
Physik-Neubau ab 2014


Ulmenstraße
Saarplatz
neue Achse
Campus Südstadt
21.10.2011
Bibliothek und
Mensa

15.1.2012
Foto nicht maßstäblich!

Projekt *Unipadd* Stand 16.1.2012

PZ: Problemzonen, EP: Entwicklungsprojekte

PZ1: Konzept für die Anbindung des Innenstadtbereichs. Evtl. zum Schröderplatz und dann Friedhofsweg
PZ2: Anbindung neue Achse des Südstadtcampus über Hundertmännerbrücke zum Saarplatz
PZ3: Umgestaltung Ulmenstraße: Breiter Radweg erforderlich
PZ4: Breiter Fahrradweg Campus Schillingallee zur Parkstraße
PZ5: Saarplatz Fahrrad-gerecht umgestalten

EP1: Hauptpadd Universitätsplatz - Saarplatz
EP2: Südstadtcampus - Ulmenstraße und Schillingallee

lecture 26.1.2012

we had so far:

- more to metallic bonding, jellium model
- photoelectron spectroscopy

today

- optical properties of metal clusters
- clusters in strong laser fields
- few more interesting features of cluster physics

end of the lecture

PES on coinage metal clusters

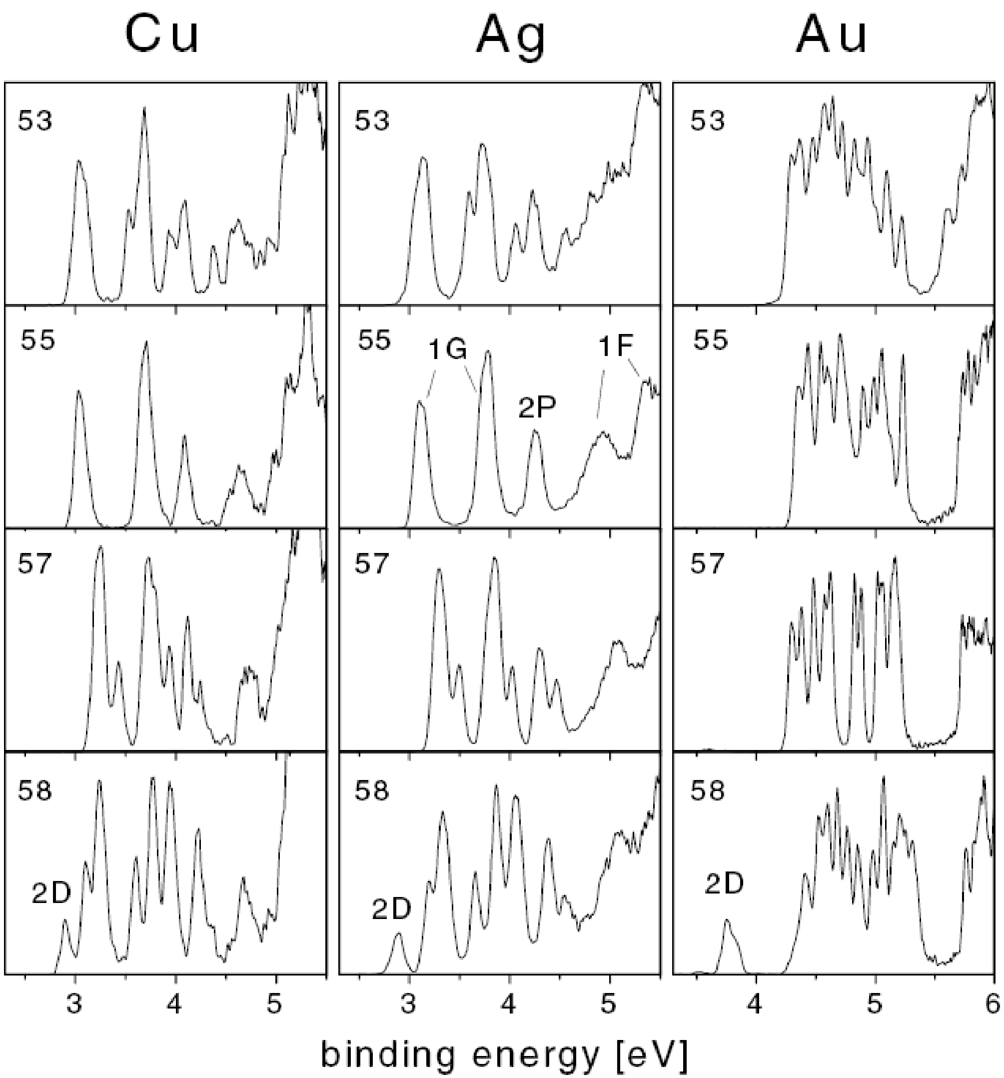
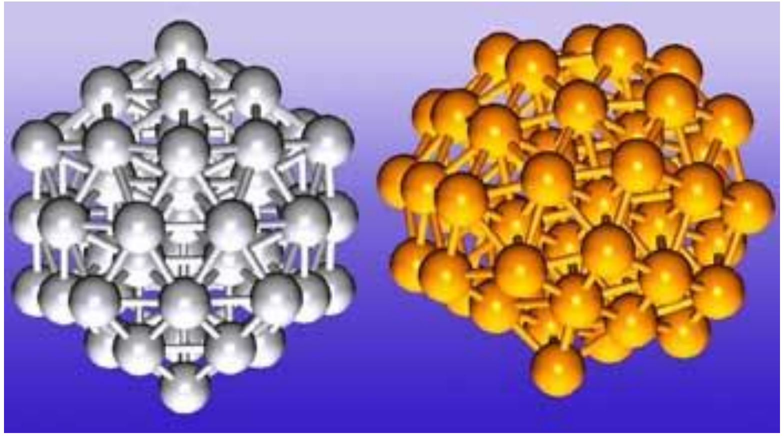


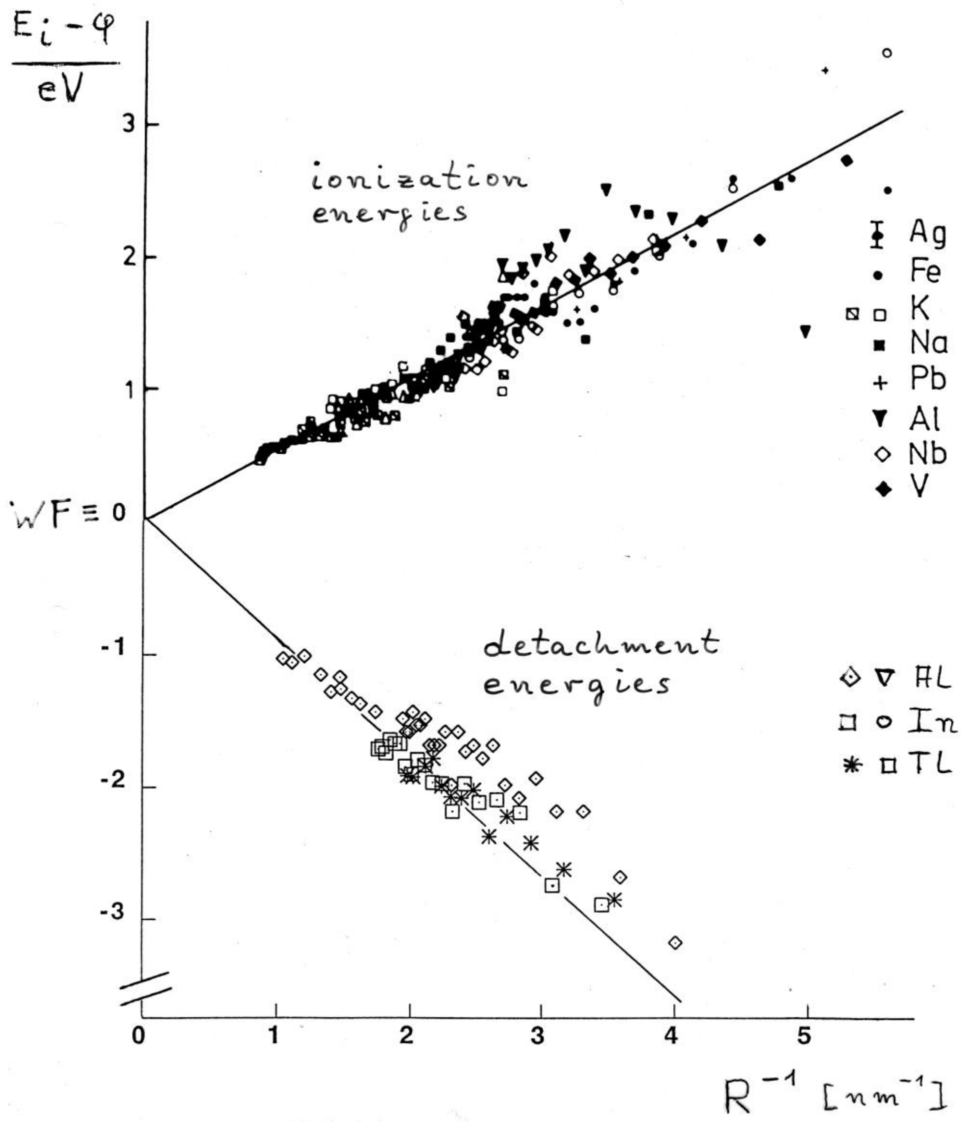
FIG. 1. Photoelectron spectra of Cu_n^-, Ag_n^-, and Au_n^- (n = 53, 55, 57, 58) obtained at a photon energy of 6.42 eV.



chemically similar systems may have similar PE spectra. Exception: Gold, due to relativistic effects

Hannu Hakkinen, Michael Moseler, Oleg Kostko, Nina Morgner, Margarita Astruc Hoffmann, and Bernd v. Issendorff, PRL 93 093401(2004)

experimental values for the IP and the EA



Meiwes-Broer, Appl. Phys. A55 (1992) 430; also Bergmann/Schaeffer

generally:
- the IPs decrease with increasing N
- the EAs rise with increasing N

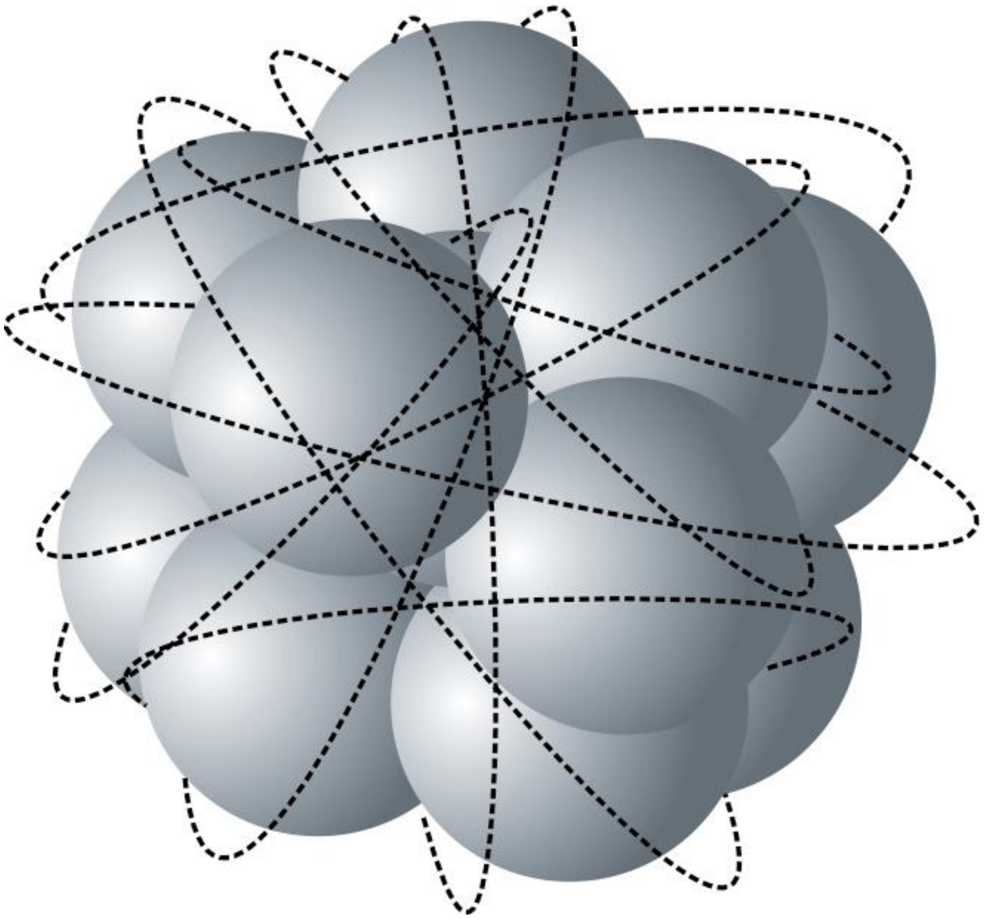
Parametrization:

$$IP(R) = WF + \alpha \frac{e^2}{R}$$ with α = 3/8 .... ½

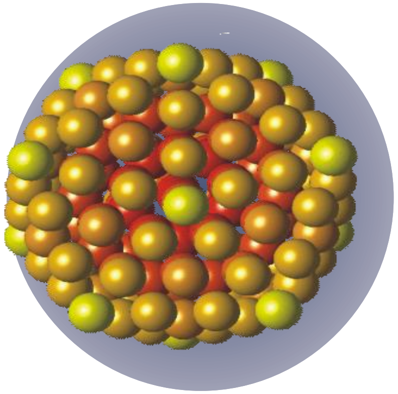
$$EA(R) = WF - \beta \frac{e^2}{R}$$ with β = ½ 5/8

