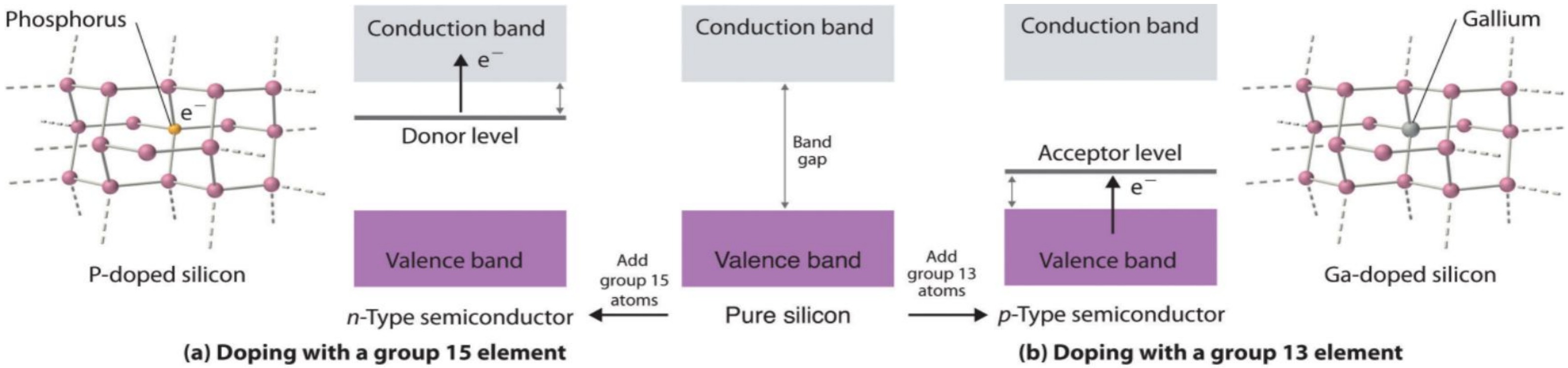




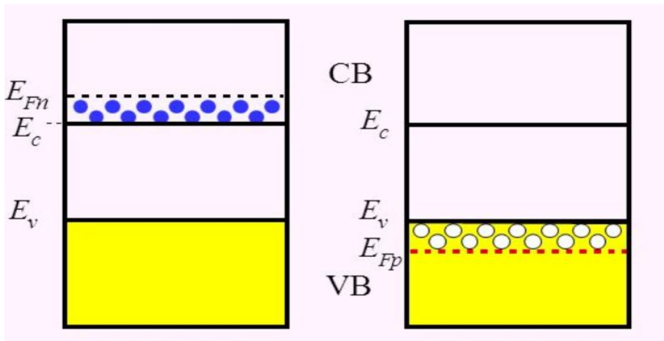
No electrons can be above the valence band at 0K, since none have energy above the Fermi level and there are no available energy states in the band gap.

At high temperatures, some electrons can reach the conduction band and contribute to electric current.

Types of semiconductors



- Intrinsic
- Extrinsic
  - Type n
  - Type p
- Degenerate:
  - $|E_C - E_D| < 3K_BT$
  - $|E_V - E_A| < 3K_BT$



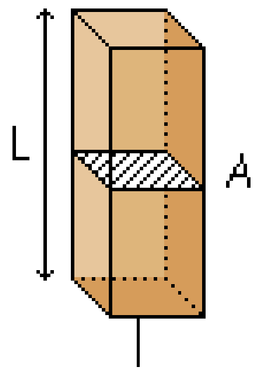
Conductivity in semiconductors

- Intrinsic carriers

$$n_i = \int_{E_C}^{\infty} f(E)g(E)dE \qquad n_i = N_C e^{-(E_C - E_F)/k_B T} \qquad n_i = N_V e^{-(E_F - E_V)/k_B T}$$

- Extrinsic carriers
  - Electrons $$n = \frac{N_D}{e^{(E_C - E_D)/k_B T} + 1} \approx N_D e^{-(E_C - E_D)/k_B T}$$
  - Electron holes $$p = \frac{N_A}{e^{(E_A - E_V)/k_B T} + 1} \approx N_A e^{-(E_A - E_V)/k_B T}$$
- Resistance: $R = \frac{\rho \ell}{A}$

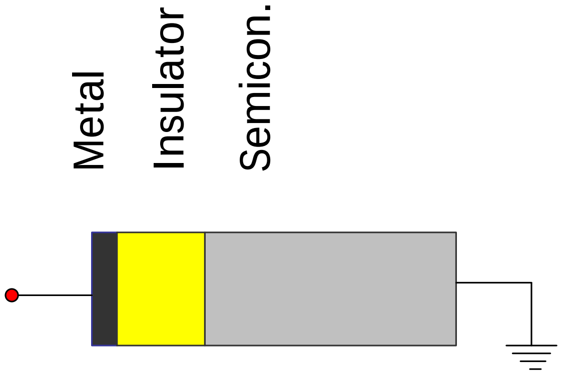
Thermo-ionic description!!

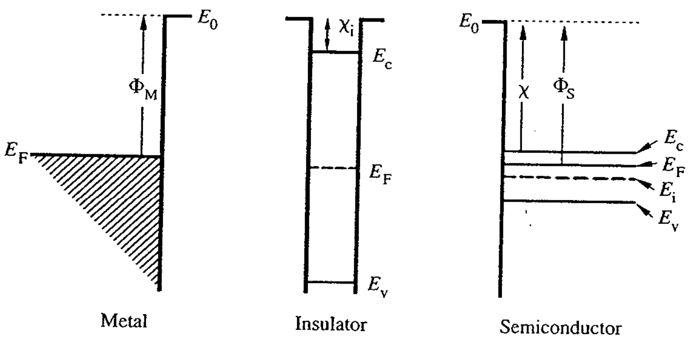


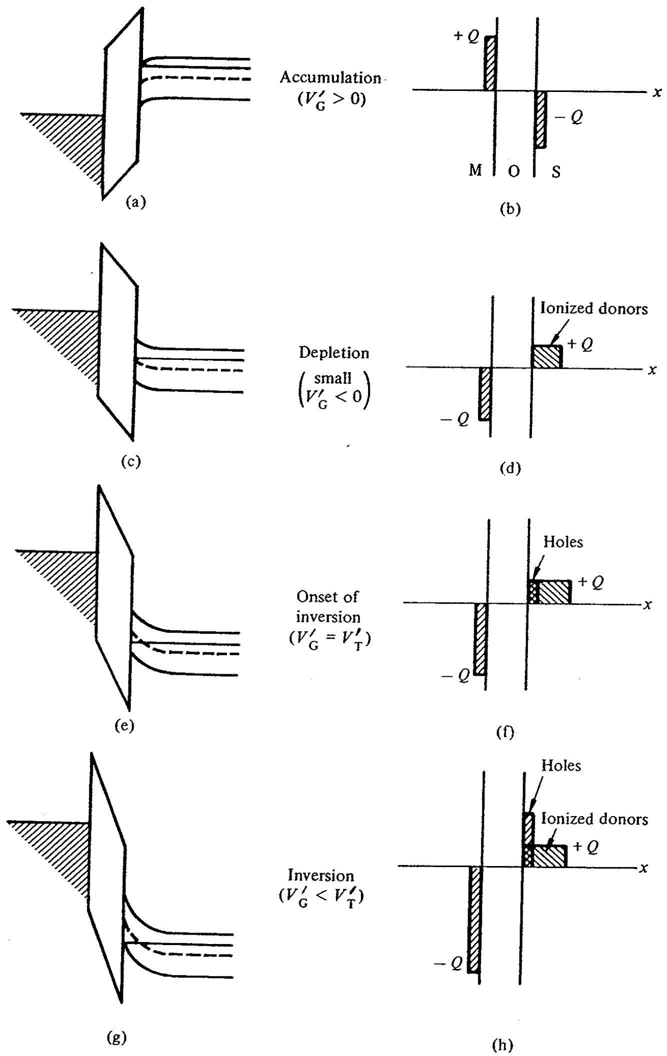
  - Resitivity

    $\rho = 1/\sigma$
  - Conductivity $\sigma = q(\mu_p p - \mu_n n)$ $\qquad \mu_n k_B T = q D_n$
- Current density $$J = qn\mu E + qD\frac{dn}{dx}$$

Band bending (MOS)

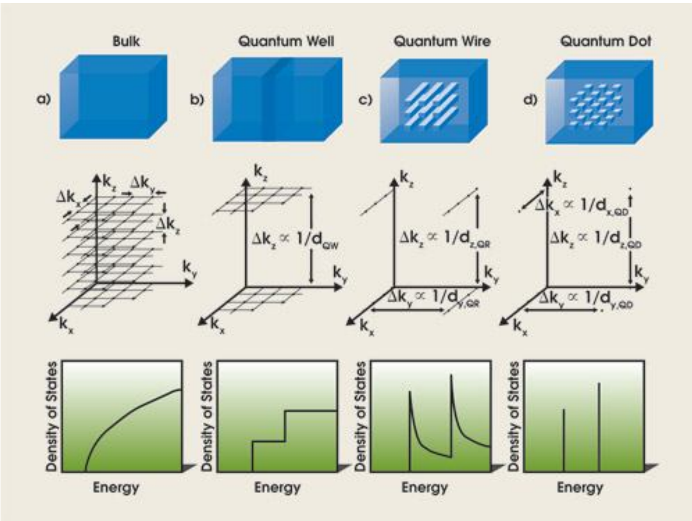


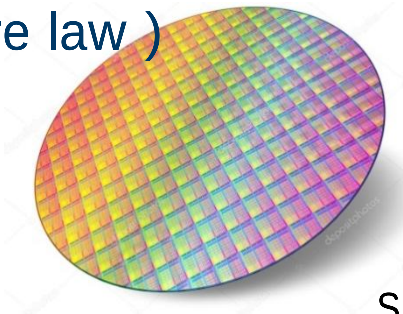


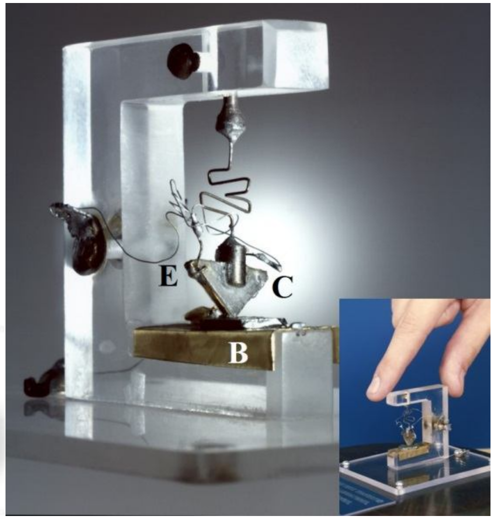


Summary

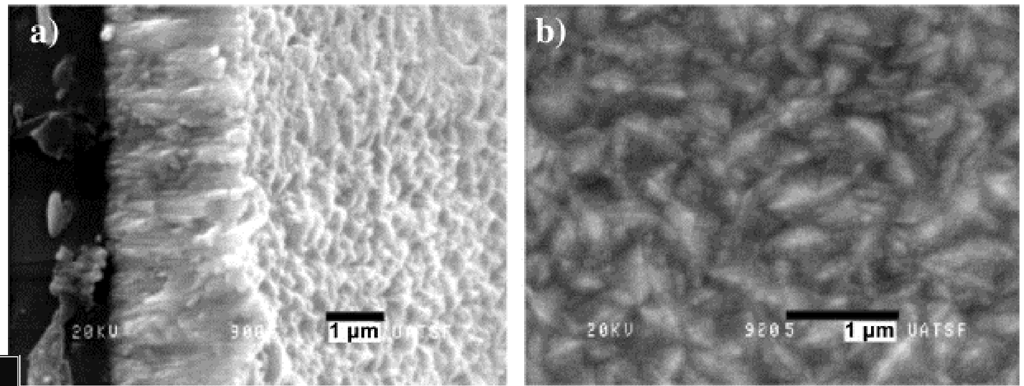
- Semiconductor technology is everywhere
  - ➢ Scaling-down Close to the quantum limit
    - ⌘L_D/2: quantum confinement:
      - ⌧5n node (end of Moore law)







Shockley, Bardeen, Brattain 1948



Thin Solid Films Volume 531, 15 March 2013, Pages 172-178

- ➢ Defects grain domains
- ➢ Surface??

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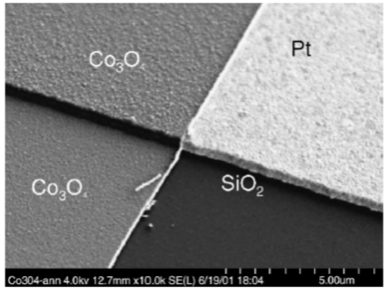
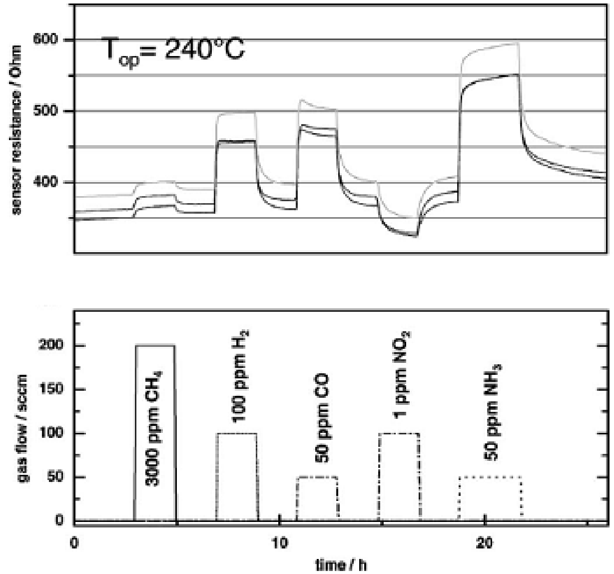
Surface physics

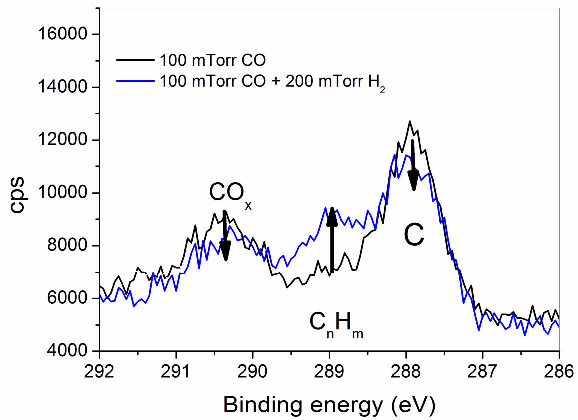
- Thin film Co_3O_4
  - Charge transfer information
    - ⌘Macro-scale