α = β = ½ corresponds to the charging energy of a jellium sphere, deviations arise from QM exchange and correlation

optical properties through confined electrons

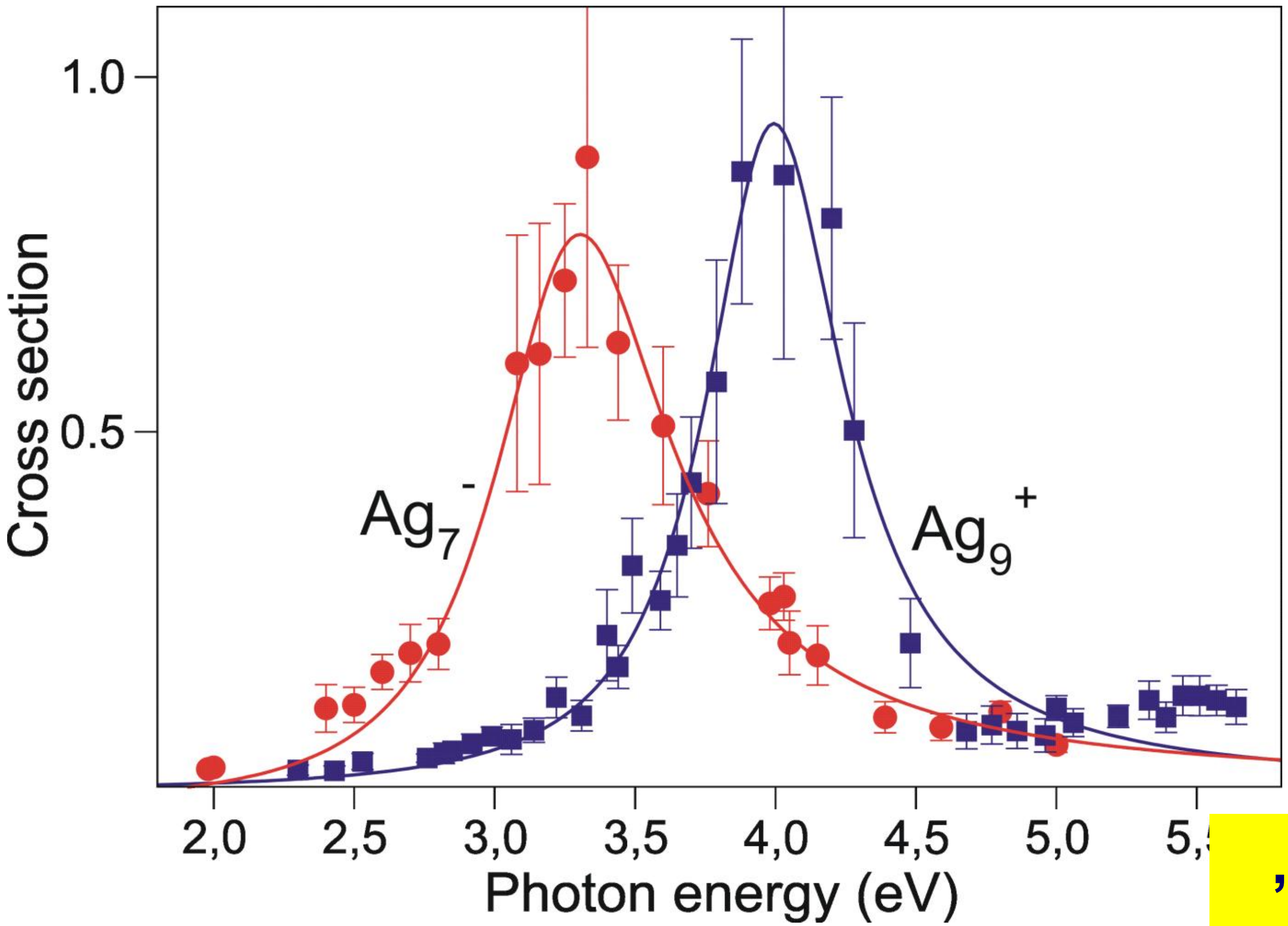


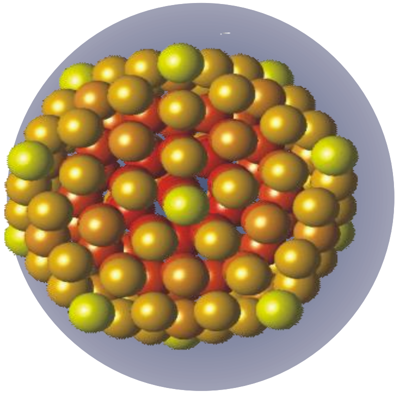
... schwingen gemeinsam im Laserlicht wie ein angetriebenes Pendel



Jellium artwork

photofragment spectroscopy on mass-selected silver clusters





„cold plasmon“

only in metals with delocalized electrons

Tiggesbäumker et al., Phys. Rev. A 48, R1749 (1993); Chem. Phys. Lett. 260, 428 (1996)

also see: Kreibig and Volmer, Springer

Optical properties of small particles

Lycurgus cup (400 AD)

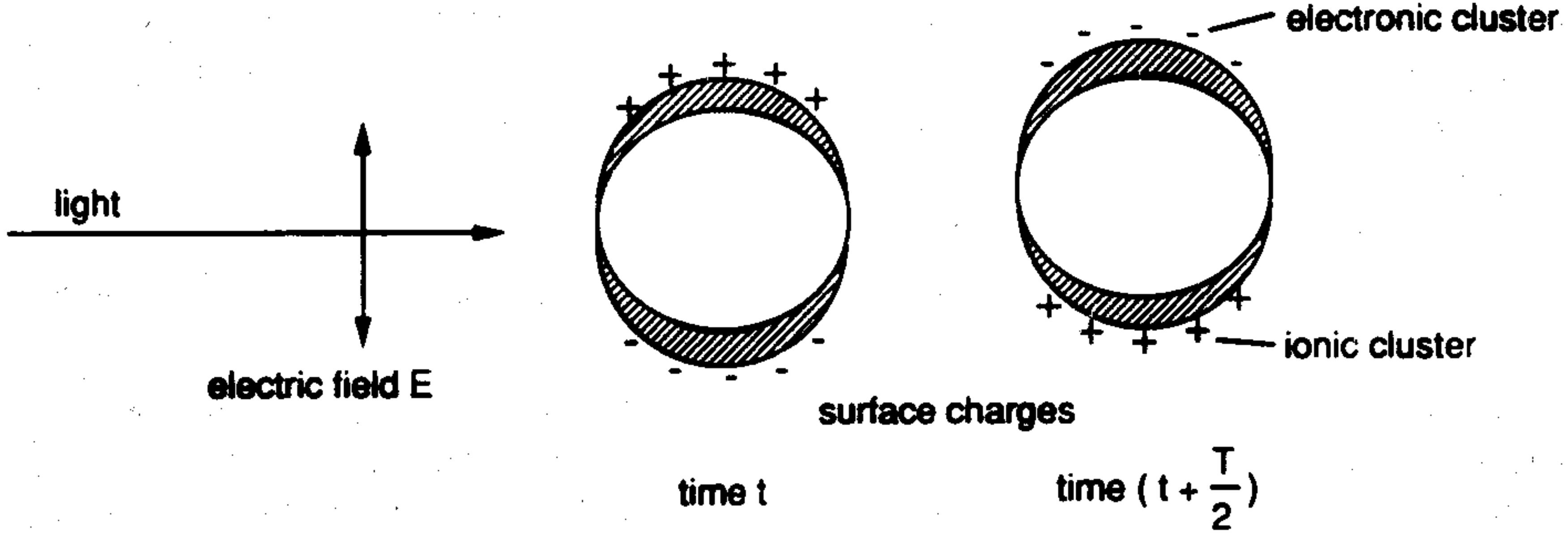
King Lycurgus is dragged into the underworld by Ambrosia

color due to a small amount of gold and silver particles

“Labors of the Months” (Norwich, England, ca. 1480).

The ruby color is probably due to embedded gold nanoparticles

physics behind the optical properties: excitation of the electron cloud in a small particle



plasmon ecxitation

- Since the ion cores are much heavier than the electrons the positive charges stay more or less at their position when a light field interacts with the particle
- The electric field of the light moves the electron cloud out of the positive background
- Below the resonance the electrons move with the direction of the electric field
- At resonance the movement of the cloud has a pi/2 phase difference to the electric field.
- This is called the plasmon resonance
- Above resonance the phase is 180 degrees

Gustav Mie 1868-1957

born in Rostock

Gustav Mie, Beiträge zur Optik trüber Medien, speziell kolloidaler Metallösungen Ann. Phys. 25, 377 (1908)

absorption and scattering of light by small particles

good book: C.F. Bohren and D.R. Huffman Wiley, NY 1983

$$\omega_{Mie} = \left(\frac{1}{3} \frac{e^2 n_o}{\varepsilon_o m_e} \right)^{1/2}$$

n_0: electron density

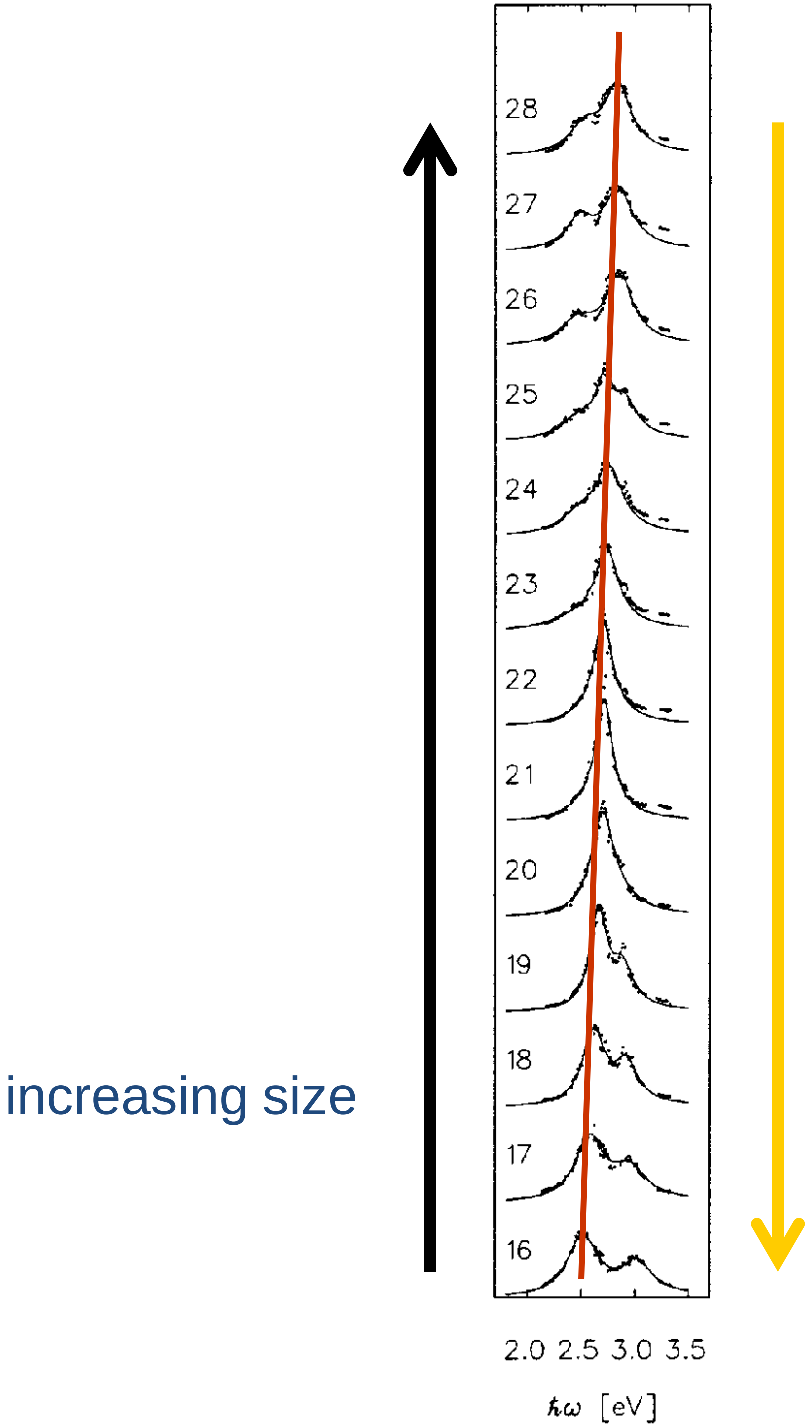
plasmon frequency of a small particle

-Everything gets simplified when the size of the particle is much smaller than the wavelength of the light

$$\omega_{Mie} = \left(\frac{e^2 N_e}{\alpha \cdot m_e} \right)^{1/2}$$

static polarizability

- The relevant value is the size R compared to the wavelength λ of the light, i.e. R/ λ



trend with small alkalies

For decreasing cluster size the plasmon energy shifts to lower values.
This is called the red-shift.