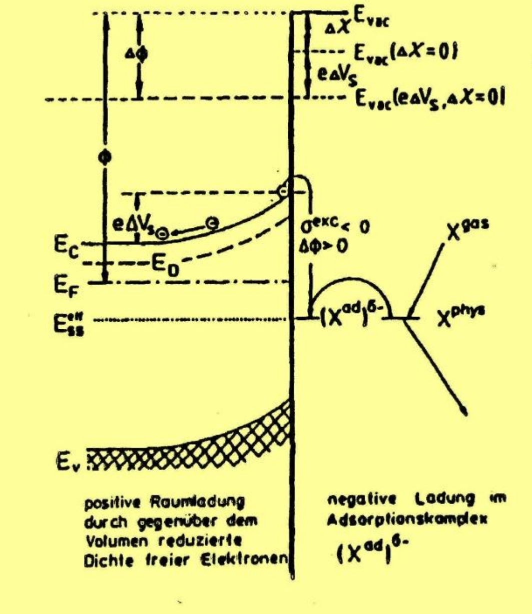


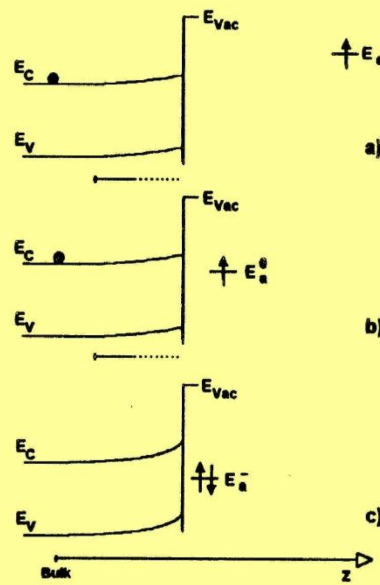
*J. Wöllenstein, Sensors and Actuators B 93 (2003) 442–448

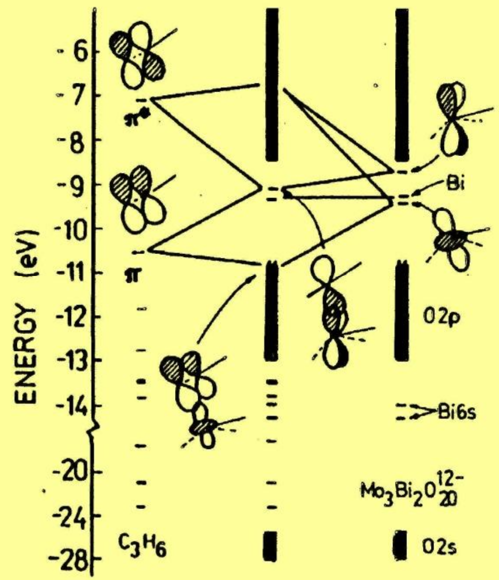
- Cobalt thin film 240°C
  - Chemical information
    - ⌘Atomic scale



Links to Theory







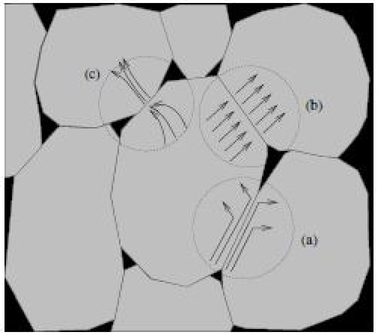
Charge Transfer Model:
Needs Electron Transfer
Band Bending
Thermodynamics

Wolkenstein Model:
Fully Quantum Mechanical
Easy Access to E_F,
Semiconductor Device
Modeling

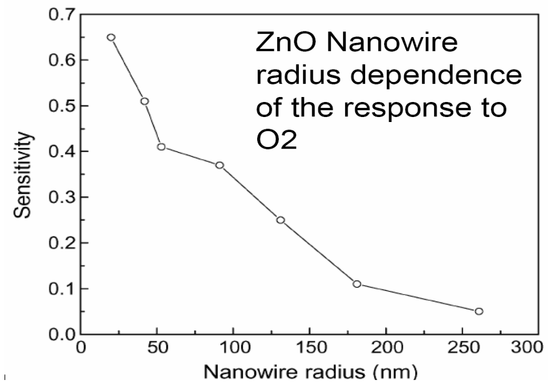
LCAO:
Quantum Chemistry
d, p and s orbitals
Overlap
Stereochemistry
Some Access to EF

Linear combination of atomic orbitals

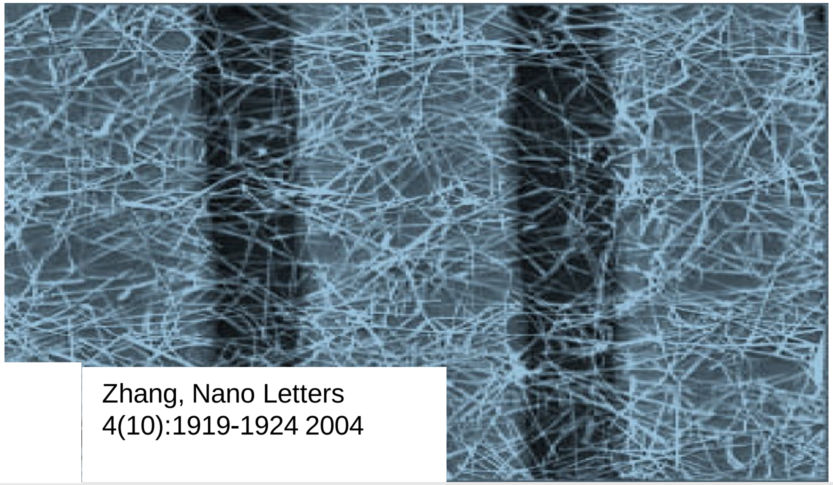
- It can not describe macro effects:
  - Different particle size
  - Or tortuosity



$$E\,\psi(\mathbf{r}) = (-\hbar^2/2m)\,\nabla^2\,\psi(\mathbf{r}) + U(\mathbf{r})\psi(\mathbf{r})$$



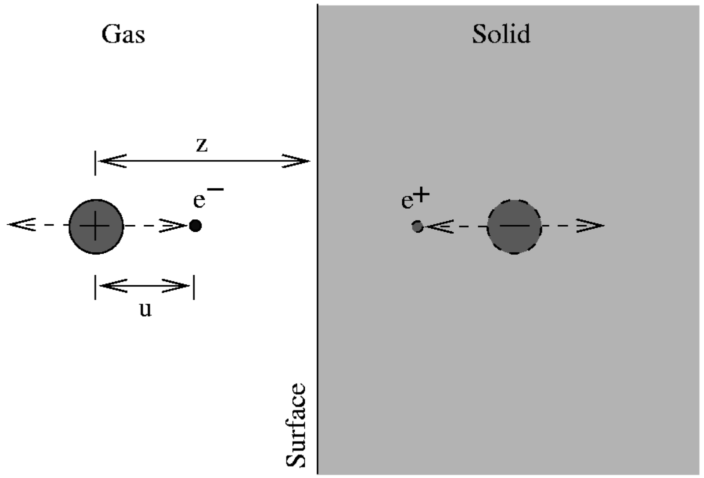
Z.Fan and Jia G. Lu IEEE Trans Nanotech. 5(4),393,2006



Zhang, Nano Letters 4(10):1919-1924 2004

Types of adsorptions

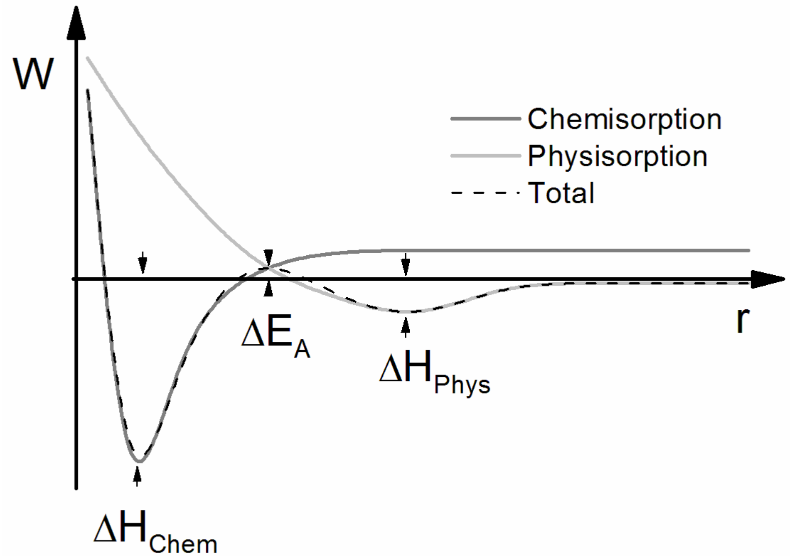
- Heterogeneous catalysis bigins with the act of adsorption.
  - Physisorption
    - ⌘Treated as dipol-dipol interaction (van der Waals interaction).
    - ⌘“Long distance interactions“, from 3E-10m to 5E-10m.
    - ⌘Novel-gas adsorbed on a metal.
  - Chemisorption:
    - ⌘Weak or neutral.
    - ⌘Strong or charged.
  - Ionsorption



$$V = \frac{e^-}{4\pi\varepsilon_0 2z}$$

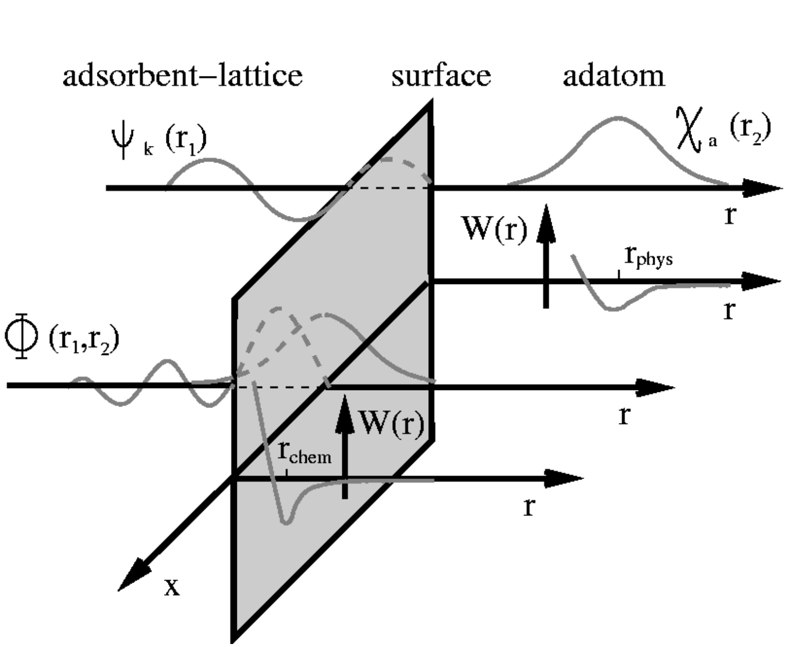
Chemisorption

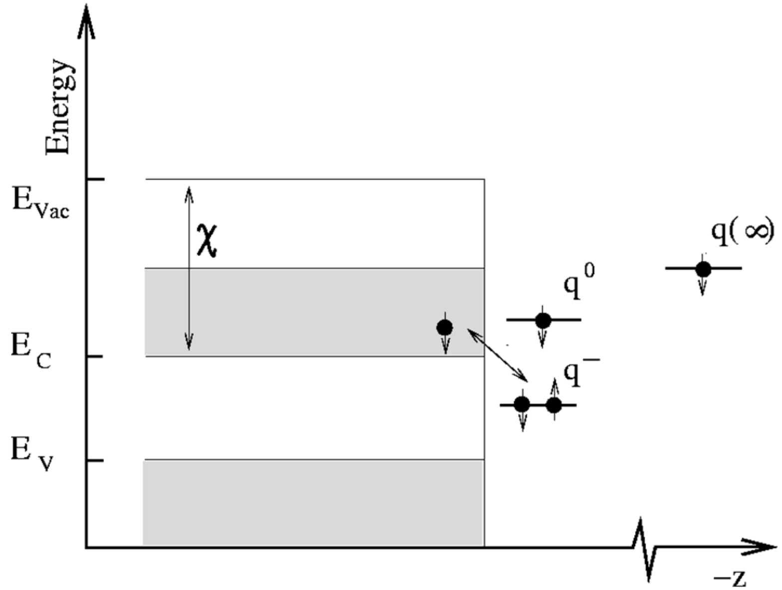
- Chemisorbed particle simultaneosly both affinity:
  - Acceptor
  - Donor
- Weak chemisorption (CL)
  - Particle remains electrically neutral.
  - Lattice electrons or holes do not participate in the bond.
- Strong chemisorption
  - Particle adsorbs free electrons or holes
    - ⌘n-bond (acceptor bond), CeL.
    - ⌘p-bond (donor bond), CpL.



Geistlinger theory

- If the electron at the conduction band and adatom has parallel spin, then a repulsive potential is produced. Therefore due to Van der Waals forces, particles are localized at a minimum potential resulting in a physisorbed state. Alternatively, if they have antiparallel spin, the wave functions are overlapped, which leads to chemisorption. Because of this, Wolkenstein postulated a subdivision into two states: Neutral or "weak chemisorbed" and charged or "strong chemisorbed". They are represented as discrete states, meaning that.





Only strong chemisorbed molecules produce change in the conductivity

Geistlinger, H. (1993). Electron theory of thin-film gas sensors. *Sensors and Actuators B: Chemical*, *17*(1), 47-60.

Fermi statistics

- When still valid the Fermi statitics?
  - Non-degenerated semiconductor, $E_V+3kT<E_F<E_C-3kT$.
- Fermi level
  - Controls the catalytics properties at the surface.
  - Once electronic equilibrium is stablished:
    - ⌘ Surface and bulk have the same Fermi level.

$$f^0 = \frac{1}{1+\frac{1}{2}\exp\left((E_F + e\Delta V_S - E_a^- + E_a^0)/kT\right)}; \quad f^- = \frac{1}{1+2\exp\left((E_a^- - E_a^0 - E_F - e\Delta V_S)/kT\right)}$$

- Many factors control the Fermi level possition.

Wolkenstien Isotherm

- The Fermi level determines the probably of weak and strong occupancy

$$f^0 + f^- = 1$$

$$\frac{N^-}{N} = f^- = \frac{1}{1 + 2 \cdot exp(\frac{E_t - E_F - e\triangle V_S}{kT})},$$

$$\frac{N^0}{N} = f^0 = 1 - f^- = \frac{1}{1 + \frac{1}{2}exp(\frac{E_F + e\triangle V_S - E_t}{kT})}$$

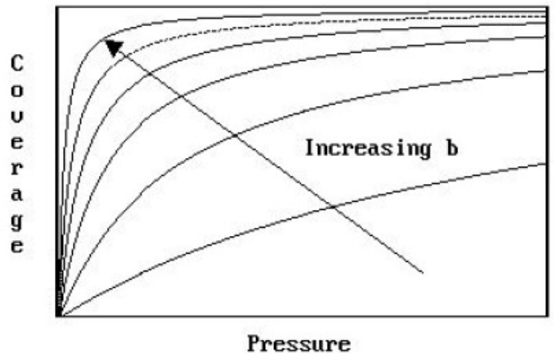
- The number of chemisorbed particles and adsorption sites are related to the coverage:

$$\theta = \frac{N}{N*} = \frac{N^0 + N^-}{N*} = f^0\theta + f^-\theta = \theta^0 + \theta^-$$

- The Langmuir isotherm is defined as

$$b = \frac{\alpha}{\nu^0}exp(\frac{q^0}{kT})$$

- The Wolkenstein isotherm is expressed as



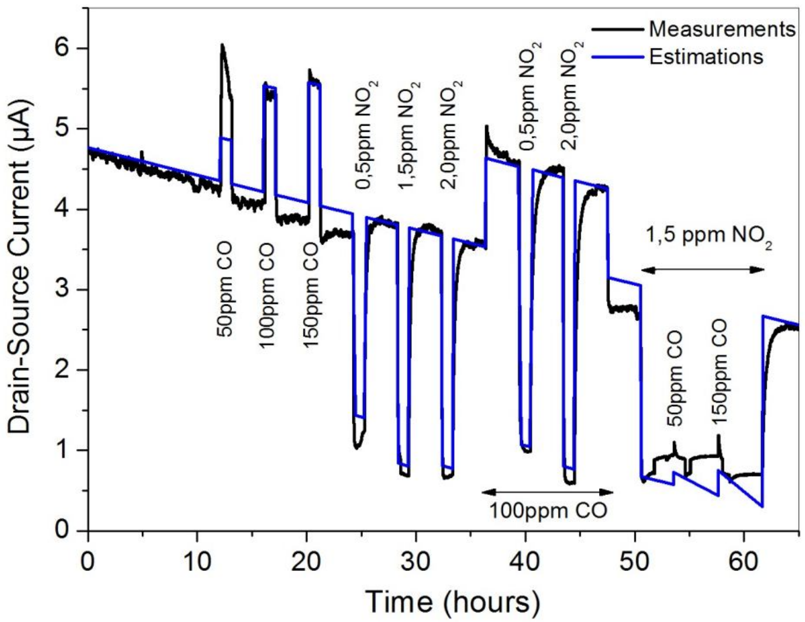
$$\beta = b\{f^0[1 + 2 \cdot \frac{\nu^- f^-}{\nu^0 f^0}exp(\frac{E_t - E_c}{kT})]\}^{-1}$$

$$\theta(\tilde{p}) = \frac{\beta\tilde{p}}{1 + \beta\tilde{p}}$$

Co-Adsorption-Modell

- They don‘t reach with each other
- Certain interaction
  - Adsorbates compite for the free adsorption centres

$$\theta_j^- = \frac{\beta_j \cdot p_j}{1+\sum_{i=0}^{n} \beta_i \cdot p_i} \cdot f^- \qquad N^- = \sum_{i=0}^{n} N_{Surf} \cdot \theta_i^-$$

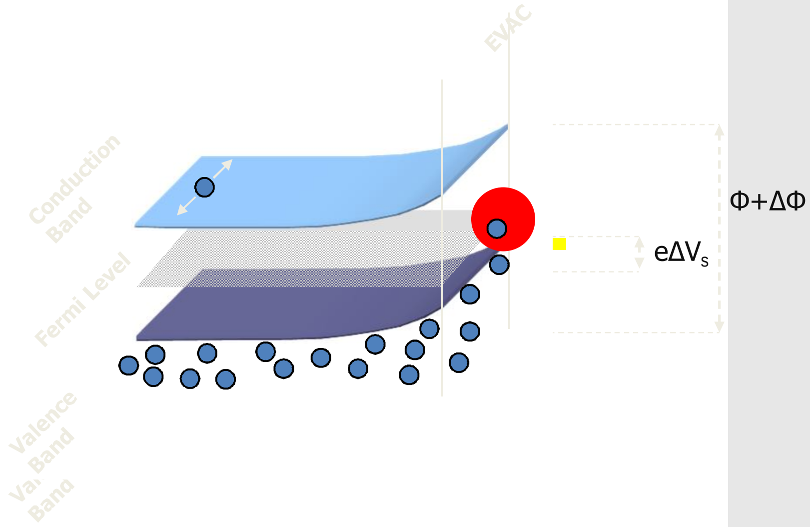


- Problem to extend this model to catalysis
  - Same problem with Langmuir isotherm

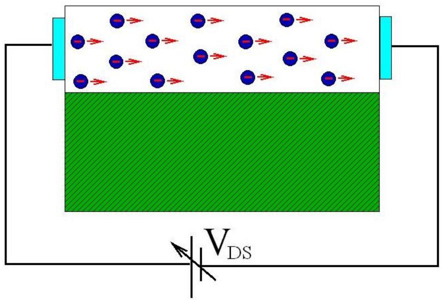
Charge transfer

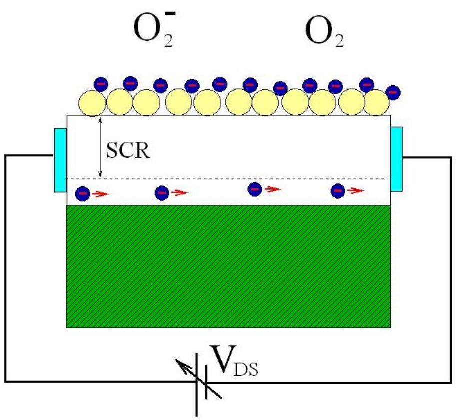
- Band bending
  - Surface potential.
  - Work funtion change
  - Depleted zone (SCR).
  - Electrical neutrality

$$Q_{SCR} = Q_{SS}$$

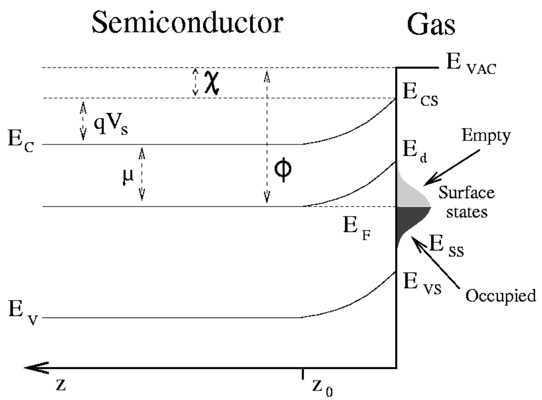


- Electric properties
  - Changes in the bulk conductivity





Charge transfer



- Charge neutrality $Q_{SCR} = Q_{SS}$
- Charge surface $Q_{SS} = e^{-} \cdot \int_{-\infty}^{+\infty} D_{SS}(E) f_{SS}(E) dE$
- Distribution function $f_{SS}(E, E_F) \rightarrow f_{SS}(E, E_{F,bulk} + e\triangle V_S(\theta^{-}))$
- Poisson equation $\frac{d^2V}{dz} = -\frac{\rho}{\varepsilon_0 \varepsilon_r}$
- Surface potential $V_S = \frac{eN_D z_{SCR}^2}{2\varepsilon_0 \varepsilon_r}$ $V_S = \frac{e(N^{-})^2}{2\varepsilon_0 \varepsilon_r N_D}$
- Movility thermally activated $\mu = \mu_0 \cdot exp(-e^{-}V_S/kT)$

Weisz limit

$$(N^- / N) = f^-\left(E_a^{0/-}, E_F, e\Delta V_S\right)$$

$$f^- = \frac{1}{1 + 2 \cdot exp(\frac{E_t - E_F - e\Delta V_S}{kT})}$$

$$V_S = \frac{e(N^-)^2}{2\varepsilon_0\varepsilon_r N_D}$$

$$Z_{SCR} = (N_S / N_D) = 1E14cm^{-2} / 1E18cm^{-3} = 1\mu m$$

$$V_S = 696V!!!!!!!!!!!!!!!!!$$

$$V_S = \frac{eN_D z_{SCR}^2}{2\varepsilon_0\varepsilon_r}$$

Self Limitation of Strong (=charged) Chemisorption

This is the Weisz Effect

Electrochemistry knows the same Effect: Mott-Schottky

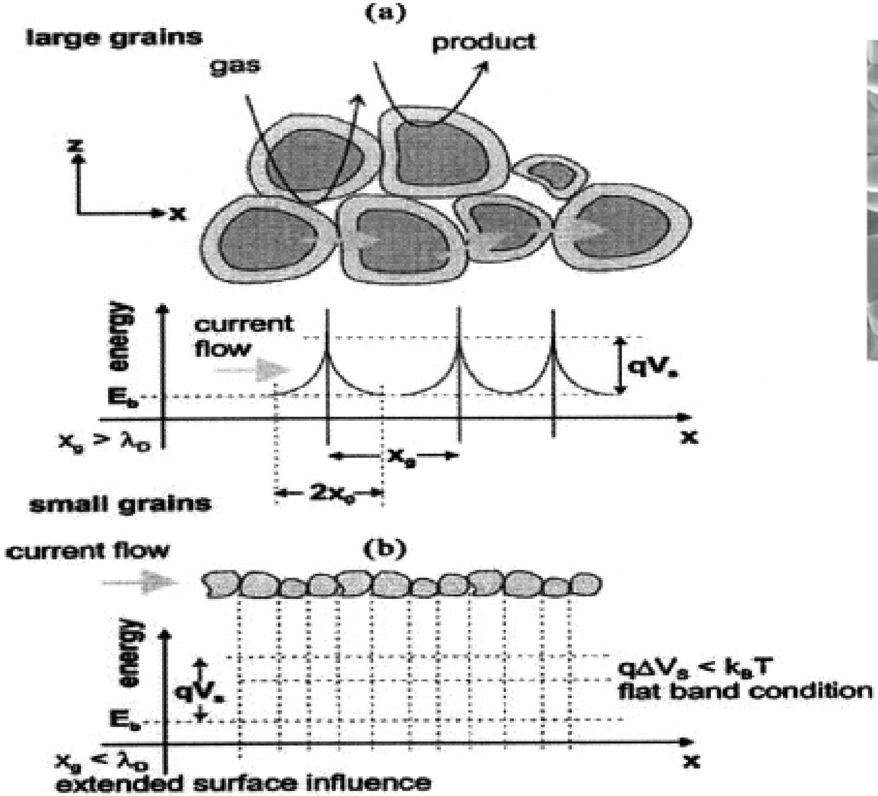
*Weisz, P. B. (1953). Effects of electronic charge transfer between adsorbate and solid on chemisorption and catalysis. *The Journal of Chemical Physics*, *21*(9), 1531-1538.

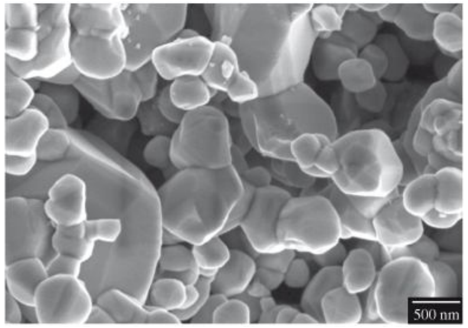
Grain size

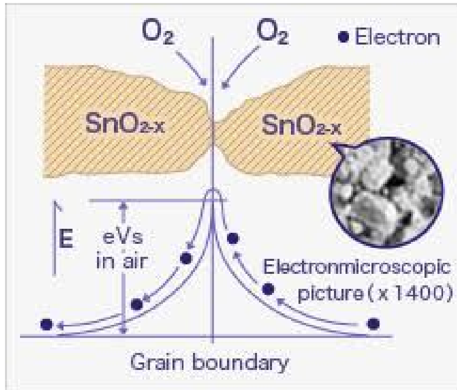
- Flat band limit
  - ➢Crystallites $d_{Grain} \sim d_{Space\ Charge}$
  - ➢Fermi statictis:
  - ➢Thermoionics vs tunneling

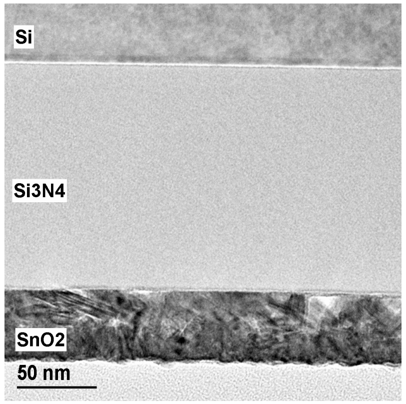
$$L_D = \sqrt{\frac{\varepsilon_0 \cdot \varepsilon_r \cdot kT}{\left(e^-\right)^2 \cdot N_D}}$$

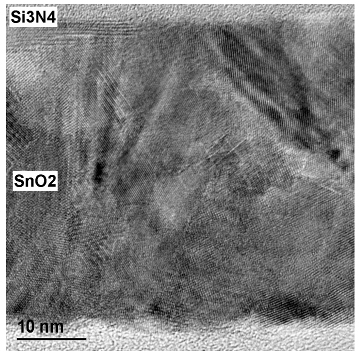
$$E\,\psi(\mathbf{r}) = (-\,\hbar^2/2m)\,\nabla^2\,\psi(\mathbf{r}) + U(\mathbf{r})\psi(\mathbf{r})$$







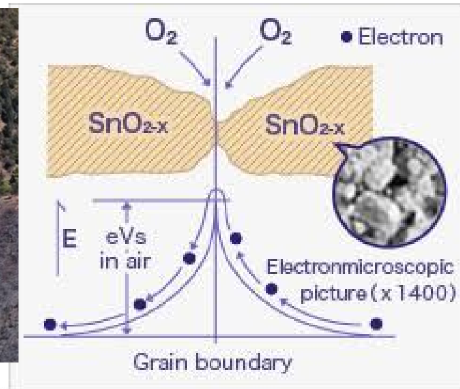




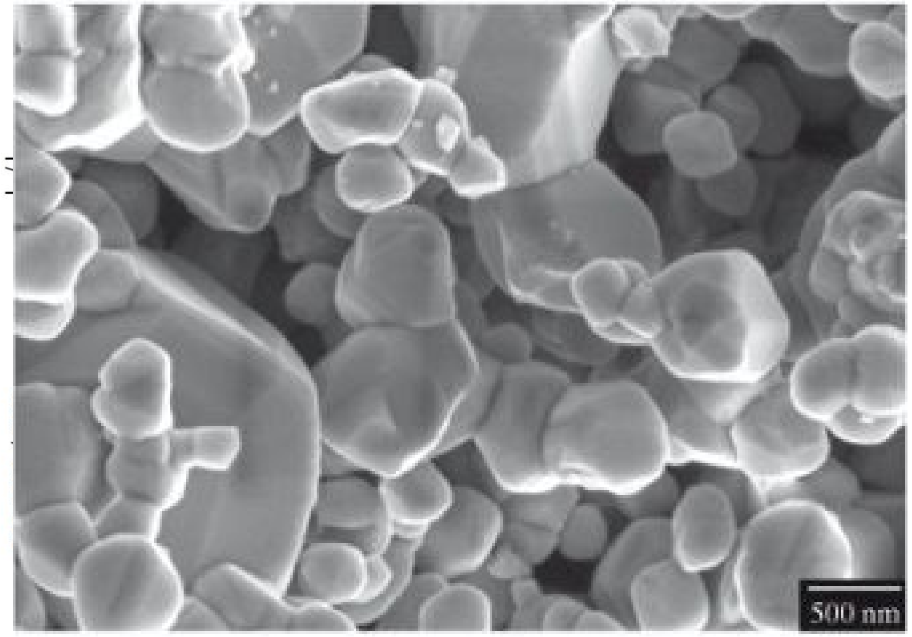
Barsan, N., & Weimar, U. (2001). Conduction model of metal oxide gas sensors. *Journal of Electroceramics*, *7*(3), 143-167.