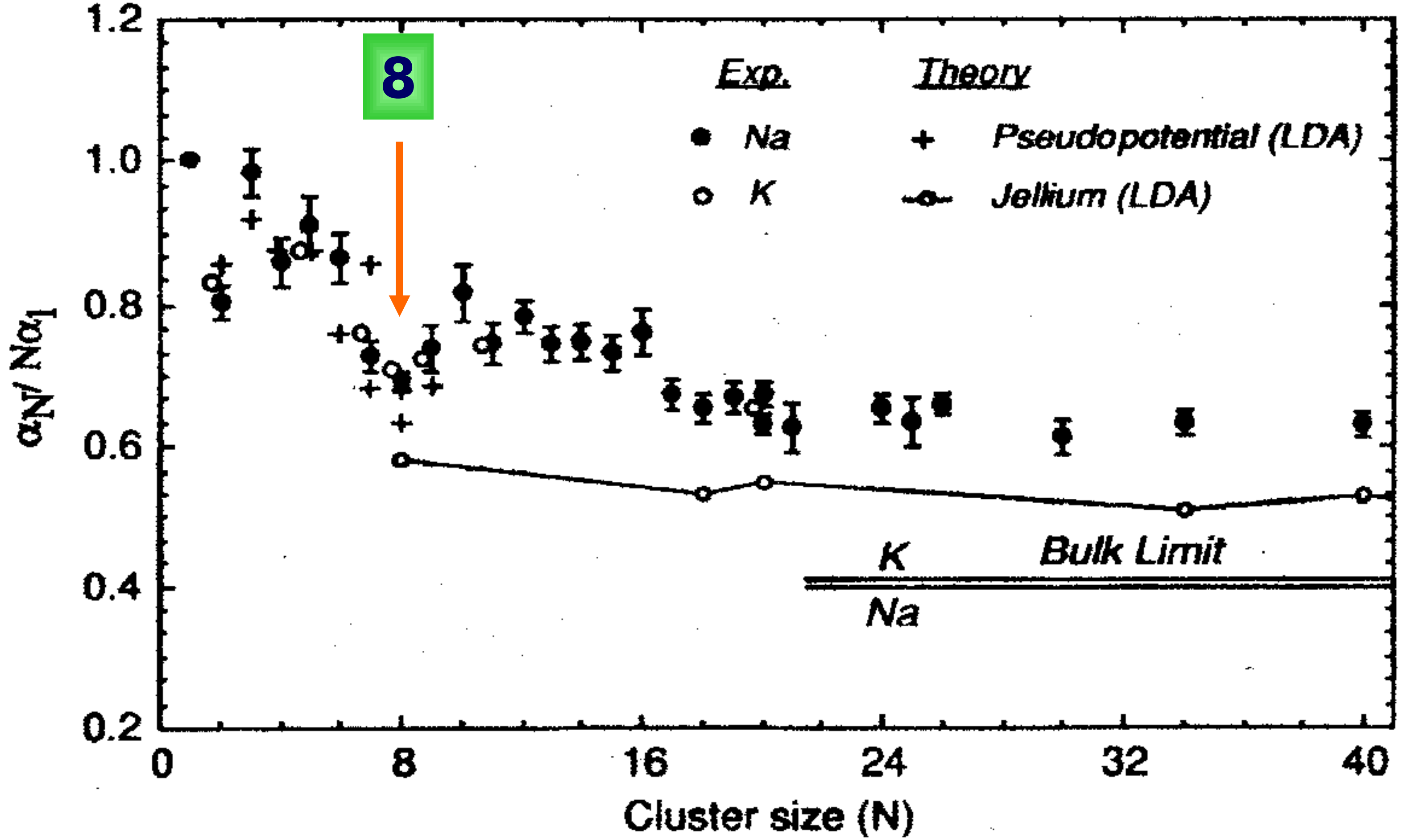
Reason: influence of the spil-out

$$\omega_{Mie} = \sqrt{\frac{n_{bg} e^2}{3m\varepsilon_o}} = \frac{\omega_p}{\sqrt{3}}$$

Mie – Plasmon

or: increasing polarizability

polarizabilities of alkali clusters

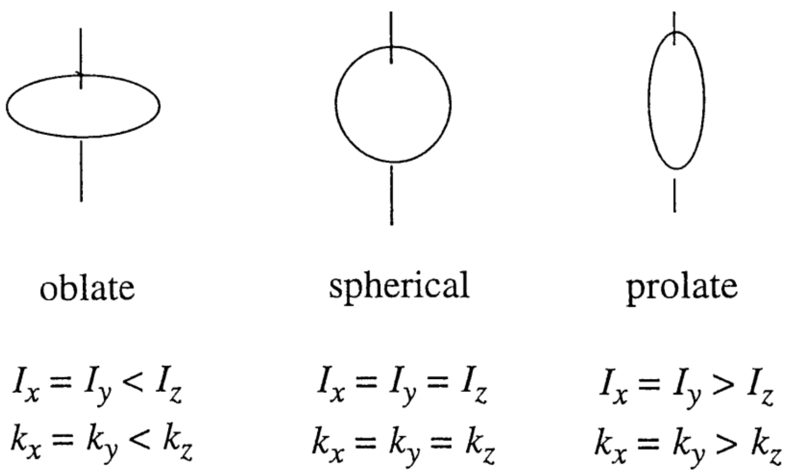


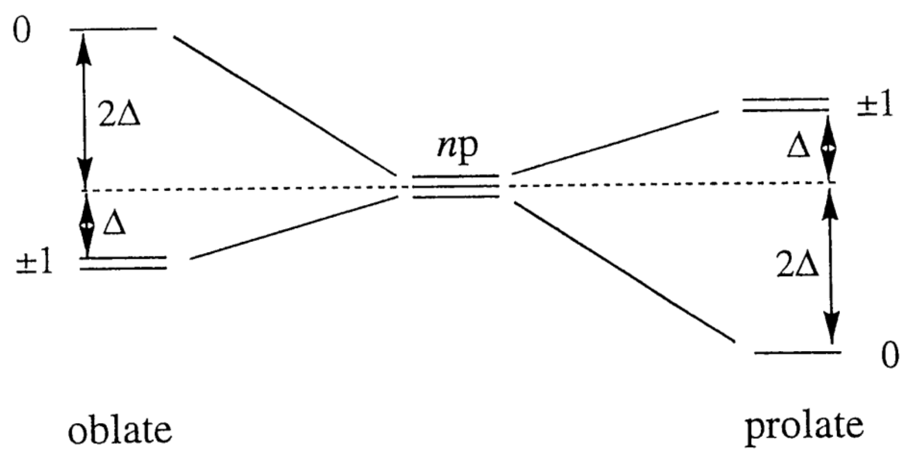
Knight et al. Phys. Rev. B 31, 2539 (1985)

not all clusters are spherical, see last lecture

ellipsoïdal shell model

Unlike an atom, the positively charged background can deform !

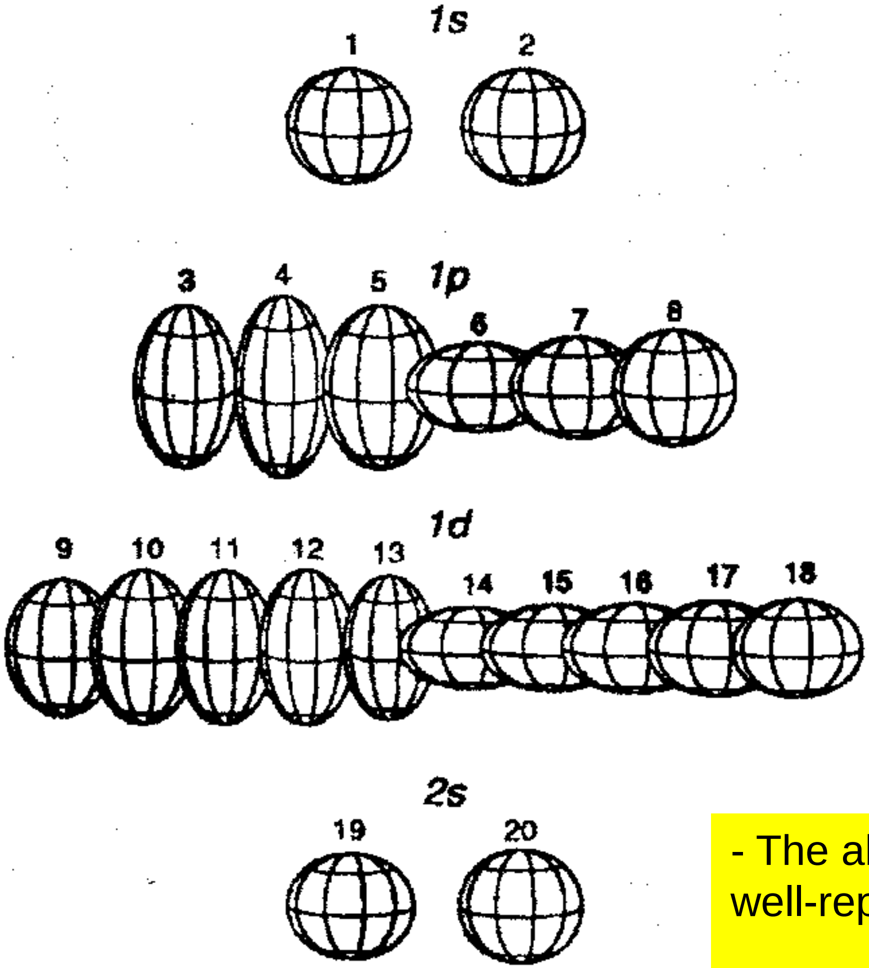




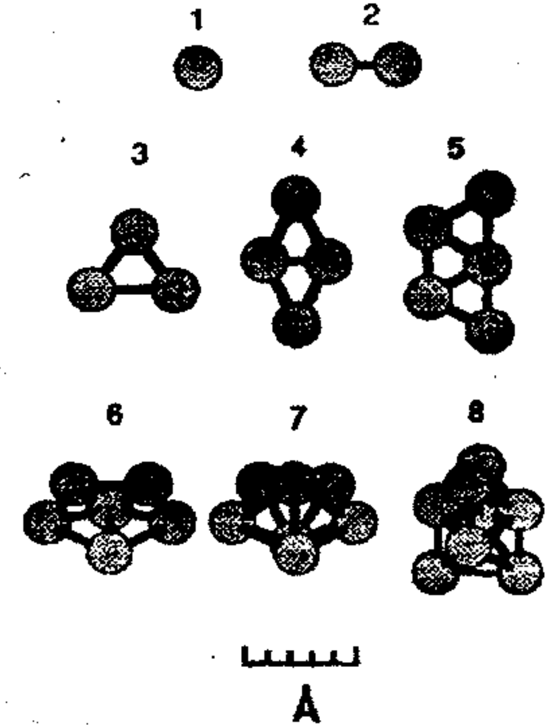
Nielsson model

not all clusters are spherical

Ellipsoidal shell model



Quantum chemical calculations

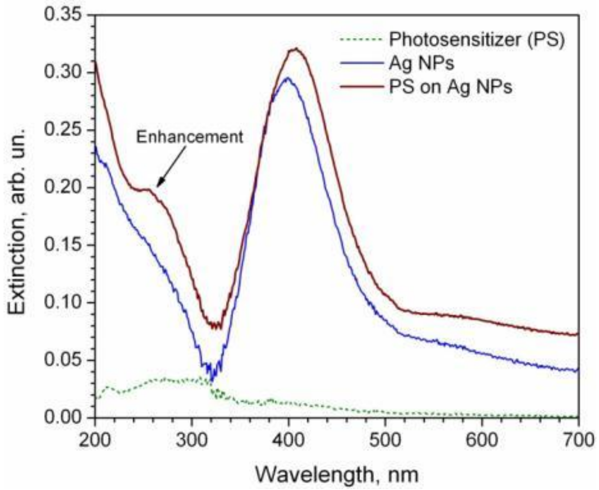


- The ab-initio cluster structures are in first order well-reproduced by the ellipsoidal shell model.

- One can calculate the plasmon profiles more easily on the basis of this model.

how do we measure the plasmon profiles?

- with high-density targets we could measure the optical absorption



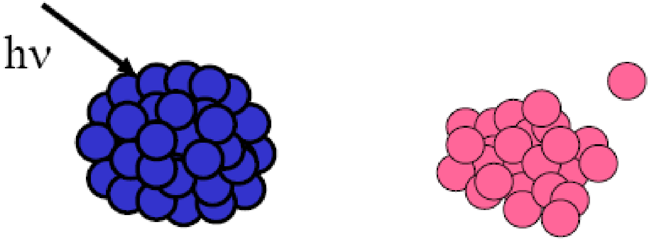
this example shows the intense Ag plasmon peak, seen by photoabsorption of Ag clusters deposited onto glass (trace: Ag NP's).
Recent work by Ingo Barke, H. Hartmann, Stefan Bartling, et al.
Aim of the project is to study the role of Ag clusters to accelerate photocatalytic processes.

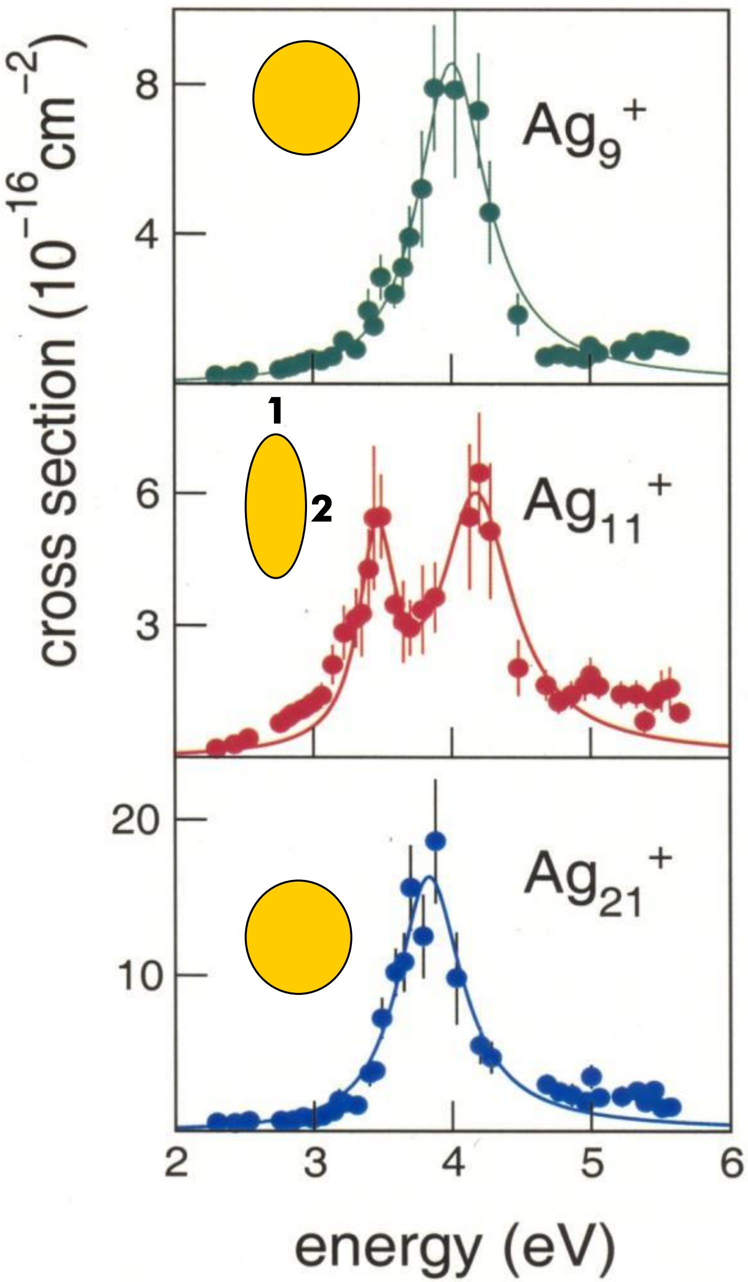
- in cluster beams we use photofragment spectroscopy:

<u>step 1</u>: observe the intensity of the not exited (parent) cluster in the mass spectrometer

<u>step 2</u>: excite the cluster with a given photon energy, measure the decrease of the parent signal or the appearence of daughter signal

<u>step 3</u>: repeat, for different photon energies





Tiggesbäumker et al.

role of deformation

- In cluster ions the number of electrons is N-1, thus closed shells appear at M_{N+1}^+

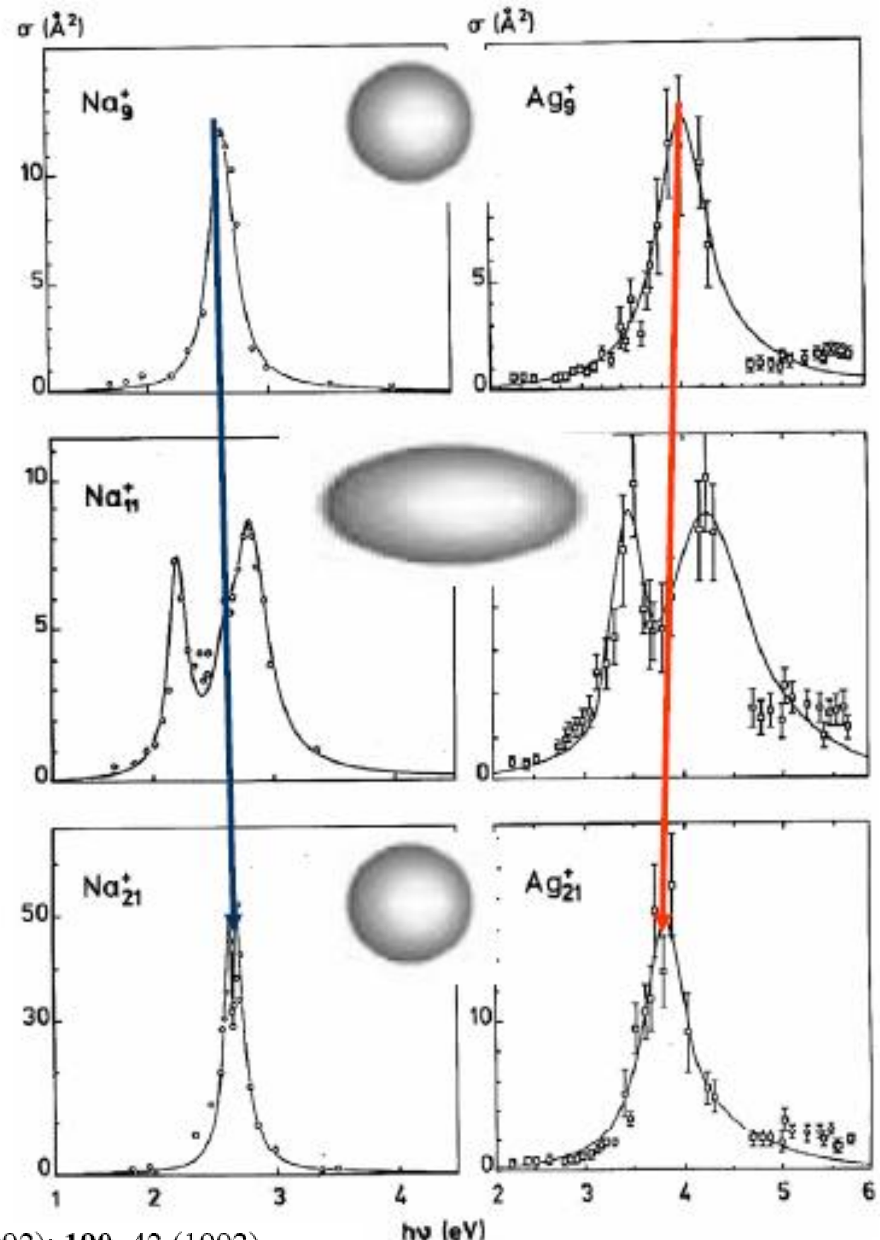
- Ag_9^+ and Ag_{21}^+ are closed-shell clusters and only a single absorption line appear in the optical spectra.

- In Ag_{11}^+ the prolate deformation of the particle results in a splitting of the absorption into two components.

- According to the ellipsoidal shell model the lower energy peak should have half the oscillator strength.

compare the optical spectra

sodium



silver

Chem. Phys. Lett. **189**, 28 (1992); **190**, 42 (1992)

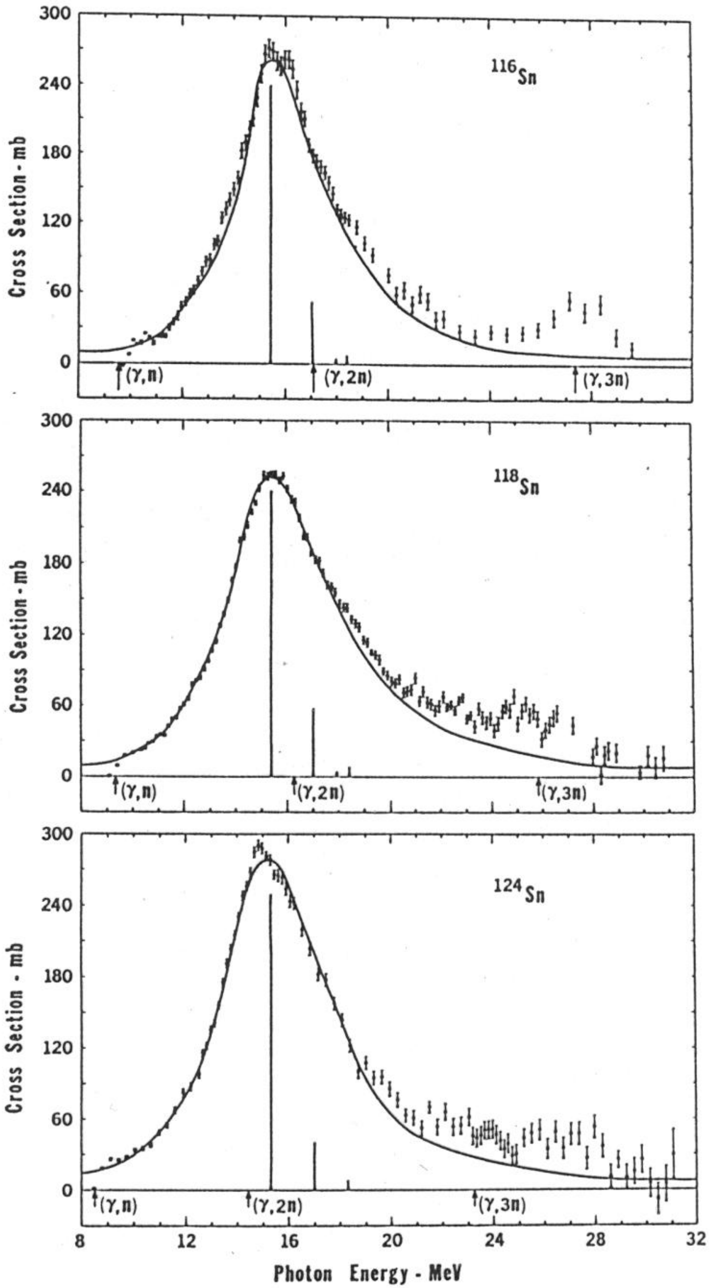


Fig. 1: Showing total photoneutron cross sections for different isotopes of tin, together with theoretical fits based on a dynamical collective model. (After reference 5)

similar:
giant resonances in atomic nuclei

- **electrons in metal clusters and the nuclear particles are fermions. Both oscillate in the confining potential**

- **therefore the same model can be used in order to describe the optical properties, e.g. the random phase approximation (RPA)**

- **closed-shell nuclei also show only a single absorption**

- **in the nuclei however the excitation energies are much higher, i.e., in the MeV range**

^{175}Lu nucleus

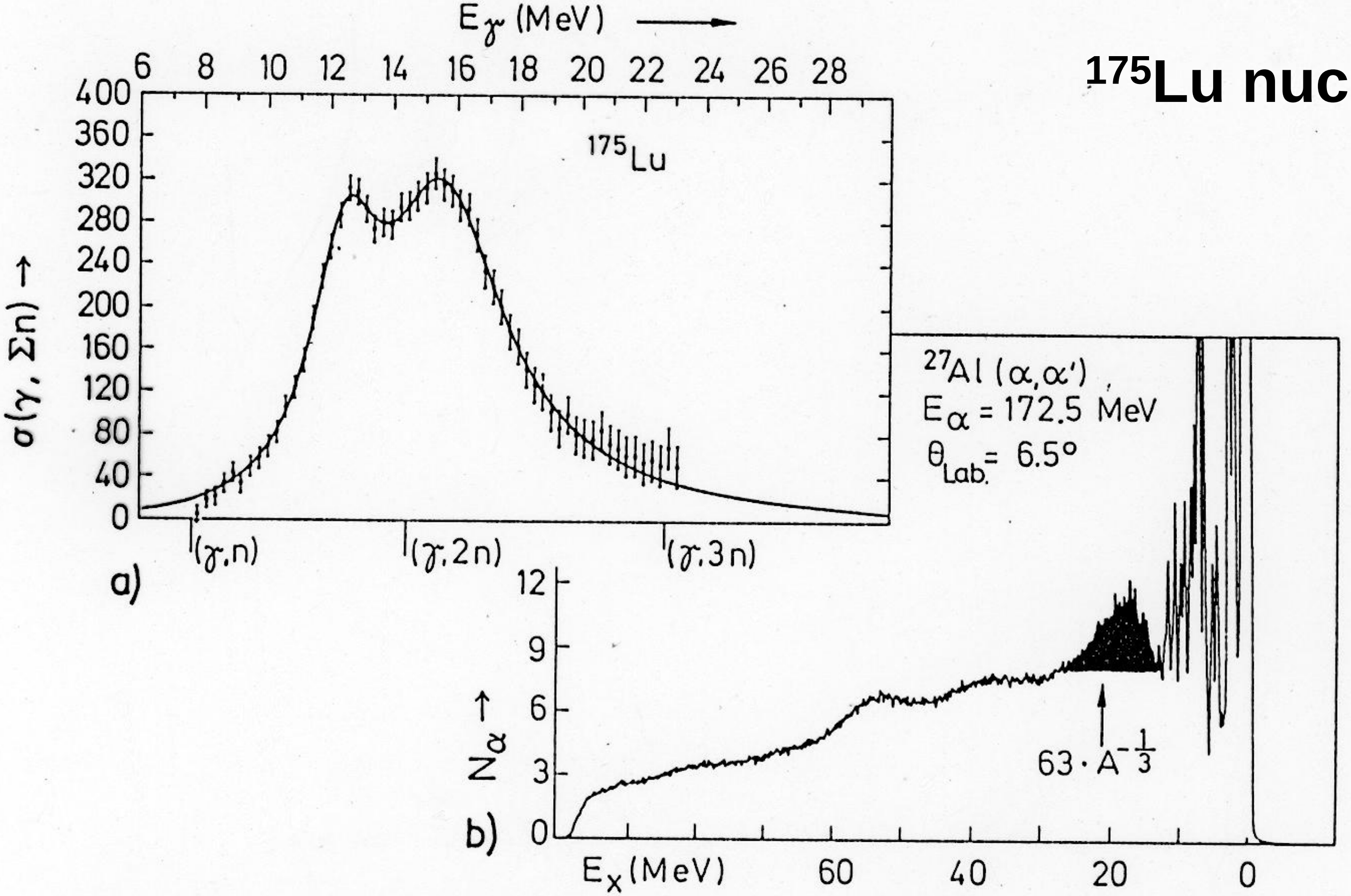
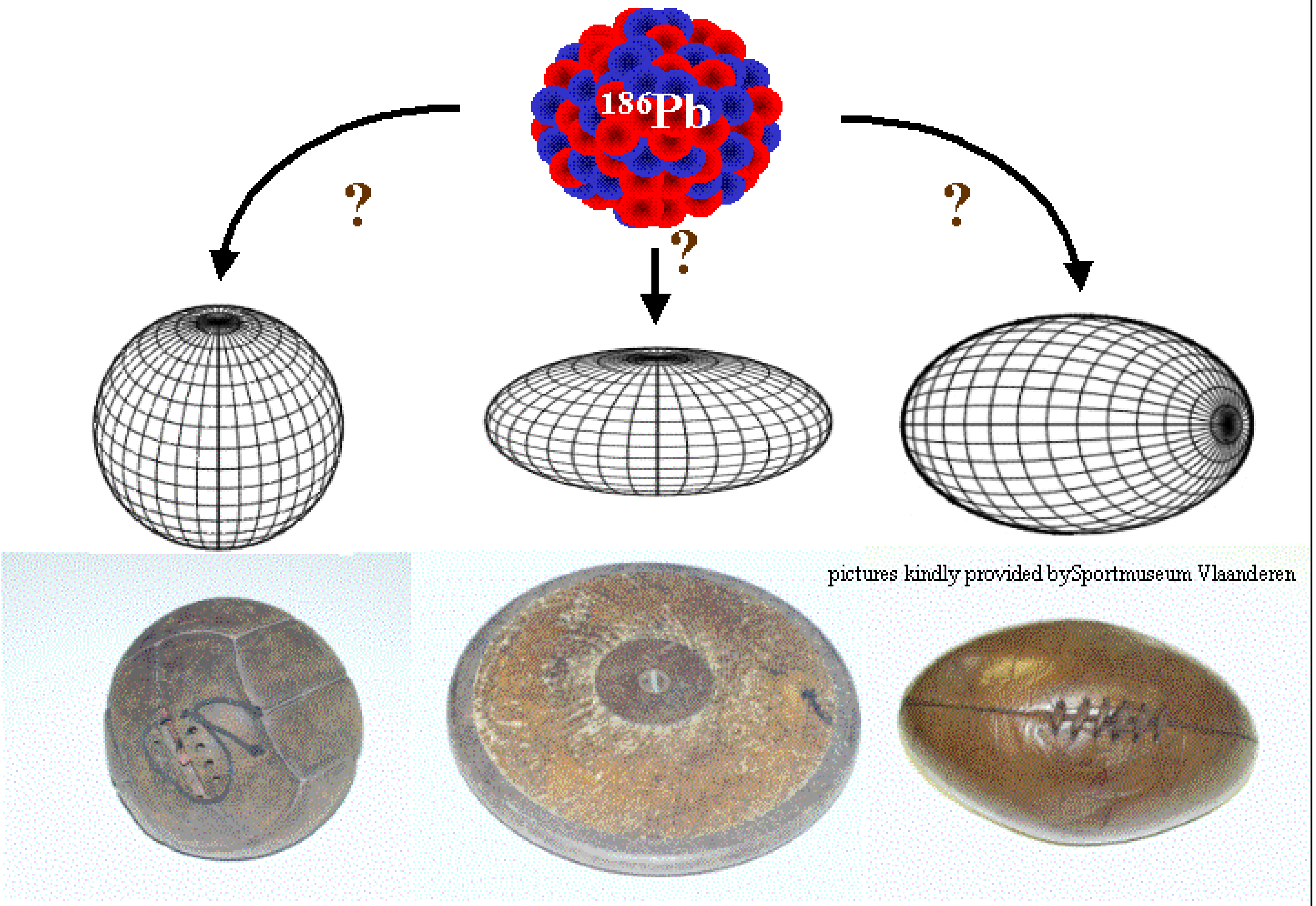