Summary

- Charge transport and transfer description
  - Flat band conditions (Fermi-Dirac statistic): Thermo-ionic
  - Wolkenstein isotherm
  - Charge transfer
- Complex description
  - Schrödinger equation
  - Shockley-Read statictics: Tunneling



V_{DS} = 2V



Outline

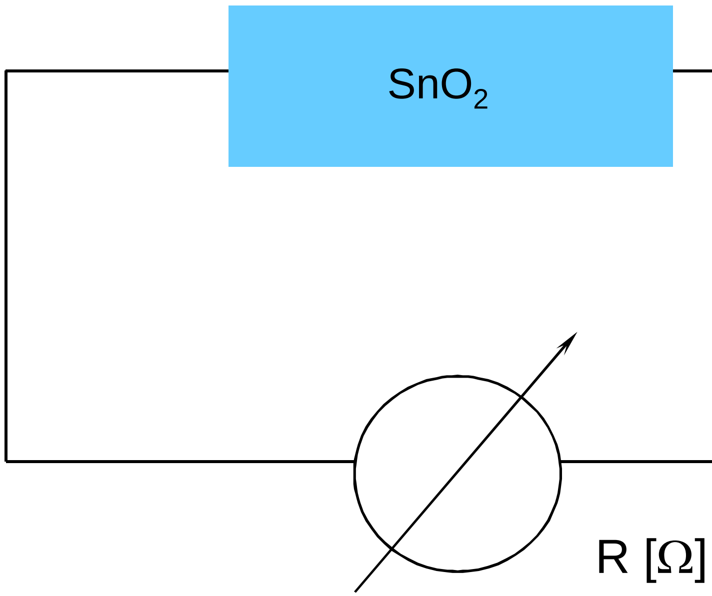
- Motivation

- Classic semicondutor theory
  - Ideal „Bulk“
- Semiconductor theory at the bulk edge
  - Semiconductor surface-gas interactions

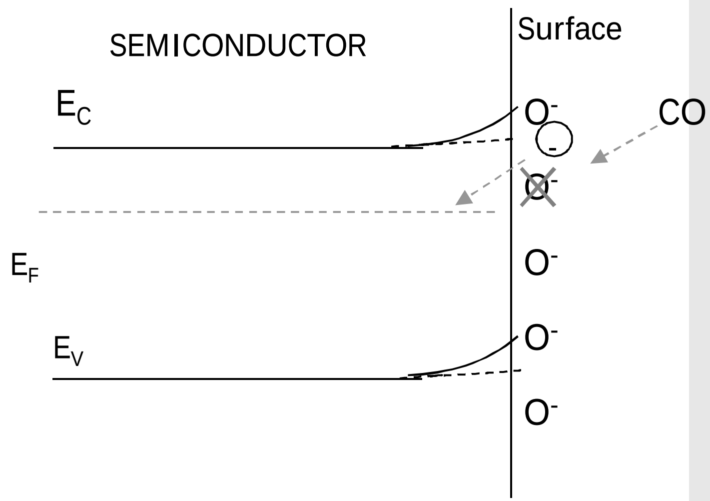
- One example of device operation
  - Electro-adsorptive effect
  - Thin film transistor
  - Drift of vacancies (dopants)

- Remarks

The challenge



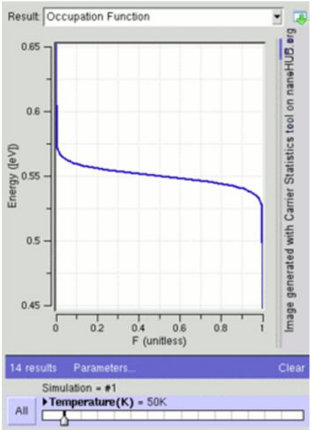
Surface:

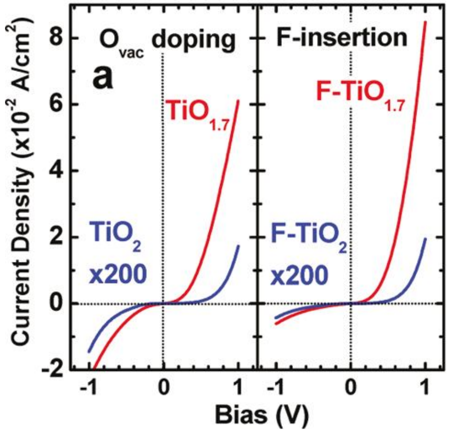


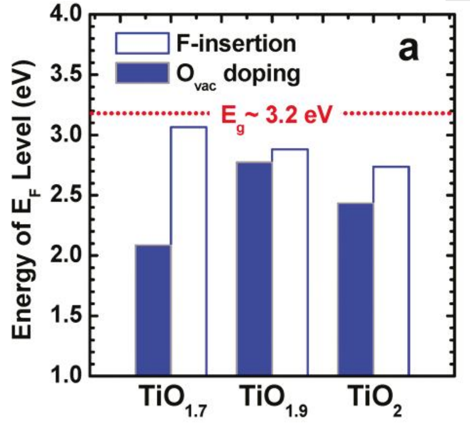
Operation temperature approx: 300°C ⟶ ~~CMOS~~ !

Control adsorption/desorption

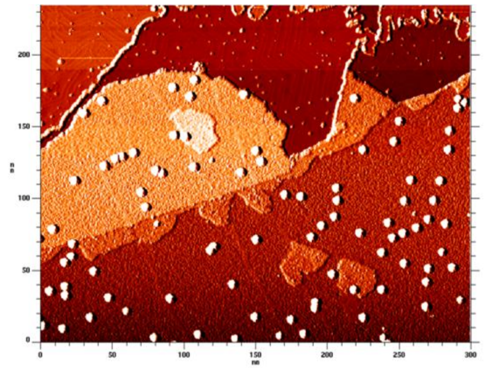
- Temperature
- Doping change Fermi level
- Surface functionalization
- External electric field

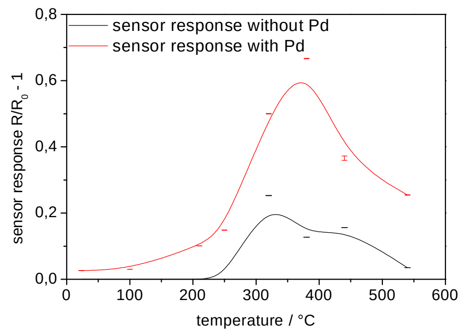






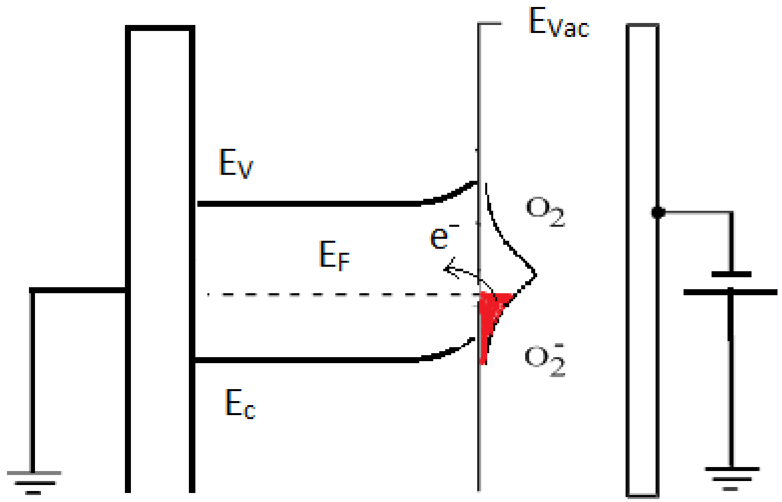
Nano Lett. 2011, 11, 751–756





Electroadsorptive Effect

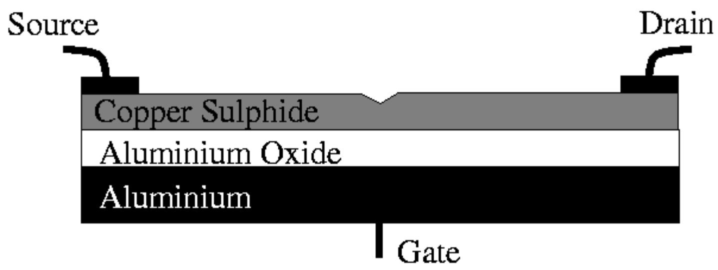
- EAE:
  - “Surface states electrical modulation on the metal-oxide surface by means of an external electric field”



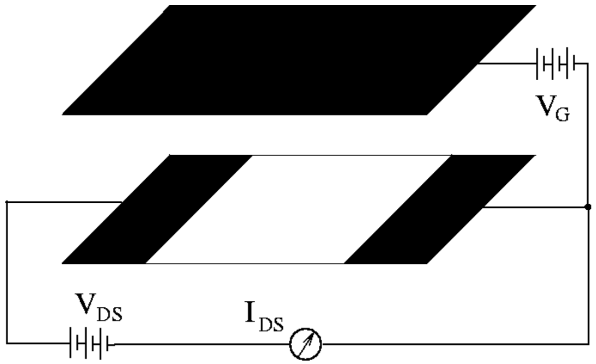
Short history: EAE

Field-Effect devices

- JSE. Lilienfeld, "Method and apparatus for controlling electric currents",US Patent1, 745, 175, 1930.
  - It dind‘t work
  - Why...
- O. Heil control of the layer conductivity by the surface states, 1935.
- W. Shockley, GL. Pearson, "Modulation of Conductance of Thin Films of Semi-Conductors by Surface Charges", Phys. Rev. Vol. 74, pp. 232-233, 1948.
  - 10% were mobile.
  - Surface states acts like electrical tramps.
- F. Wolkenstein, 1958.



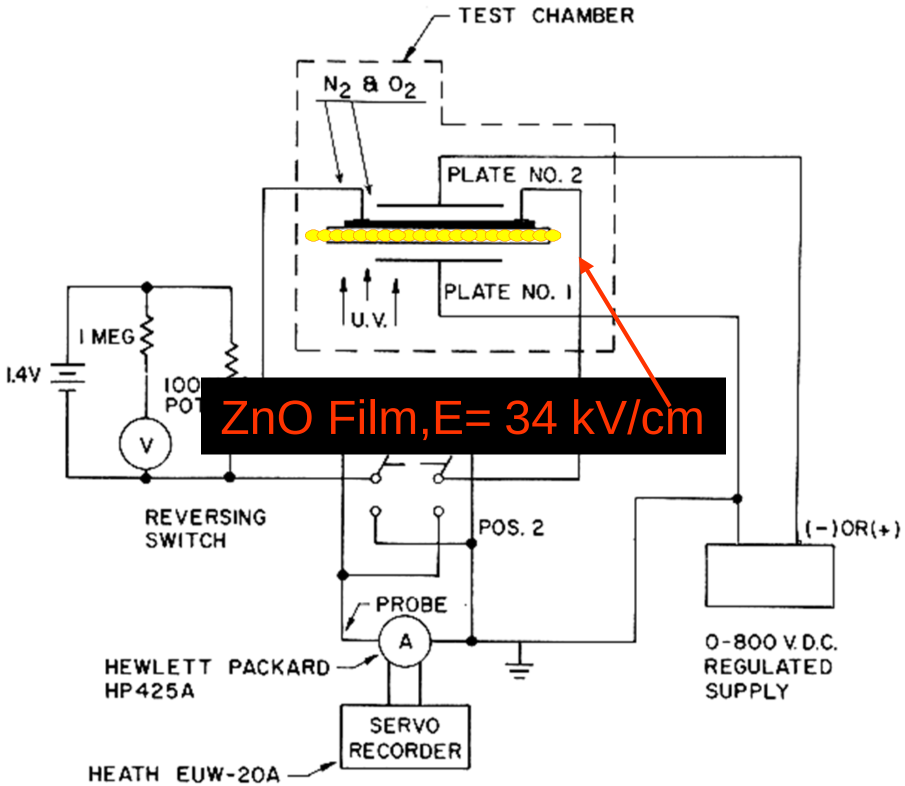
Patente de Lilienfeld (1930)

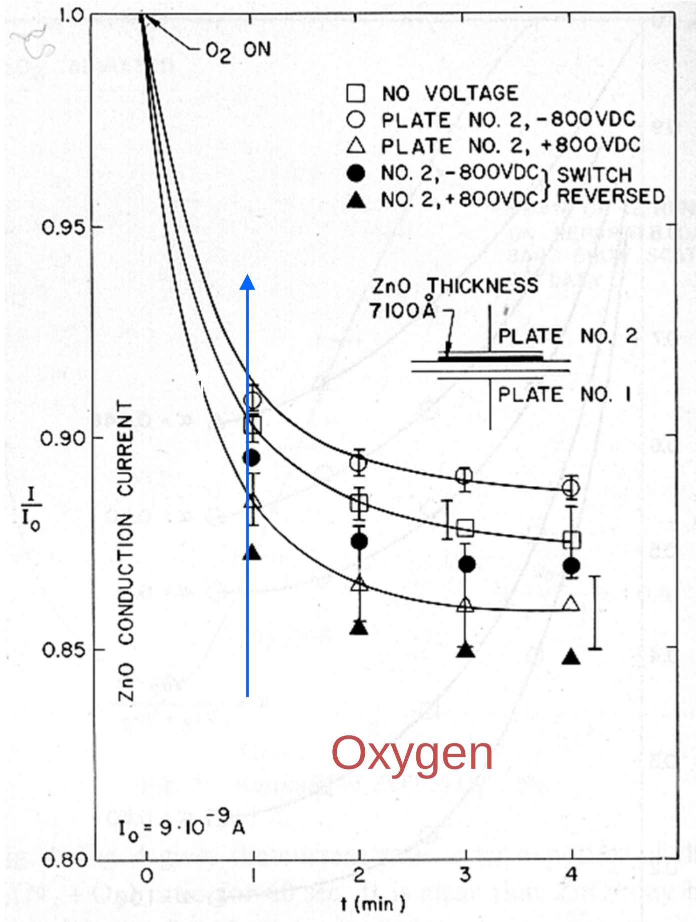


Patente de Heil (1935)

Adsorption of Methanol in Ge: Keier and Mikheeva 1964

O_2 in ZnO: Hoenig and Lane, Surf. Sci 11, 1968

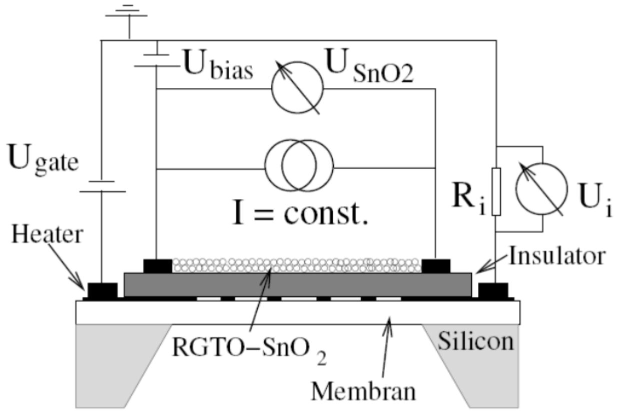




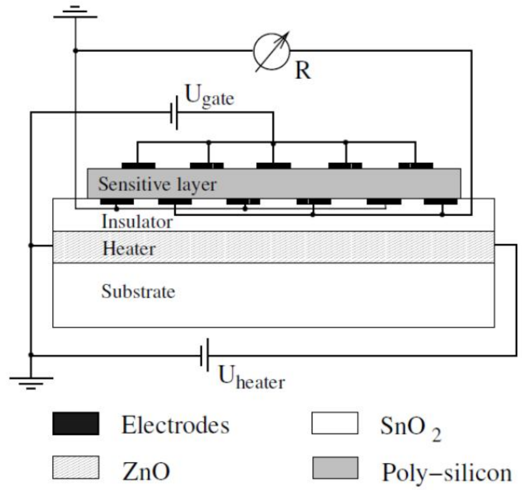
Short history: EAE

With micro always high electric field

SnO_2 Gate Transistor: Popova and Stoyanov	1994
Consistency to the theory: Geistlinger	1994
SnO_2 thin film: Hellmich and Müller	1996
SnO_2 multi-electrode Sensor:	
Hausner and Binder	1997
Thin film transistor gas sensor:	
Jaegle and Wöllenstein	1998



Hellmich and Müller (1996)

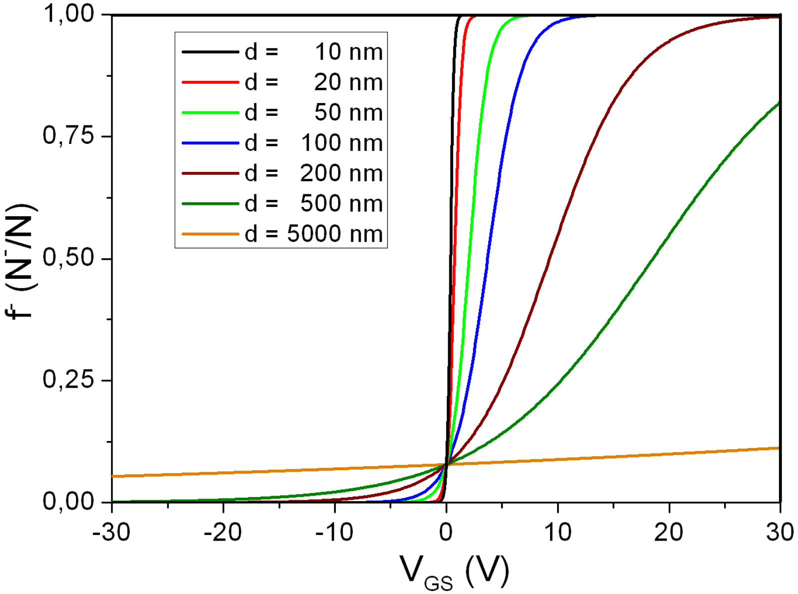


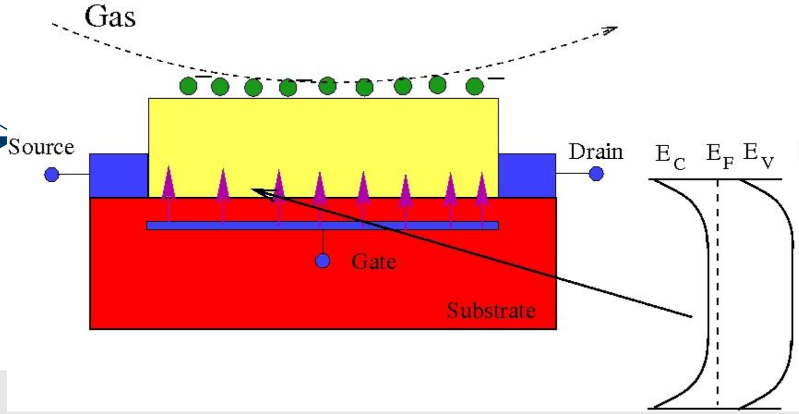
Hausner and Binder (1997)

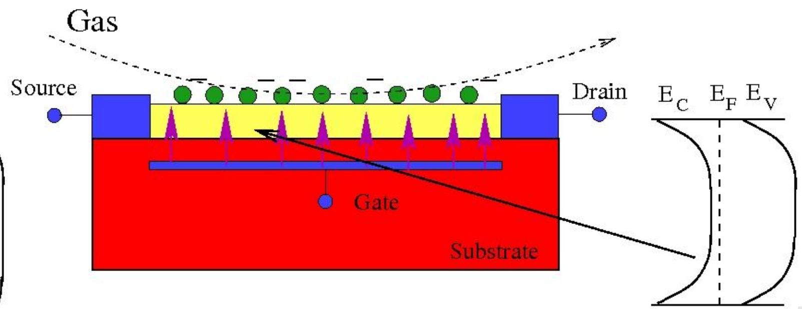
Thickness

- **Thickness:**
  - Must be in the Debye Length range.
  - Energy levels are modulated
  - Reaction on the

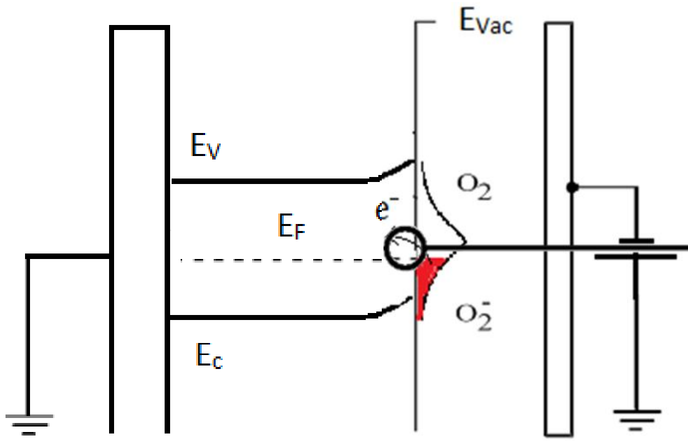
Estimated strong occupancy probability vs. gate voltage:

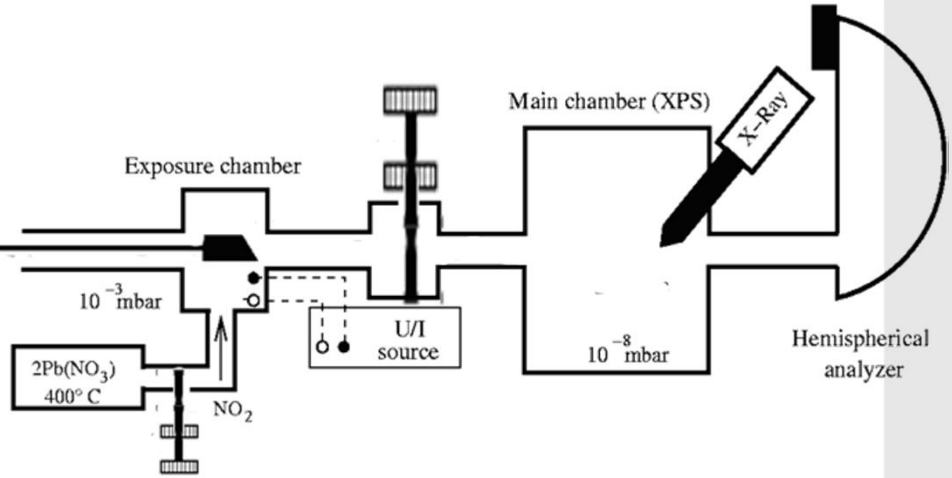


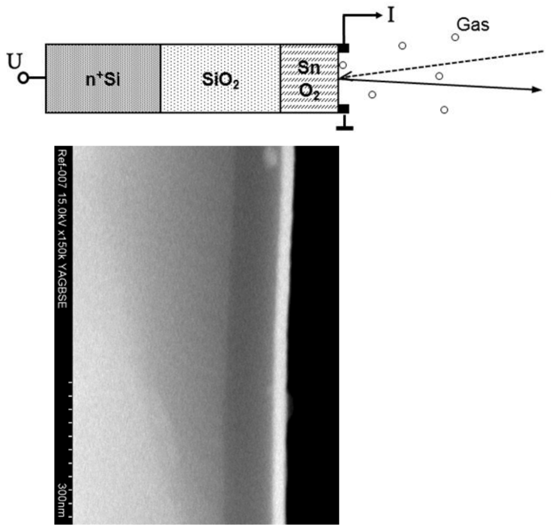


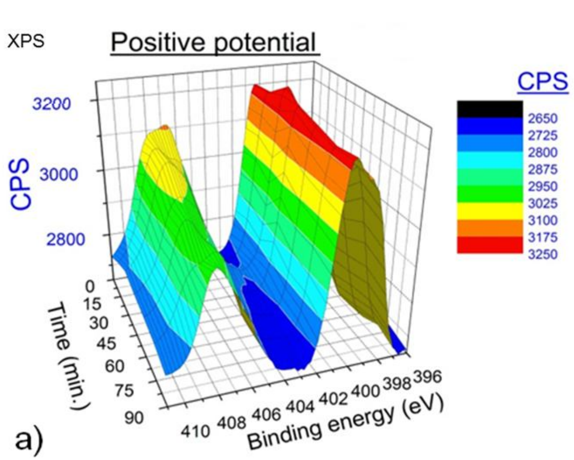


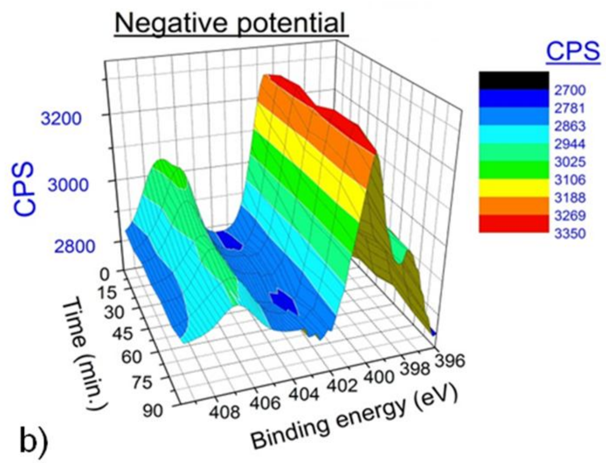
Example at UHV: Fast transfer, EAE







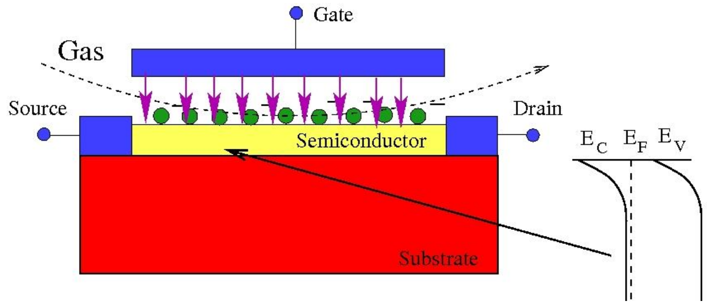




*ChemPhysChem, 14(11), 2505-2510, 2013

Development

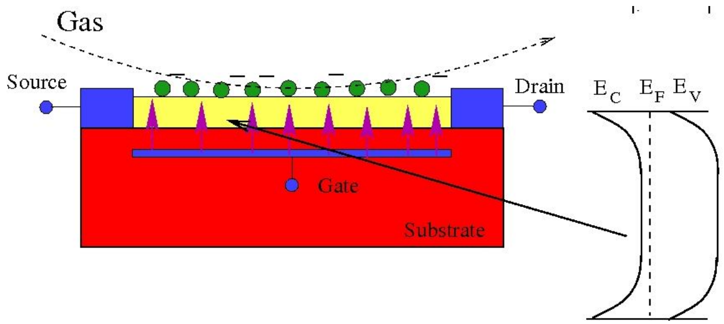
- Suspended Gate
  - ❿ Effective control on the surface states
  - ❿ Very difficult to realice this geometry with standard CMOS processes.