Fig. 99 Beispiele für Riesenresonanzen: a) In der Anregungsfunktion für (γ,n)-Prozesse an ^{175}Lu. Für deformierte Kerne ist der doppelte Höcker charakteristisch, da in der Deformationsachse eine andere Schwingungsfrequenz auftritt als senkrecht dazu [nach Ber 75a]; b) im Spektrum von unelastisch an Aluminium gestreuten α-Teilchen [Kis 76]

Mayer – Kuckuck p.237

nuclear shape isomerism : ^{186}Pb



Andreyev et al., Nature405 (6785) 430, 2000

Nobel prize in Physics 1963,
for their work on the nuclear shell model

Maria Goeppert Mayer
(1906-1972)

German-American

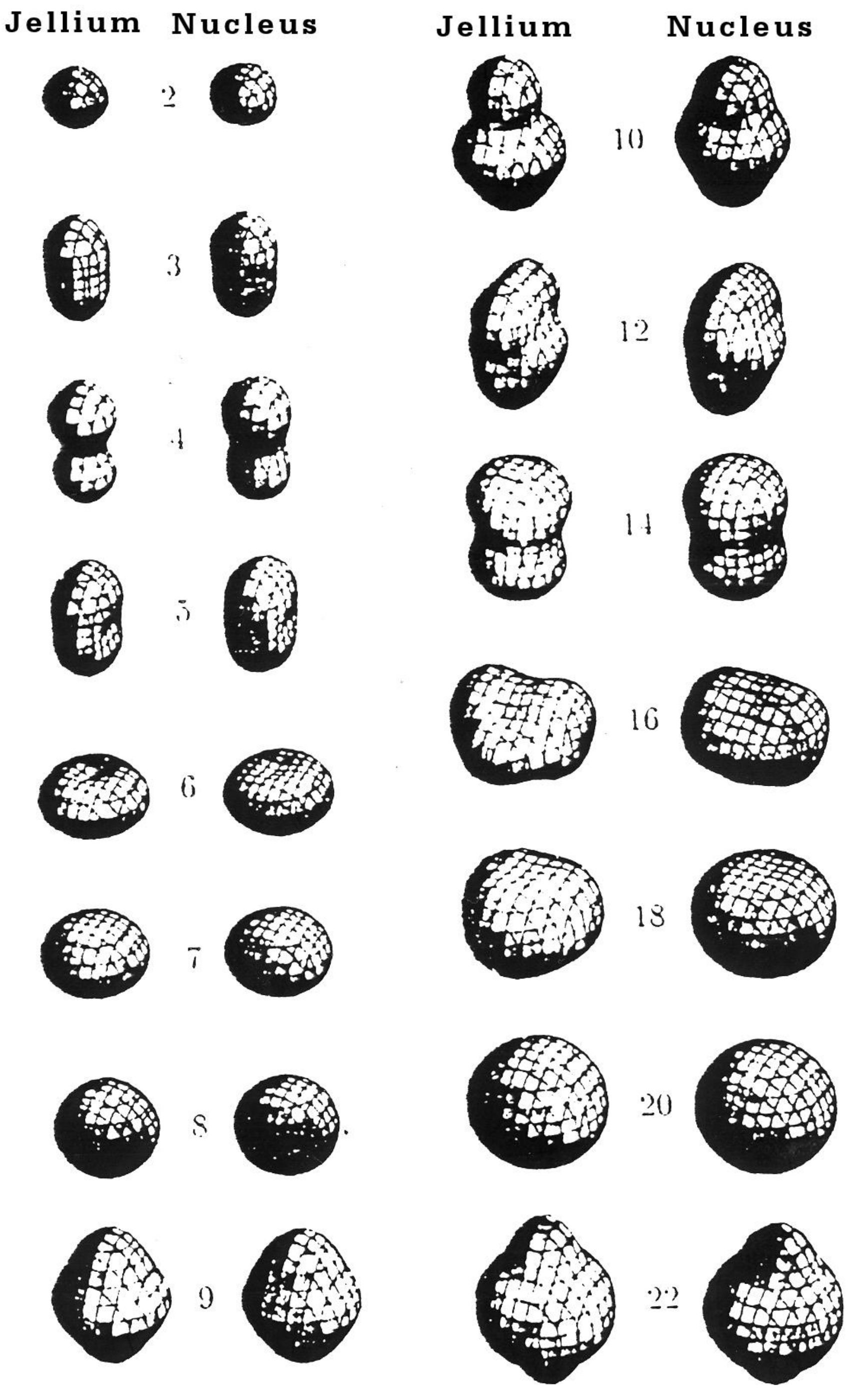
Hans Jensen
(1907-1973)

Hamburg

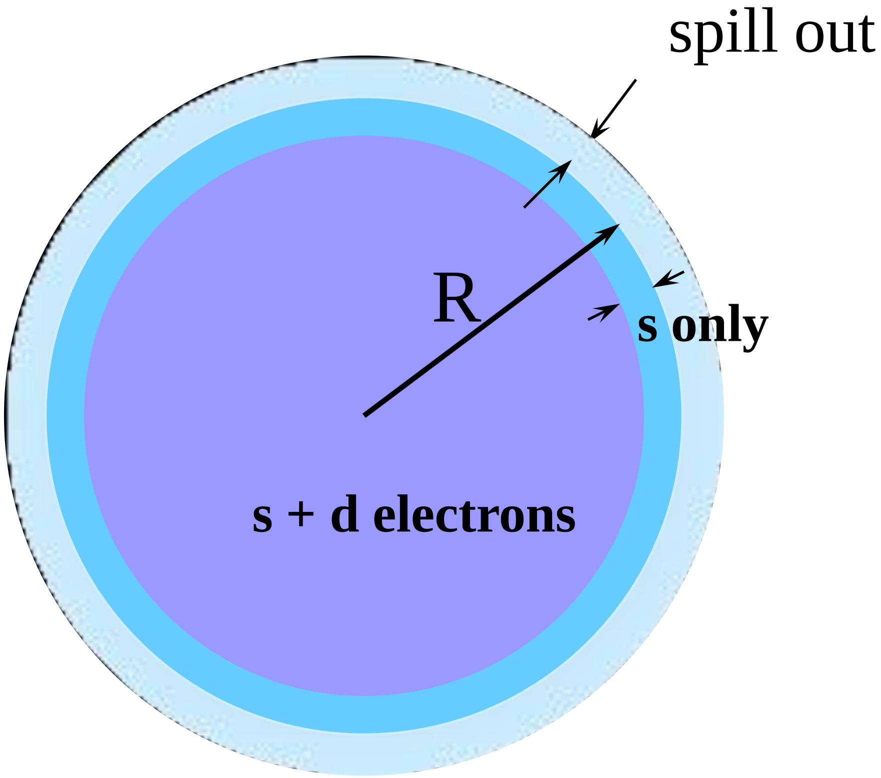
constant-density surfaces

for electron clusters and nuclei.

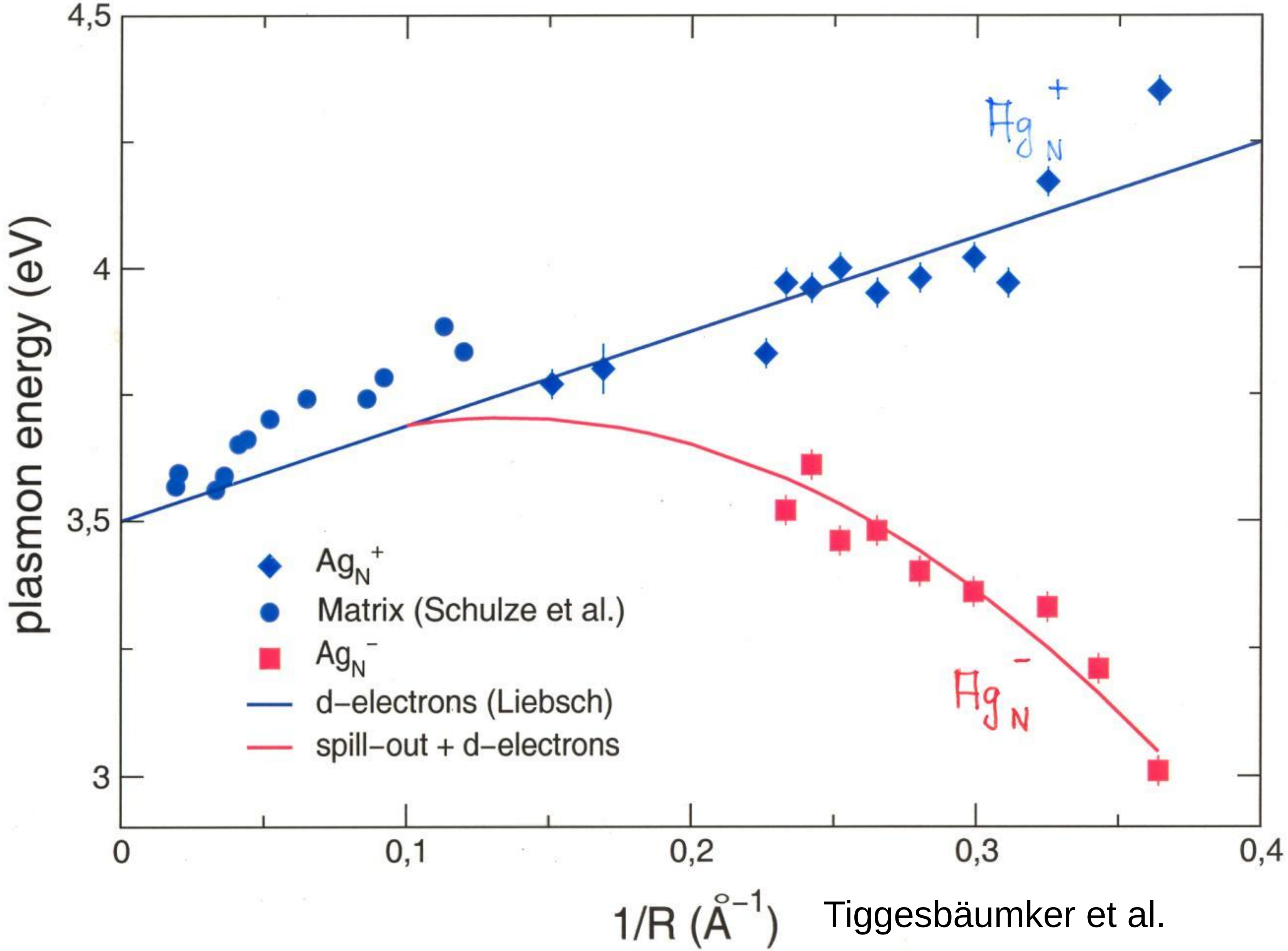
The density of the surface is 0.00125 atomic units (or scaled units for nuclei), which corresponds to 38% of the bulk density



Silver clusters: s- and d-electrons

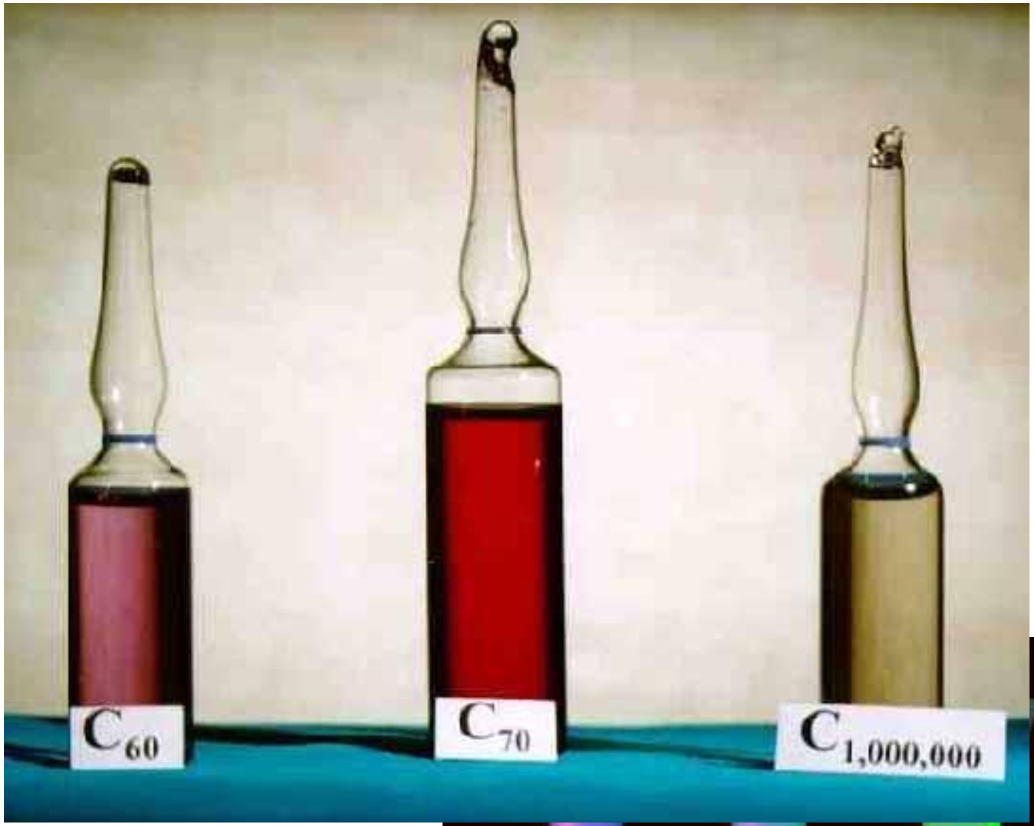


complex situation in Ag clusters



Tiggesbäumker et al.