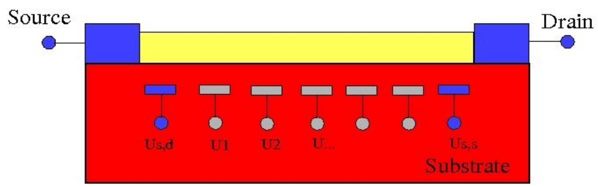
- Thin-Film-Transistor Gas Sensor
  - ➢ Debye-Lenght:

$$L_D = \sqrt{\frac{\varepsilon_0 \cdot \varepsilon_r \cdot kT}{\left(e^-\right)^2 \cdot N_D}}$$

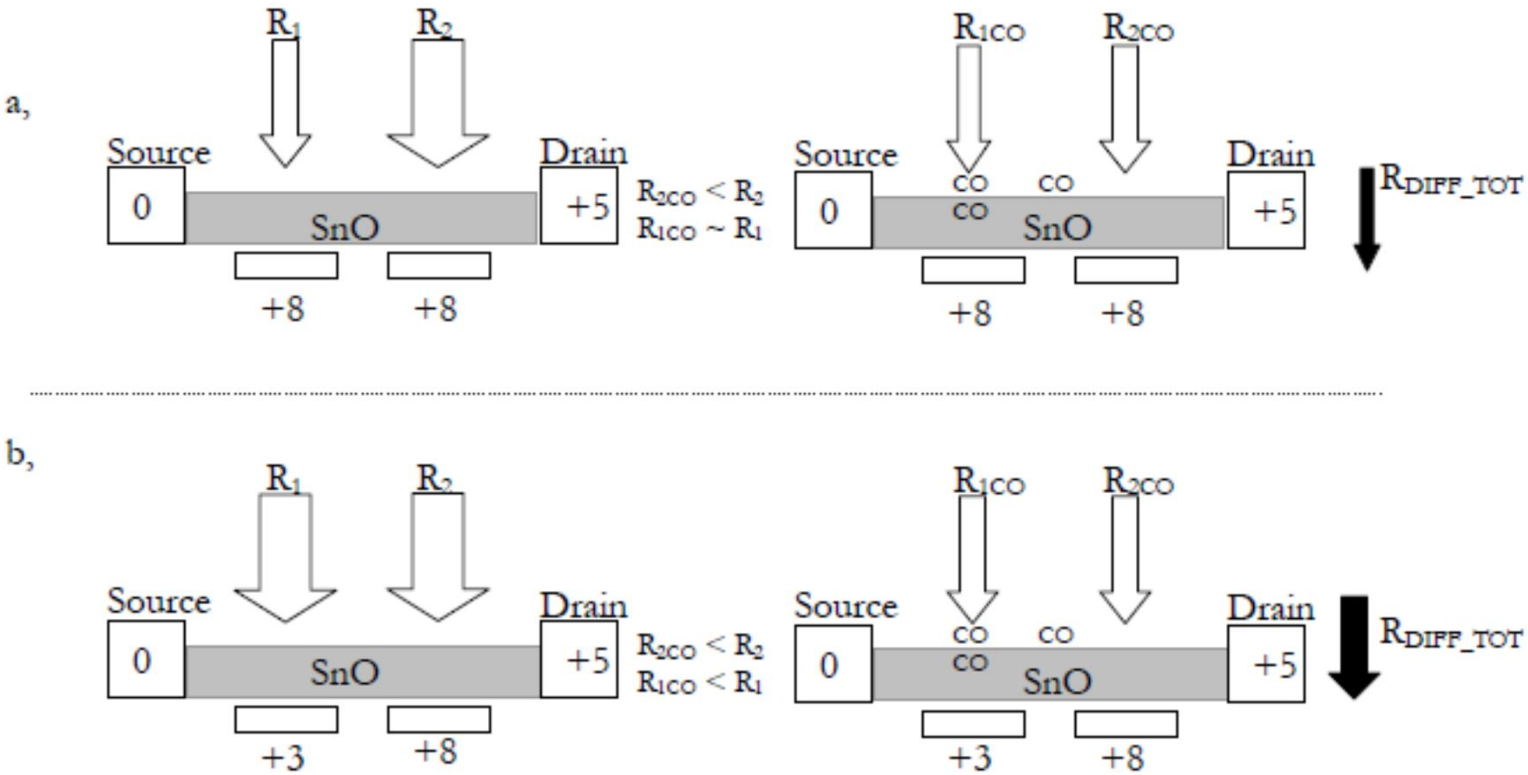
  - ➢ Effective control of the Fermi-Level
  - ➢ Technology: CMOS-Standard processes



- Multi-Gates
  - ➢ Homogeneous surface reaction

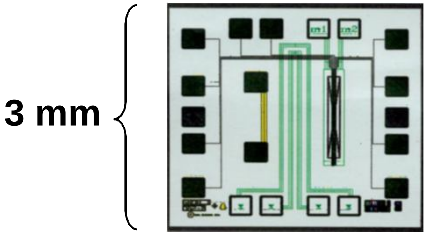


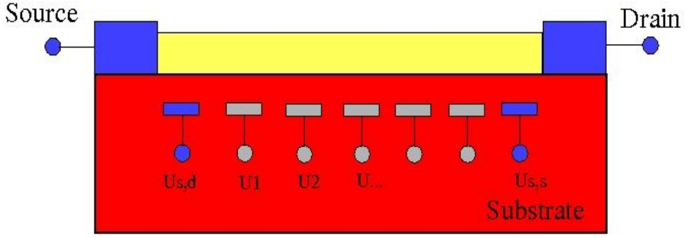
Multigate concept



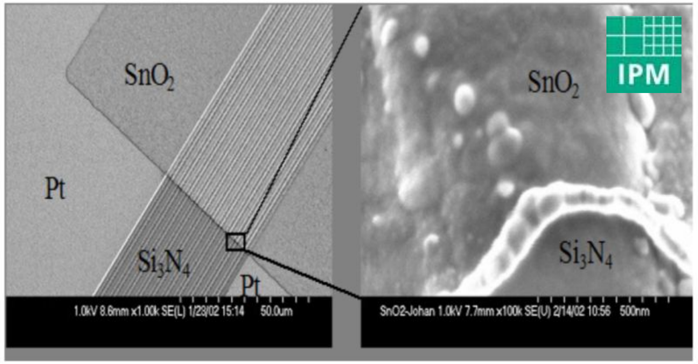
Technology

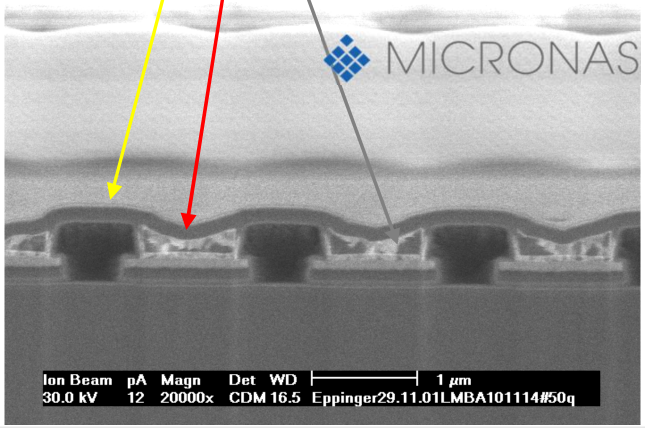
- SEM image „Multi-Gates“ Structure*
  - Source-Drain TFT
  - Semiconductor SnO_2 and insolator Si_3N_4
  - Contact Pt
- Compact poly-crystalline**
  - Non predominant orientation
  - Grain size from 5 to 25 nm (Flat-band condition)
- Sensitiven layer 55 nm





Sensitive layer (55 nm)
Multi-Gates
Si_3N_4: 250nm

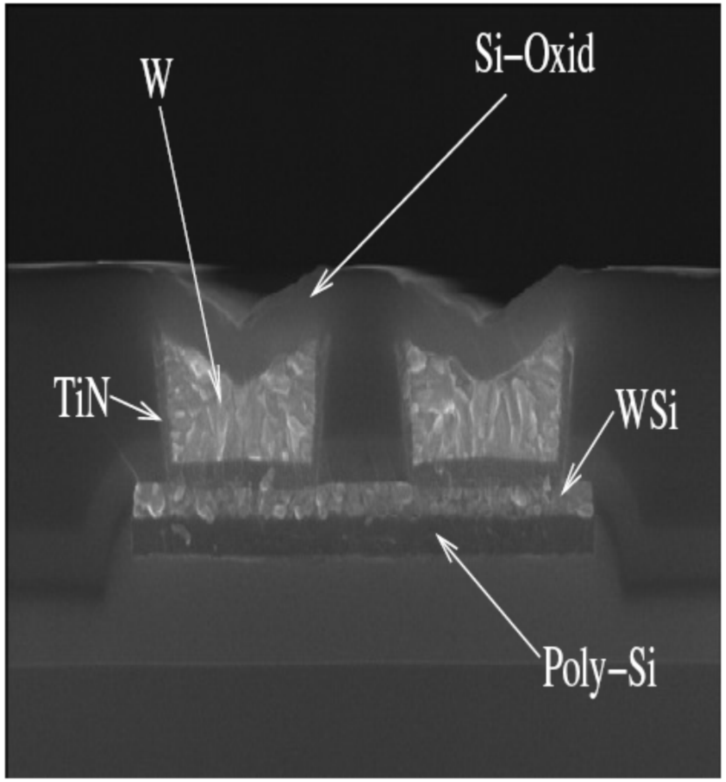


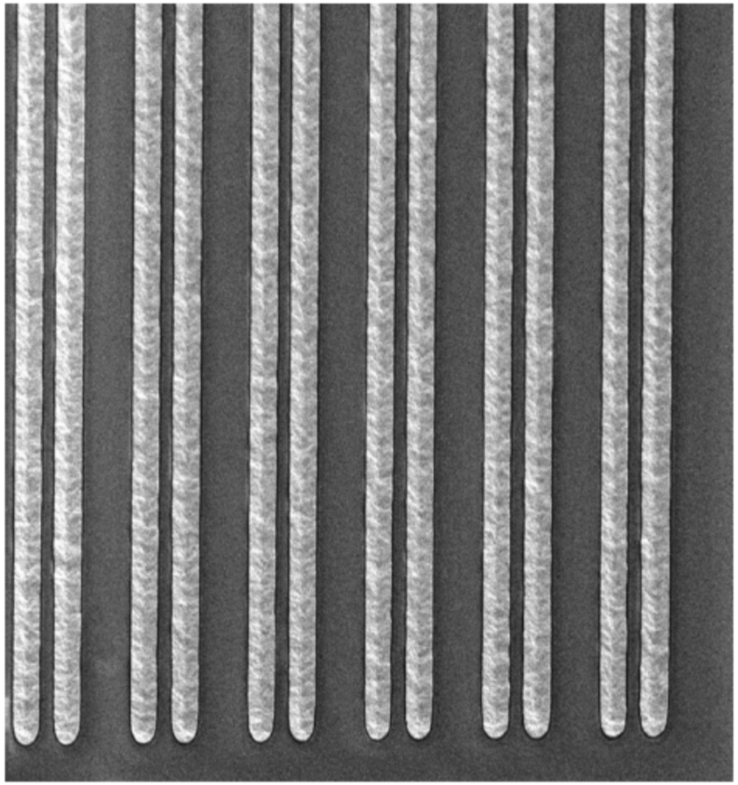


*M. Lehmann, H. Frerichs **J. Wöllenstein

Technology

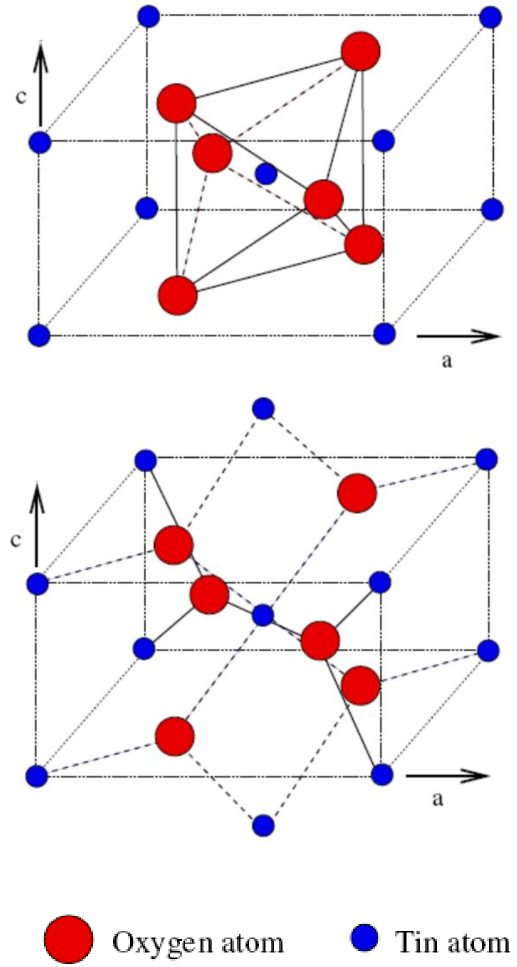
- Gate electrode
  - Multi-Gate-Structure, top view (Right)
  - Gate electrode, Crossection (Left)





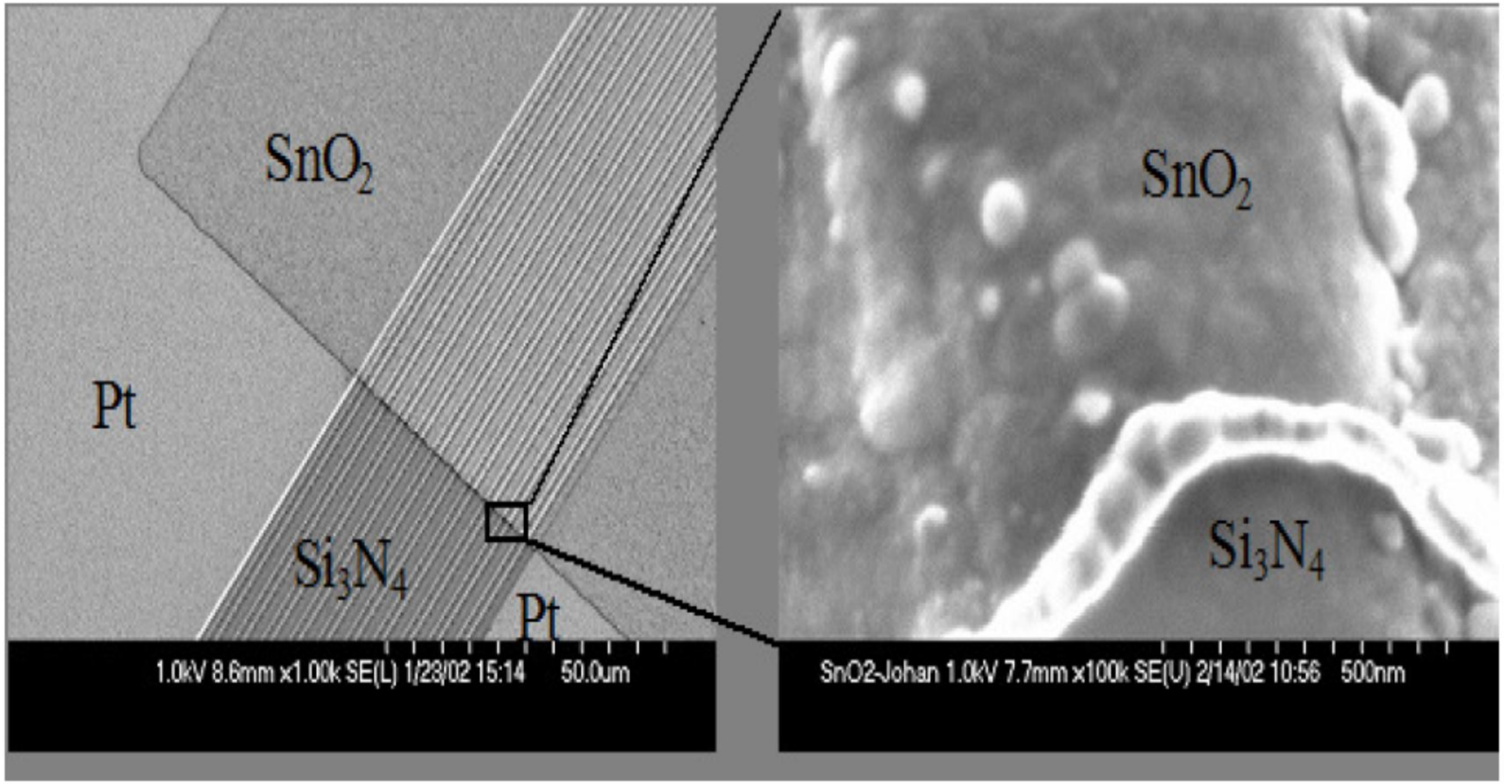
Material SnO2

- Octahedron and rutile (a=b=0.47nm, c=0,32nm, O-Sn-O=77°20´)
- Inert to the acid-base reaction
- Heat of formation ΔH=1.9E13(J/mol)
- Density at 300 K is 6,95 (g/cm^3)
- Melting point at 1630°C
- Direct band gap 3,5 eV
- Small electron effective mass m*=0,275 m0 , good conductivity
- Neutral oxygen vacancies (V_0, they play role as donators) form energy states at 0.035 eV and 0.140 eV
- It is a n-type semiconductor due the existence of native donators levels