Fullerenes and quantum dots

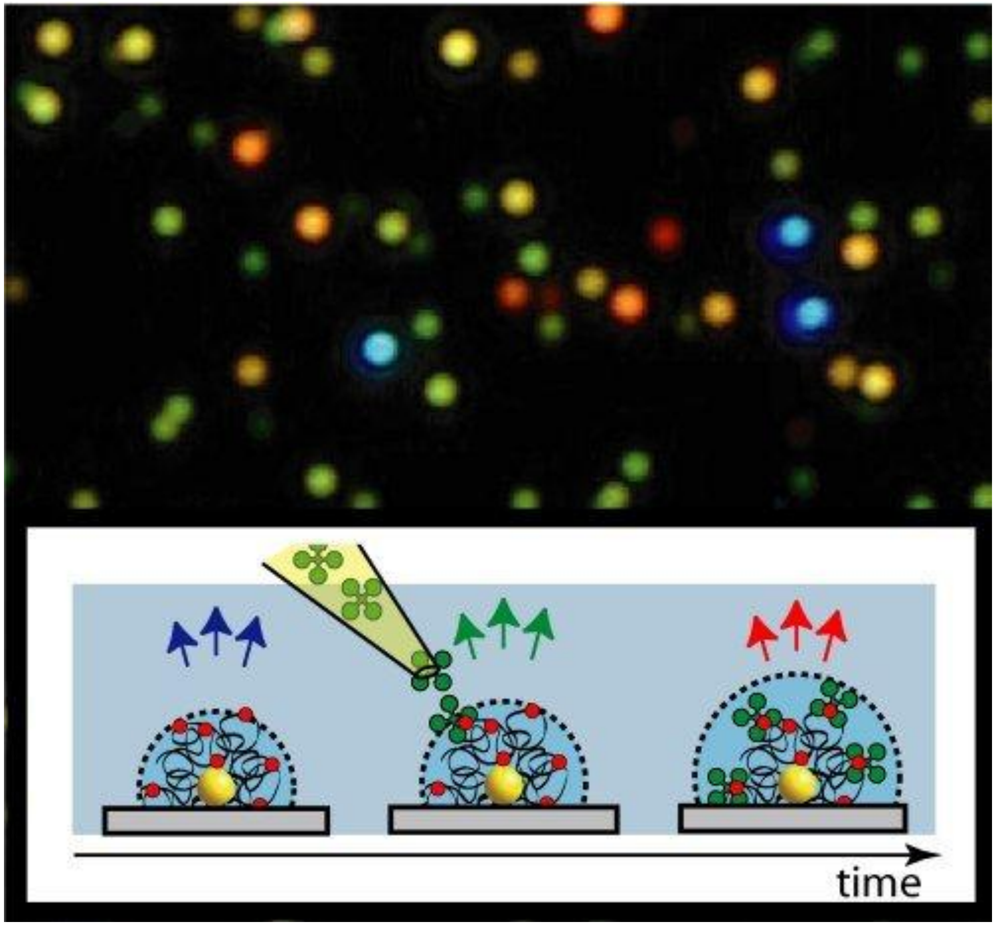


-Such absorption features can also be found in other small particles
- It is therefore not restricted to metal particles but occur also in particles like fullerenes or semiconductor quantum dots
- It turns out that we only need to know the optical properties of the material in order to calculate the optical properties

CdSe

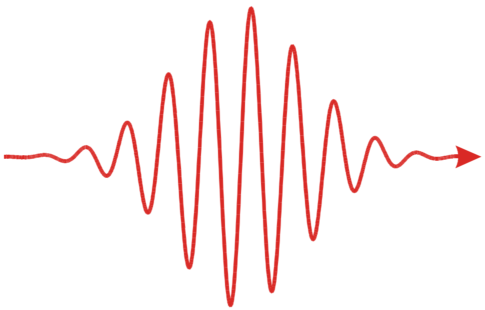
quantum dots

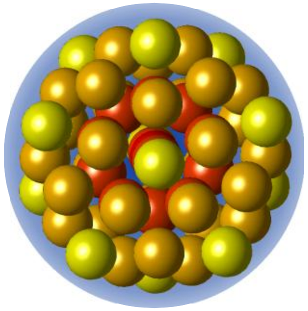
technical application as fluorescent labels

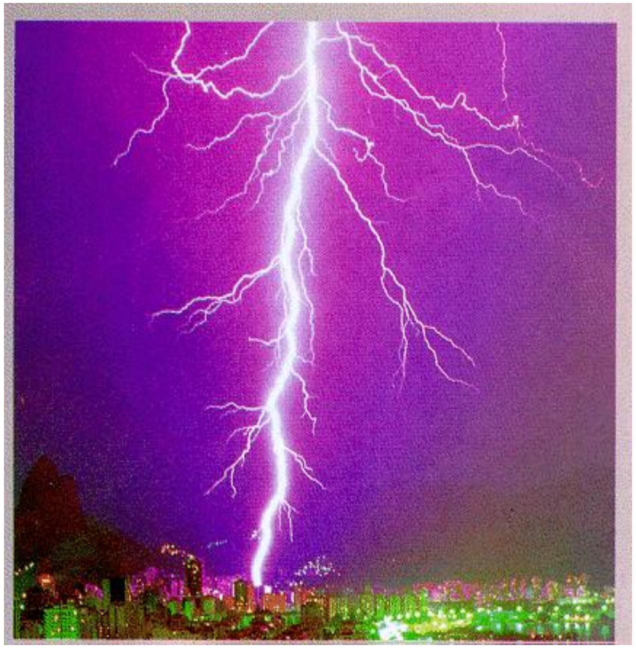


response to strong laser pulses: clusters act as strong nano antennas

Gigawatt laser pulse







Pulse length: 100 femto seconds
power densities up to 10.000 Gigawatts per cm^2

SFB 652

➔ minuature plasmas

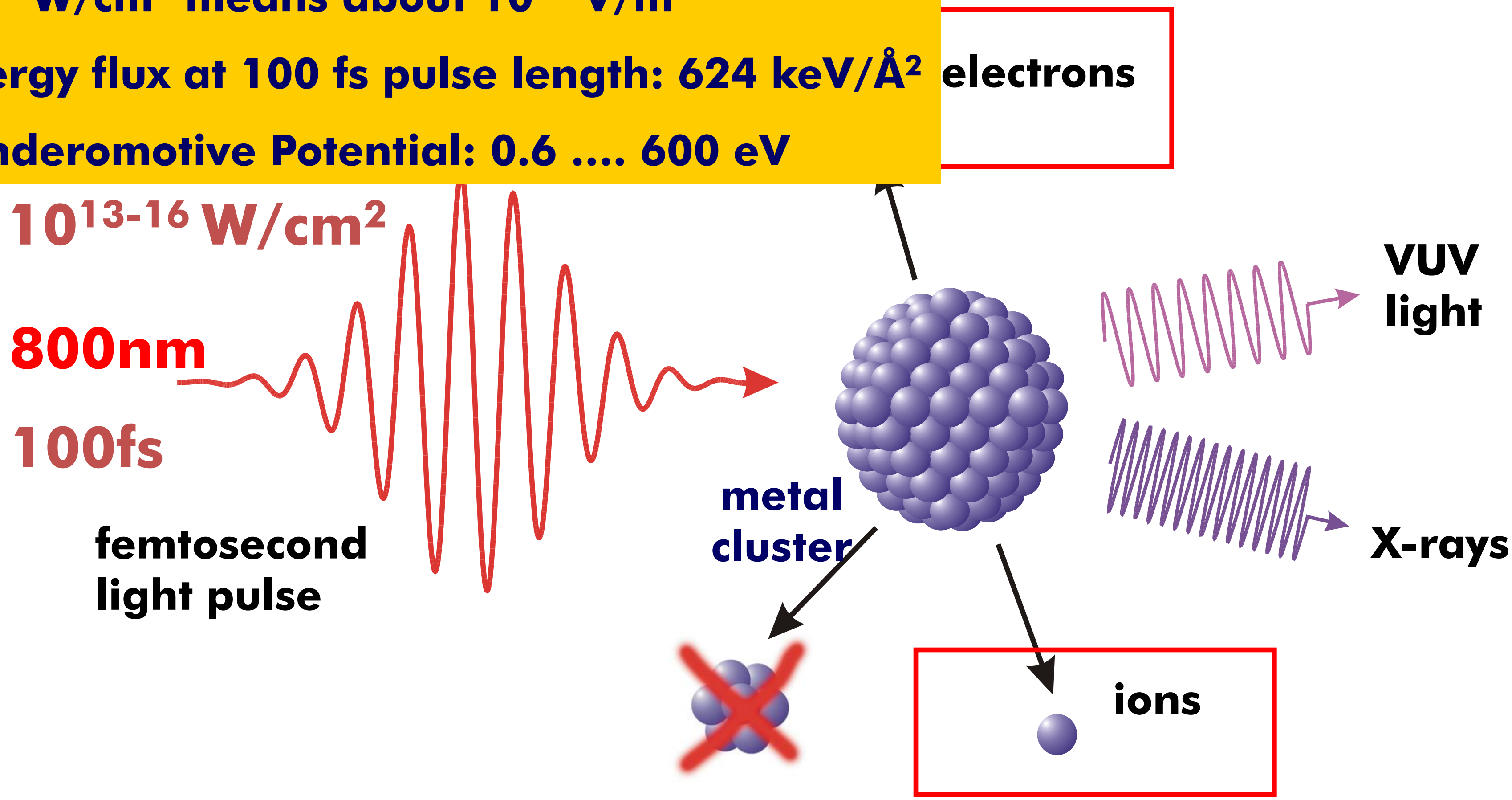
clusters in intense laser fields: nanometer-sized plasmas far from equilibrium

properties of the radiation field

10^{16} W/cm² means about 10^{11} V/m

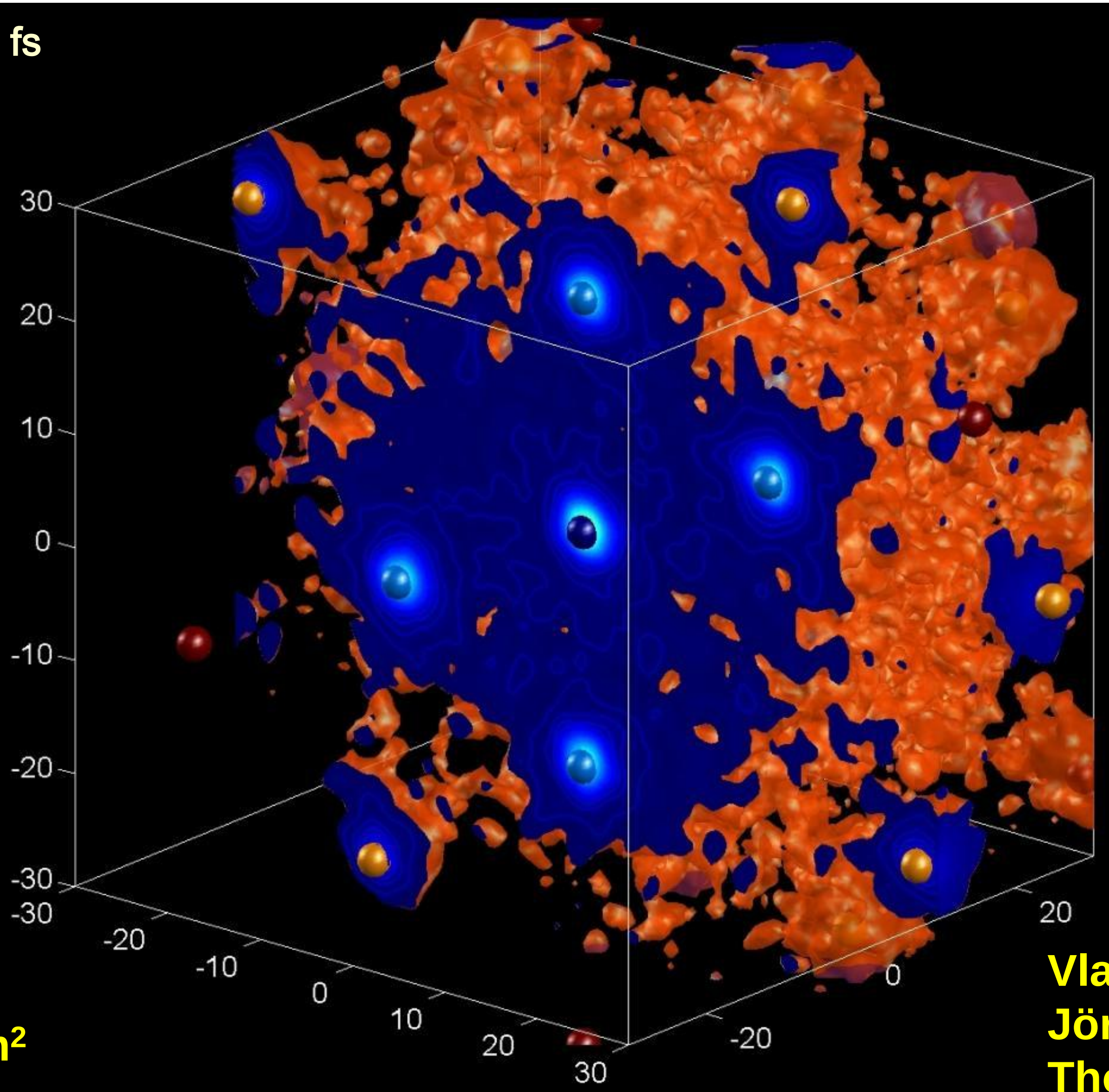
Energy flux at 100 fs pulse length: 624 keV/Å²