Technology

- SEM view of a multi-gate structure
  - Source and drain area of a multiple TFT (left)
  - SnO_2 and Si_3N_4, on right



Electric model

- Sensor-Modell:
  - Poisson +continuity equation: Signal behaviour

$$\Delta \cdot \psi = -\frac{\rho}{\varepsilon_0 \cdot \varepsilon_r}$$

$$\nabla \overrightarrow{J_{th}} + e^- \cdot \frac{\partial}{\partial t}(p-n) = 0$$

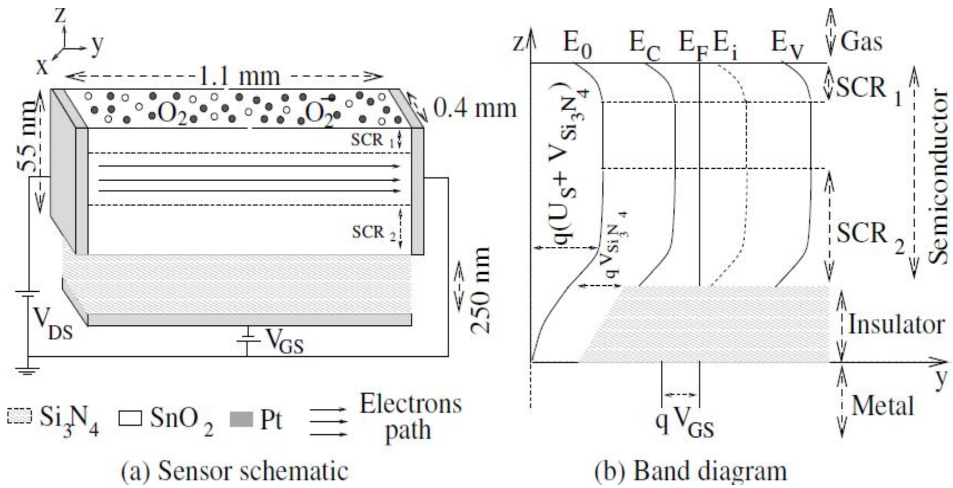
$$Q_{SS} = Q_{SCR}$$

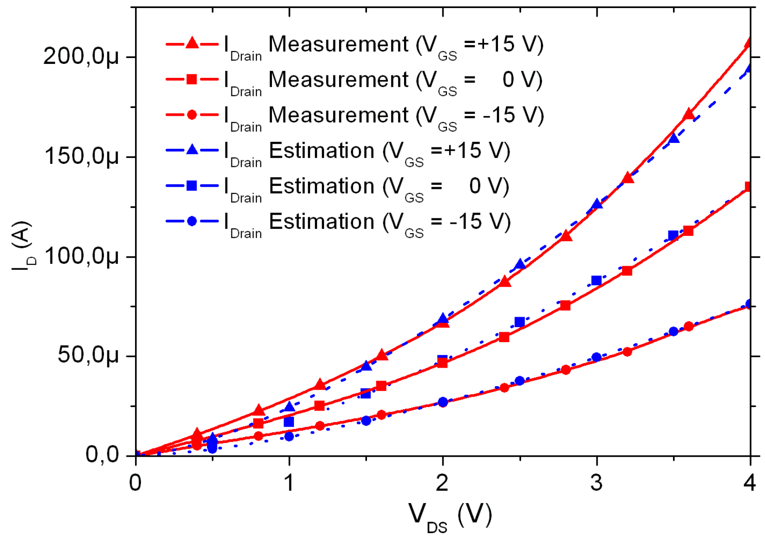
  - Wolkenstein-Modell: Surface reaction

$$\theta(p) = \frac{\beta \cdot p}{1+\beta \cdot p}$$

$$\beta = b\left\{ f^0 \left[1 + \frac{\upsilon^- f^-}{\upsilon^0 f^0} \cdot \exp\left(\frac{E_t^f - E_c}{kT} \right) \right] \right\}^{-1}$$

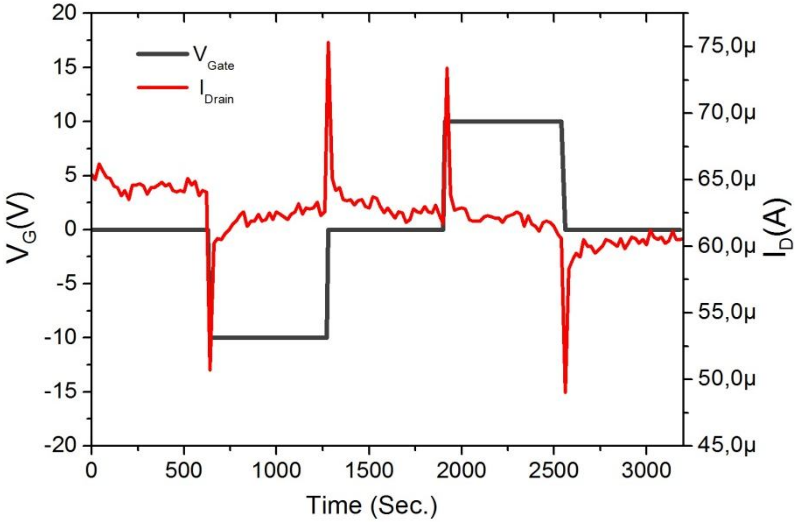
- Drain-current: Controled by V_{DS}
- Sensor works like MOSFET with reaction to gases

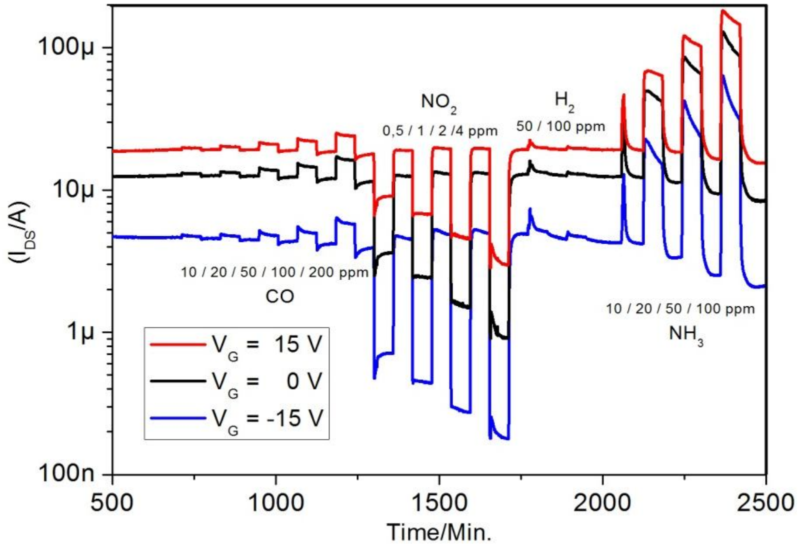




Gas measurements

- Ideal current signal
  - Proportional to the gate potential
  - Not time depending (stable)
- Measurements with GasFET
- Solution: Pulse at the gate
- Parameter:
  - Synthetic ait 80% N_2 and 20% O_2, RH 40%, Work temperature 200°C, V_{DS}=1 V

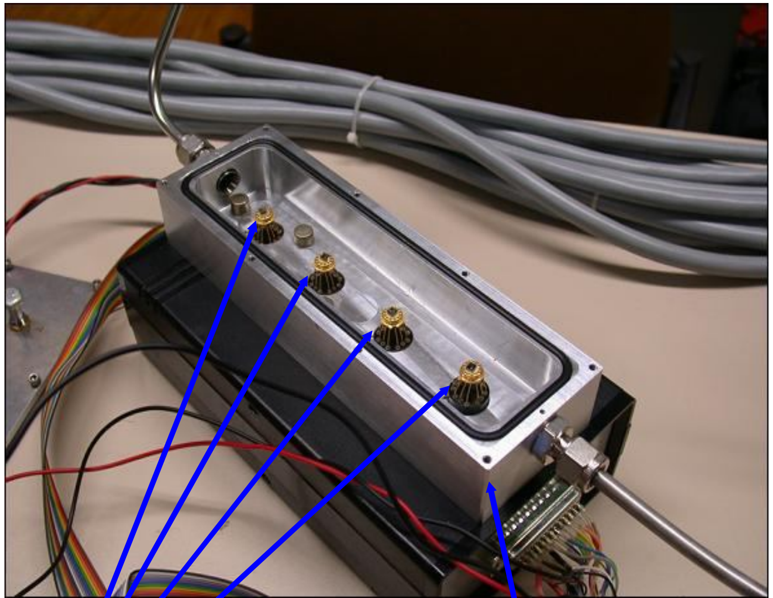


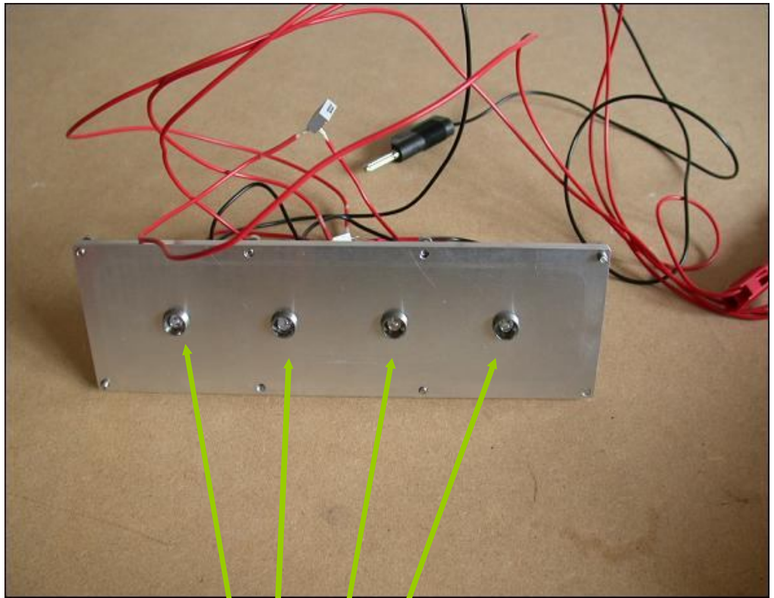


AP 2 Gasabhängige Charakterisierung der Schichten

Lichteinfluss auf gassensitive Nanoschichten

Messaufbau:



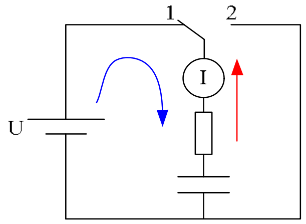


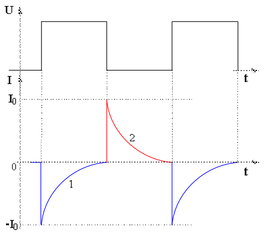
Gassensoren Messkammer

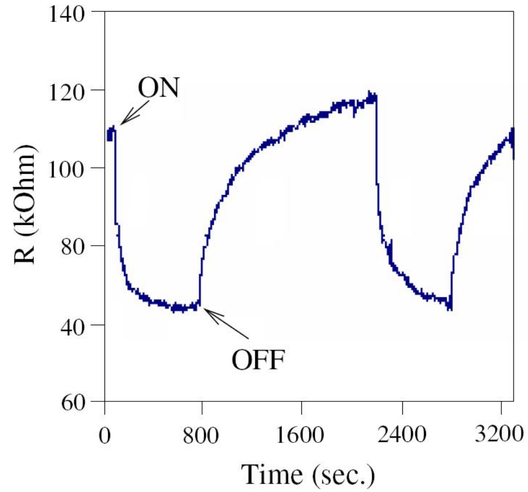
LEDs bei verschiedenen Wellenlängen
(blau und UV)

Signal drift

- MIS-condensator
  - ➢ Low: C = 2.2 pf



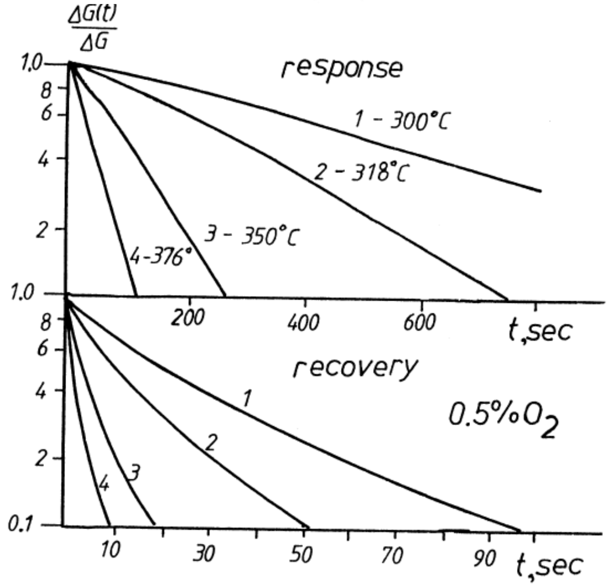




- Charge carriers Generation
  - ➢ Ilumunation (LED 340 nm)
    - ⌘t_{ON}: 40-60 s
    - ⌘t_{OFF}: 80-120 s

- Surface kinetic reaction*
  - ➢ Simulation parameters: 10^{10} times smaller than real paremeters