Ponderomotive Potential: 0.6 .... 600 eV



simulated Coulomb explosion Na_{55}

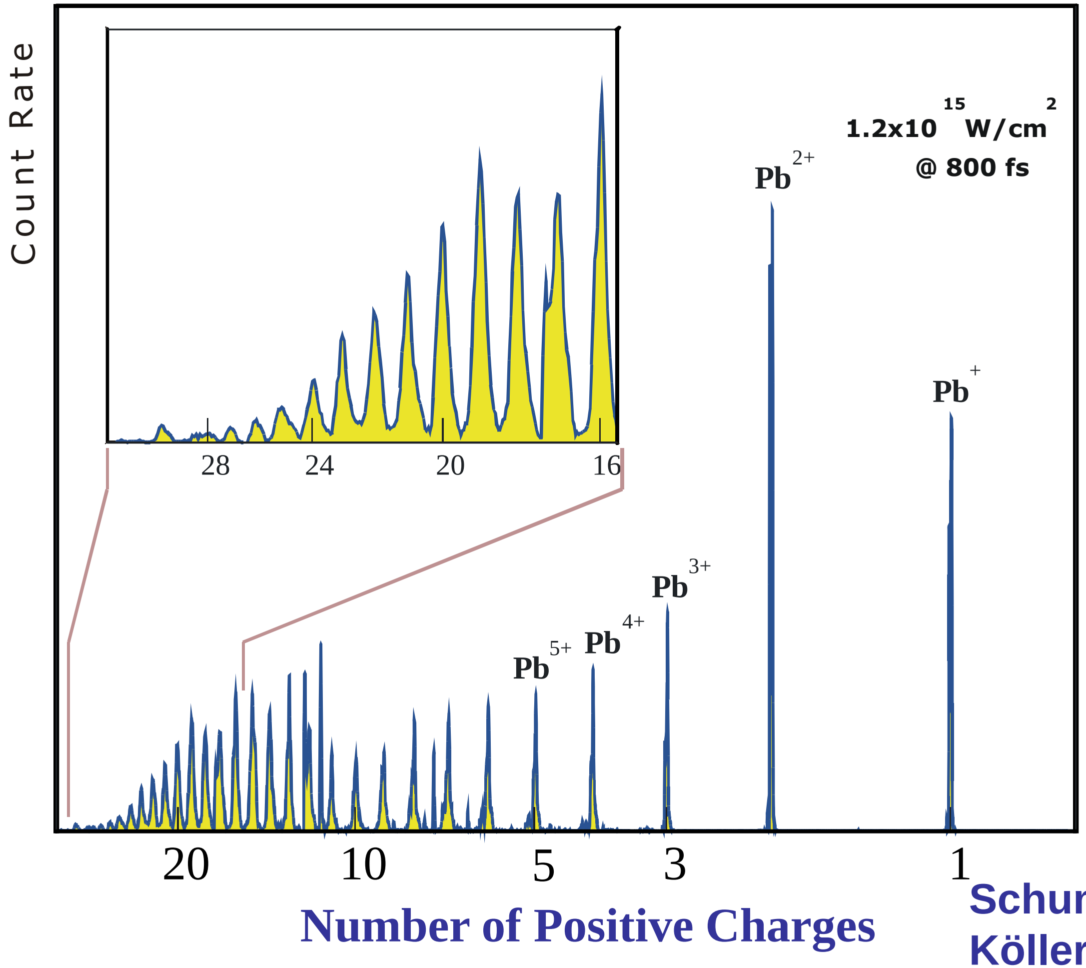
Modellzeit: 350 fs



E_{Photon} = 2.7 eV
t = 50 fs
I = 4×10^{12} W/cm^2

Vlasov-VUU
Jörg Köhn
Thomas Fennel

atomic charge distribution from Coulomb explosion



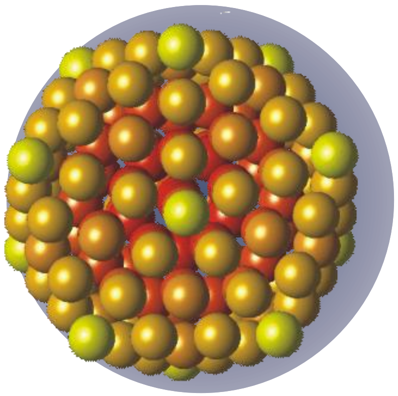
- Max. charge state z=30!
- Highest charging at 800 fs
- Max recoil energy E_{max} = 180 keV

Schumacher et al., EPJD 9, 411-414 (1999)
Köller et al., PRL 82, 3783-3786 (1999)

the following material has not been discussed in detail.
It demonstates further interesting issues
of cluster physics

mechanism for the high absorption

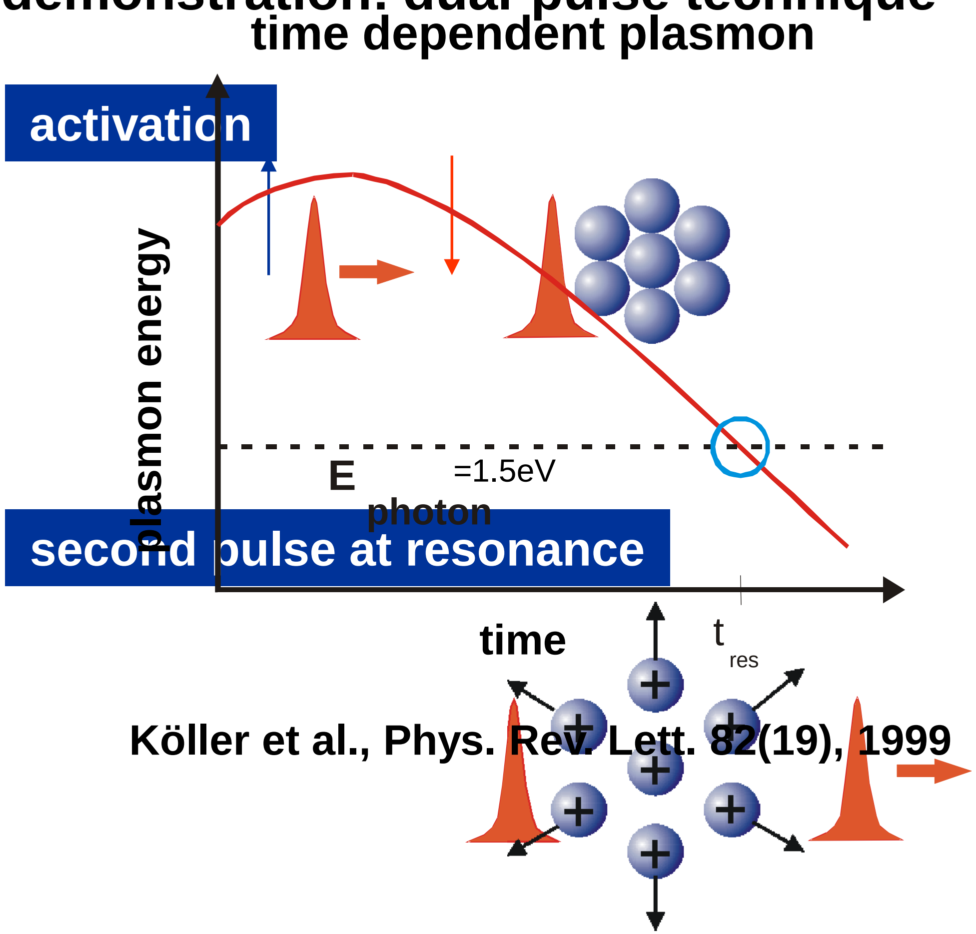
plasmon-enhanced ionization



$$\omega_{Mie} = \sqrt{\frac{n_{bg} e^2}{3m\varepsilon_o}} = \frac{\omega_p}{\sqrt{3}}$$