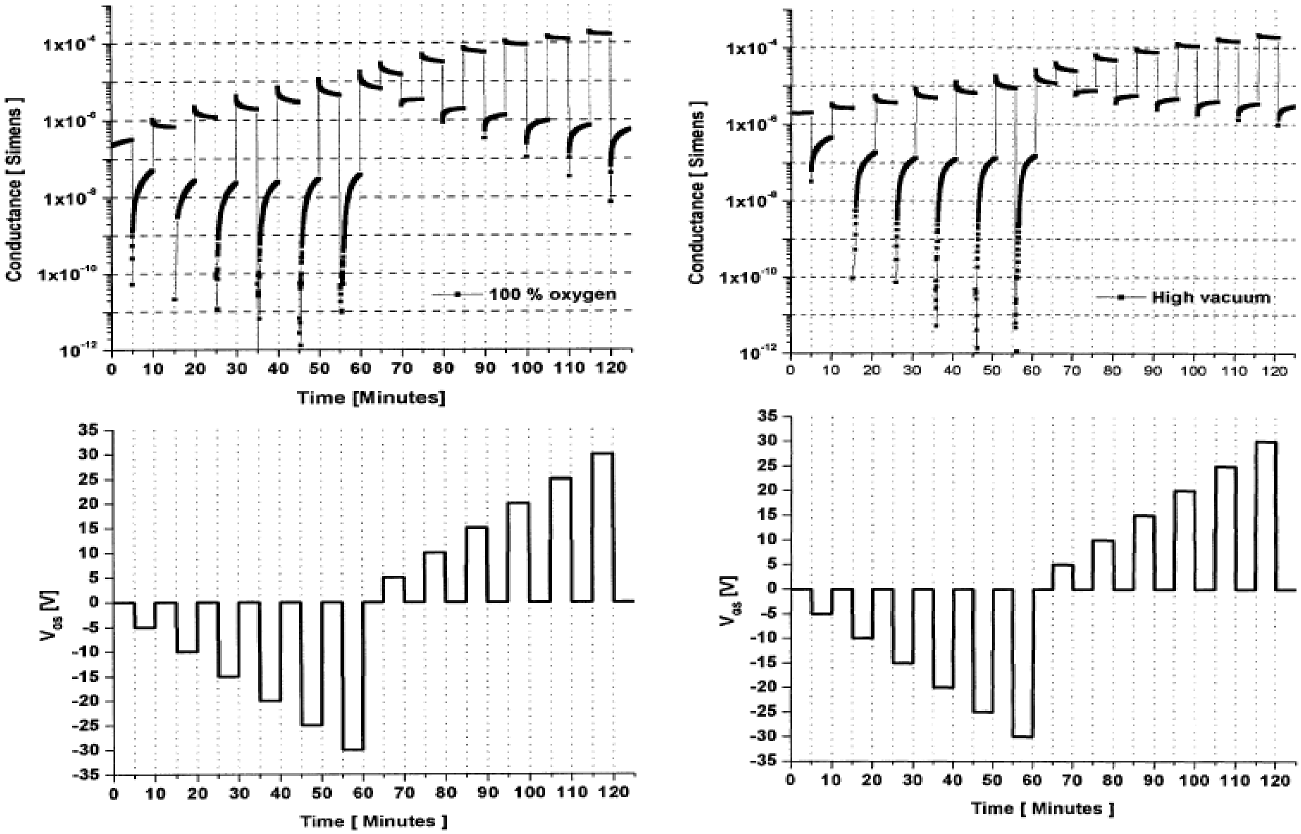
- Chemical reaction
  - ➢ Vacuum measurements, same signal behaviour



*V. Bryznari et al. Sensors and Actuators B,61,143-153, 1999

High vacuum

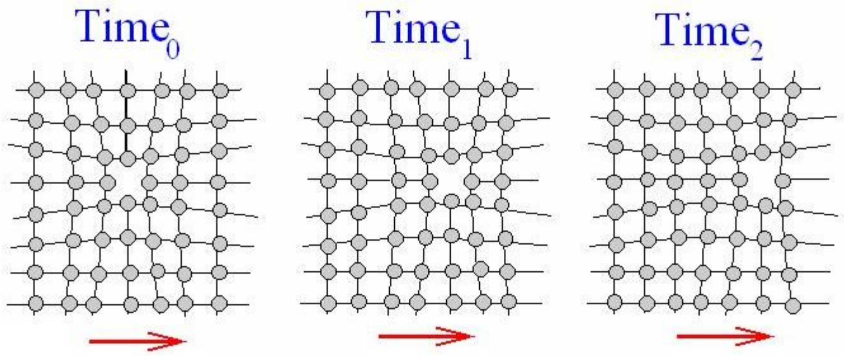
H.L. Tuller, Massachusetts Institute of Technology*



„We're all in the same boat“

Oxygen vacancies diffusion

Doping diffusion by electric field



- Fick diffusion

$$\frac{\partial N}{\partial t} = D_{vac}\Delta \cdot N$$

- Fokker-Planck-Equation

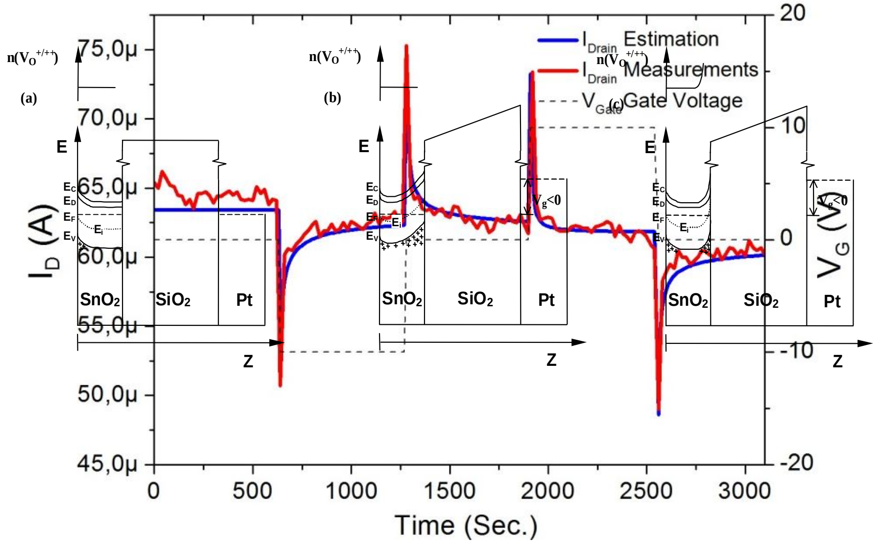
$$\frac{\partial N}{\partial t} = \frac{e^-}{m_{vac}^{eff}}\nabla \cdot (E \cdot N) + D_{vac}\Delta \cdot N$$

- Drift-Diffusions-Equation

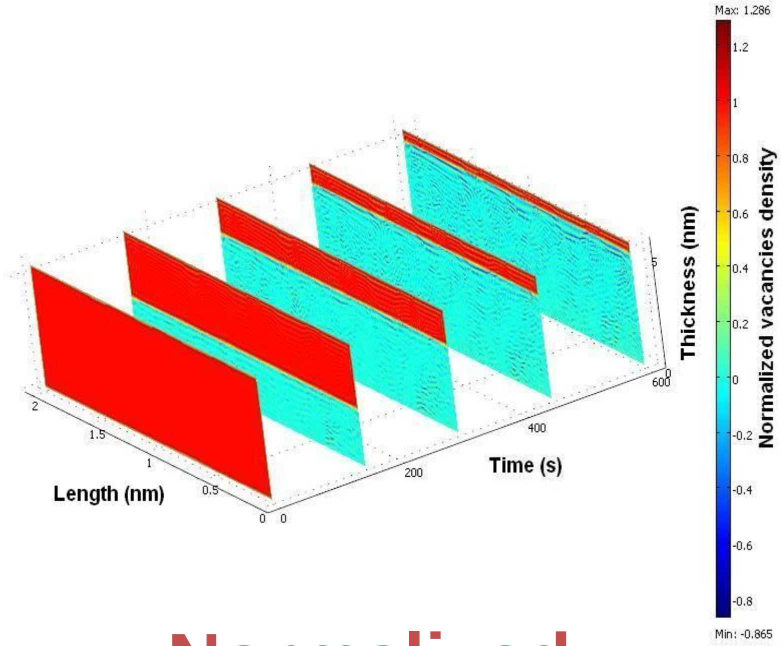
$$\frac{\partial n}{\partial t} = -\mu_n \nabla \cdot (E \cdot n) + D_{e^-}\Delta \cdot n$$

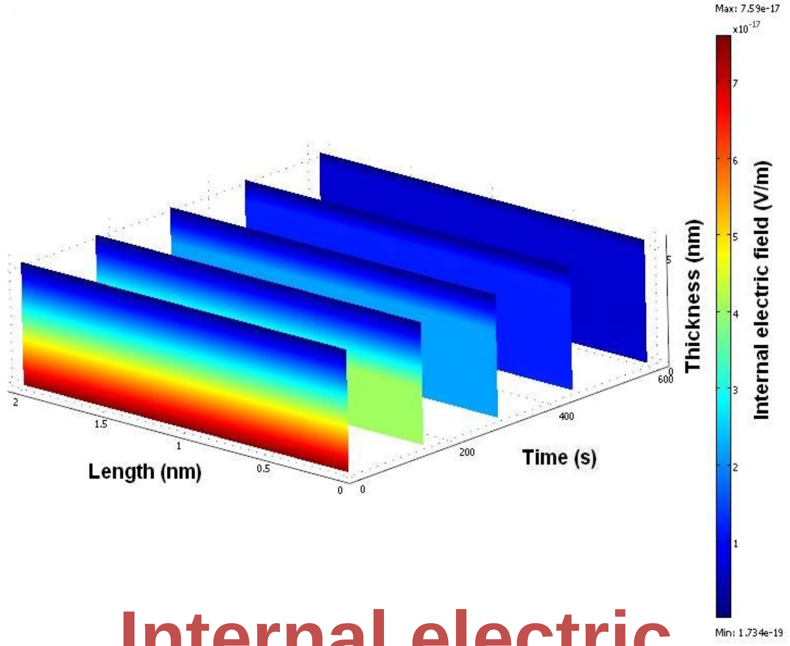
- Coupling of FP+DD by Poisson-equation

$$\Delta \cdot \psi = -\frac{e^-}{\varepsilon_0 \cdot \varepsilon_r}(n - N)$$



Drift-Diffusion simulations VG>0 V



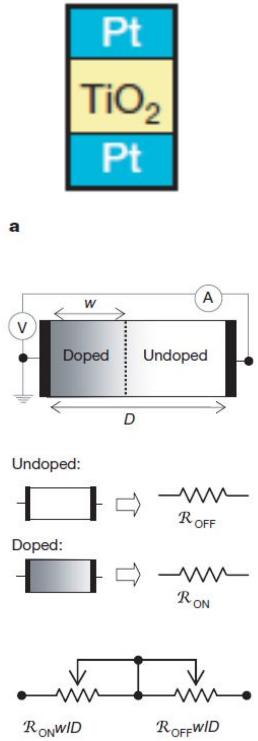


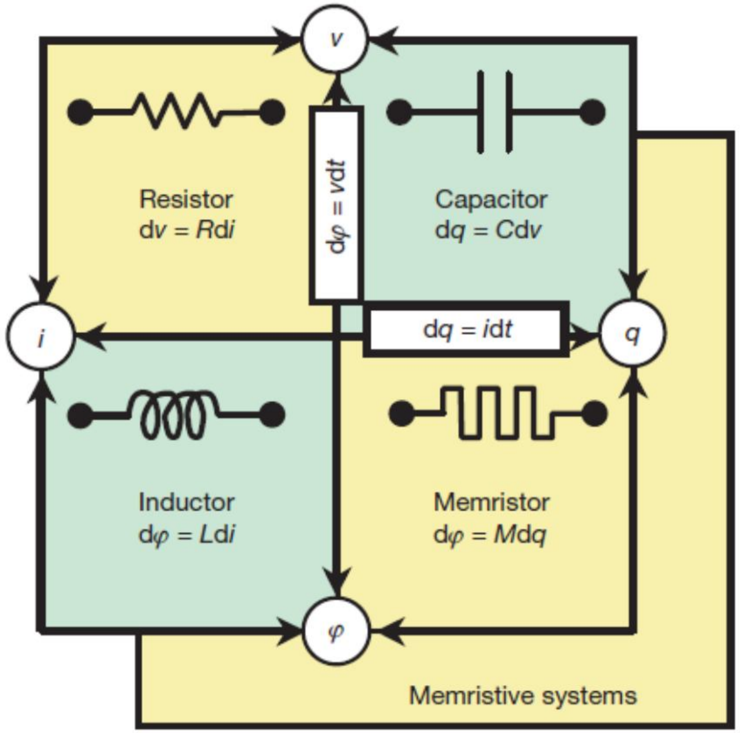
Normalized vancies distribution.

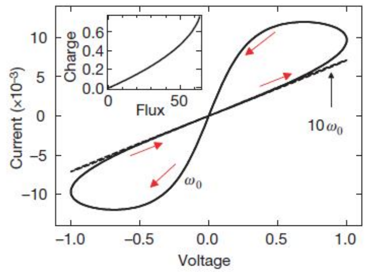
Internal electric field distribution.

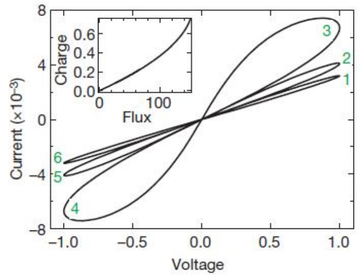
Memristor

D.B. Strukov, Nature, Vol. 453, pp. 80-83, 2008.









Summary

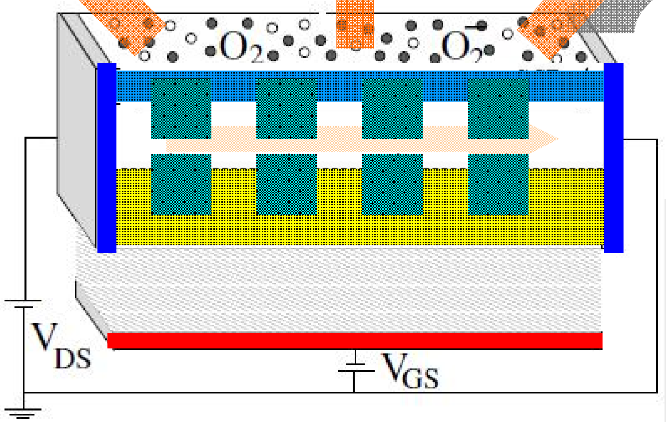
EAE Adsorption

Co-Adsorption

Gas-adsorption

EAE+Gas-adsorption

Drift



Doping

Continuty equation

Poisson-equation

Electric Potential

Standard Halbleiter-Theorie

	N_D	I_{DS}	SCR_1	SCR_2
Standard	✗	✓	✗	✓
Gasad.	✗	✓	✓	✗
Co-Ads.	✗	✓	✓	✗
EAE	✗	✓	✓	✓
Drift	✓	✓	✓	✓

Outline

- Motivation
- Classic semicondutor theory
  - Ideal „Bulk“
- Semiconductor theory at the bulk edge
  - Semiconductor surface-gas interactions
- One example of device operation
  - Electro-adsorptive effect
  - Thin film transistor
  - Drift of vacancies (dopants)
- Remarks

Conclusions

- Charge transport in semiconductors can be accurately described
  - Semiconductor theory

- IMPORTANT: Not only the material properties are important in the charge transport also geometry!

- Gas-solid intereaction influences the transport of charge in solid due to a charge transfer process.
  - Atomistic intereaction at the surface modifies the conductivity of solid
  - It can be accurately described by an extended semiconductor theory
    - ⌘ Wolkenstein theory
    - ⌘ Charge transfer (band bending)
  - IMPORTANT! Check if this model is applicable directly to your system
    - ⌘ Grain size effects (Debye lenght)

- Note: In catalytic processes the surface change continuosly:
  - Dynamic phase transitions

AC FHI

Greatings

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Thank you!

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