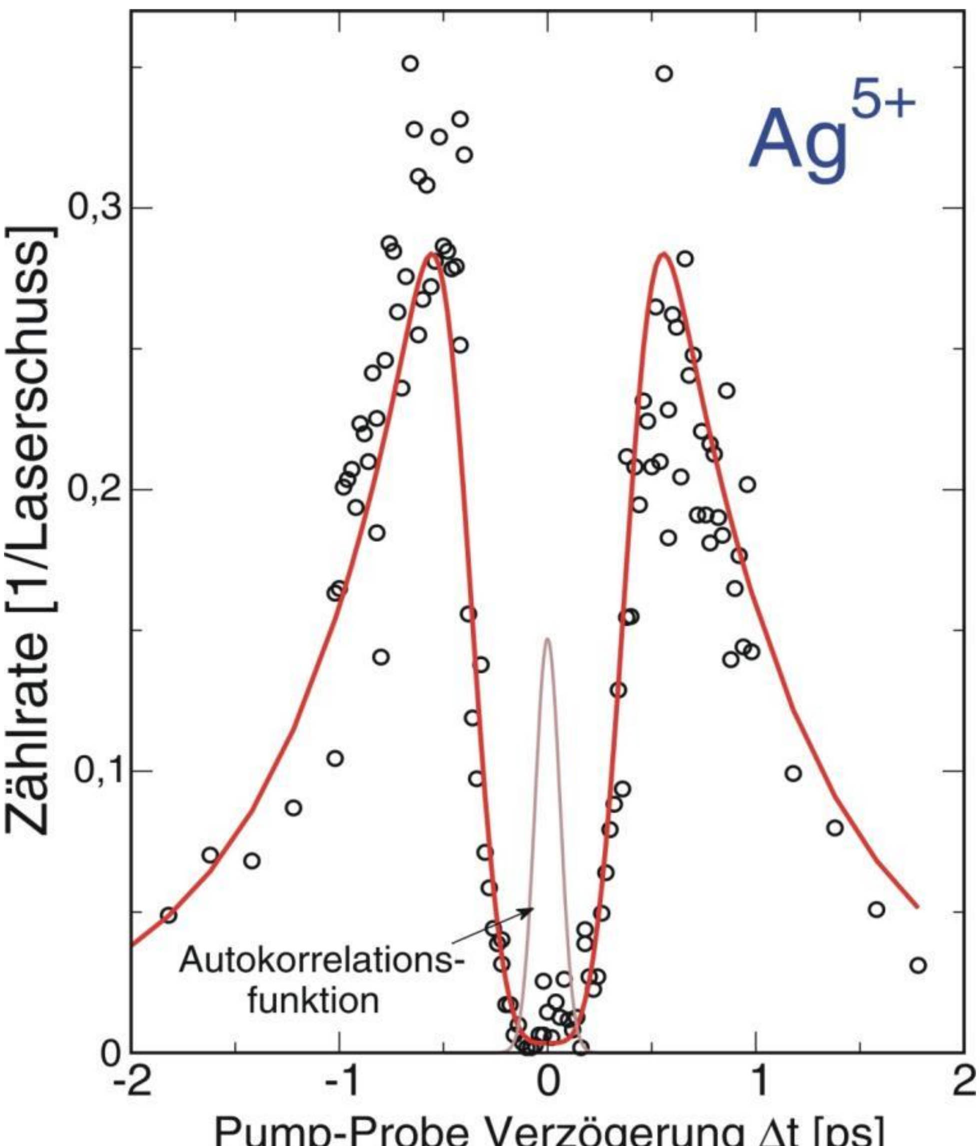
Mie – Plasmon

demonstration: dual-pulse technique

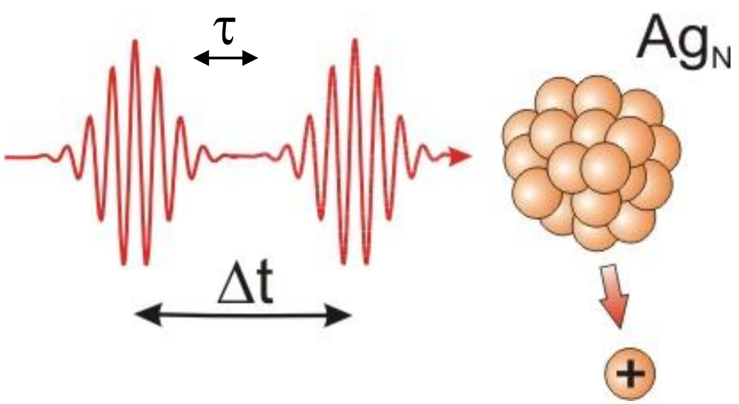


Köller et al., Phys. Rev. Lett. 82(19), 1999

dual-pulse excitation of Ag_N@He-droplet



delay



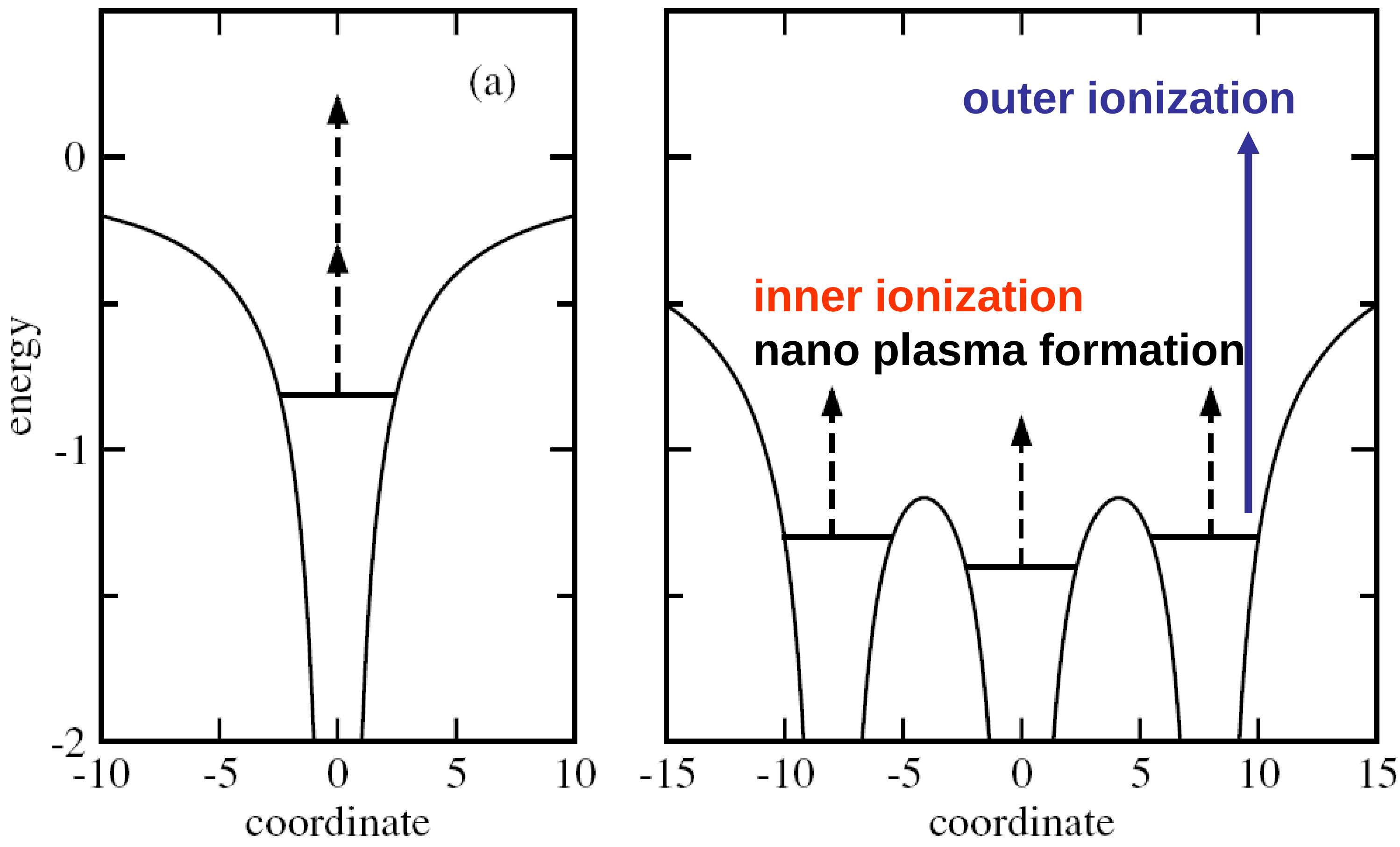
Ag_5^+

Laser parameters:

- λ = 800 nm
- τ = 120 fs
- I = 1.8 x 10^{13} W/cm^2

Döppner et al., Phys. Rev. Lett 94(13401), 2005

inner *vs.* outer ionization

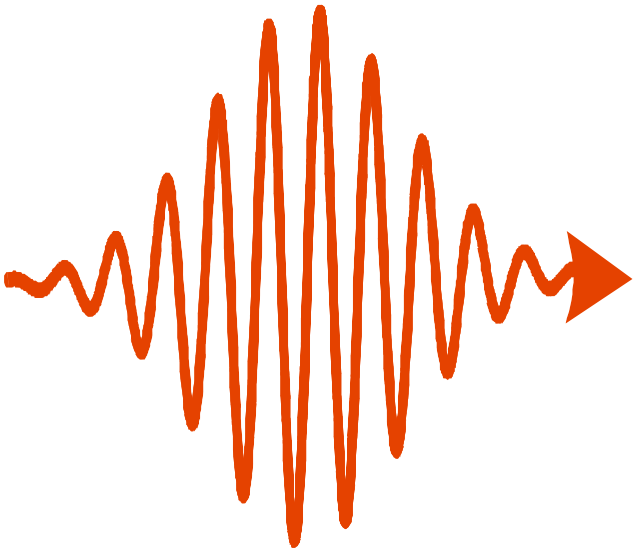


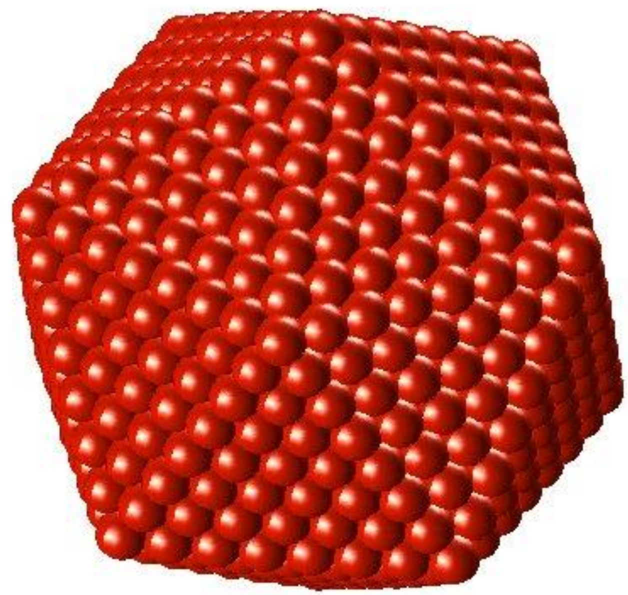
<u>several heating mechanisms</u>:
inverse bremsstrahlung
laser driven electron-ion collisions

after: Ch. Siedschlag and J M. Rost *PRL* **93** 043402 (2004)

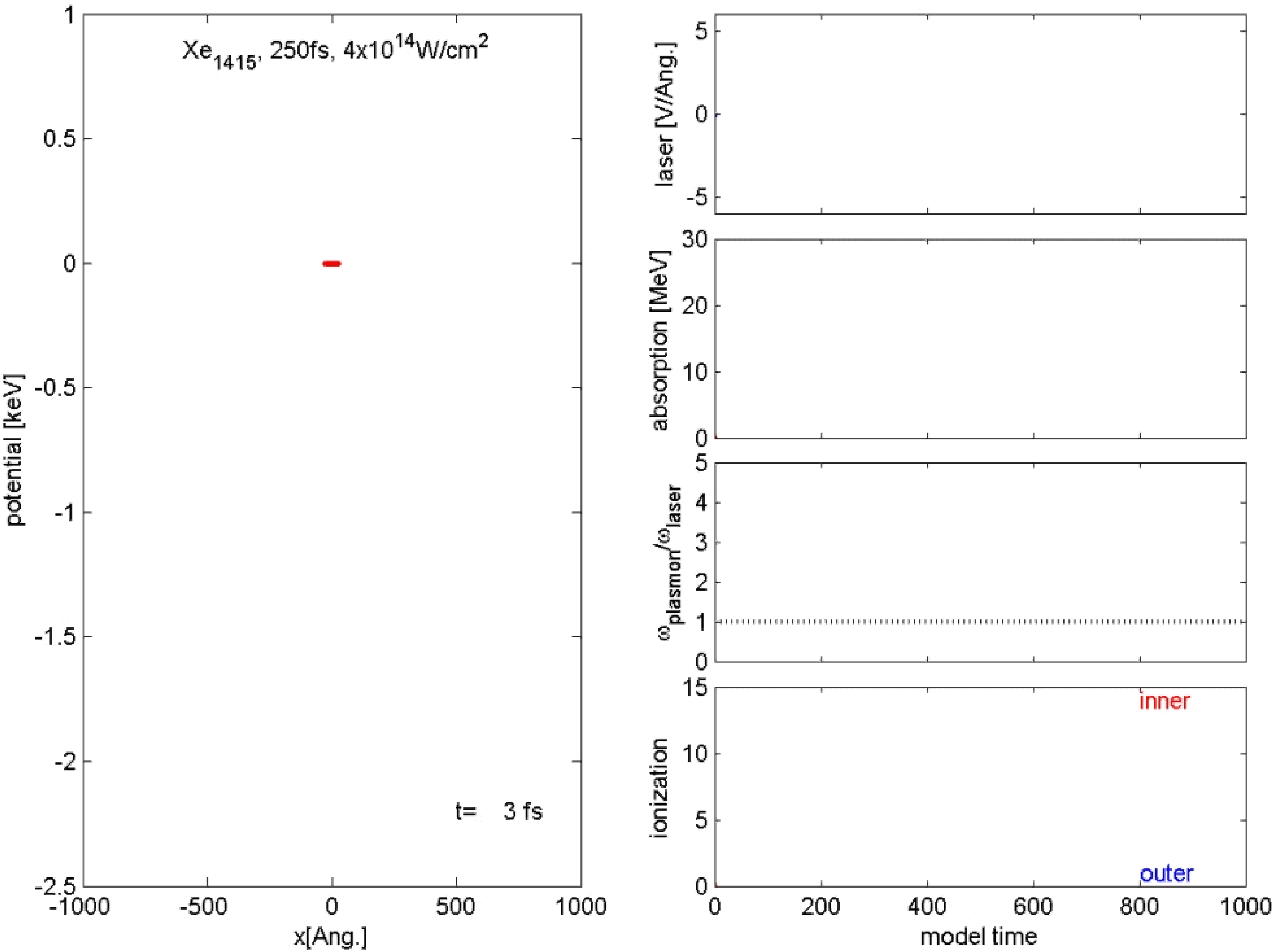
computer experiment: laser excitation of Xe clusters

250fs, 800nm,
4×10^{14} W/cm^2



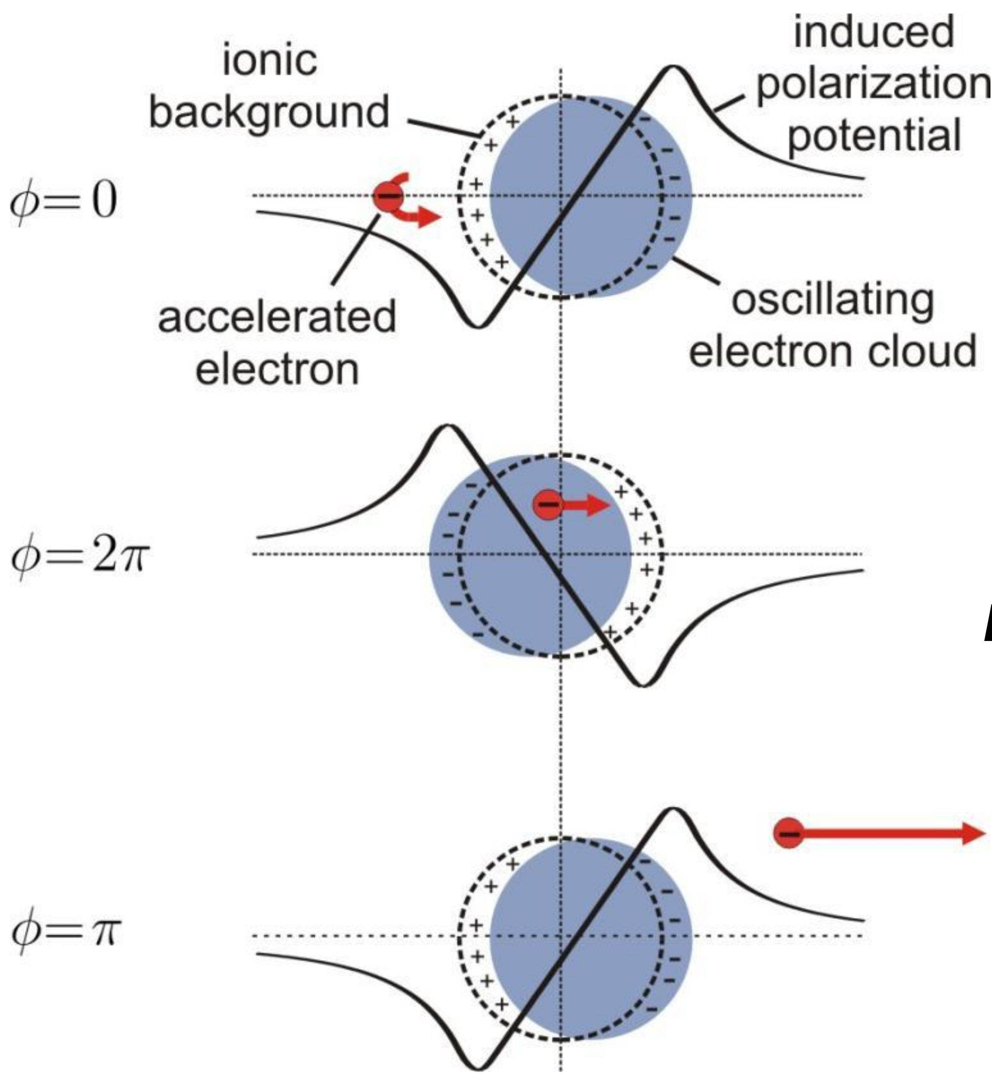


Xe_{1415}



Xe_{1415}, 250fs, $4x10^{14}W/cm^2$
potential [keV]
x[Ang.]
t= 3 fs
1
0.5
0
-0.5
-1
-1.5
-2
-2.5
-1000
-500
0
500
1000
laser [V/Ang.]
5
0
-5
absorption [MeV]
30
20
10
0
$\omega_{plasmon}/\omega_{laser}$
5
4
3
2
1
0
ionization
15
10
5
0
inner
outer
0
200
400
600
800
1000
model time

on plasmon resonance: dynamic electron acceleration



field strenght for N = 500:

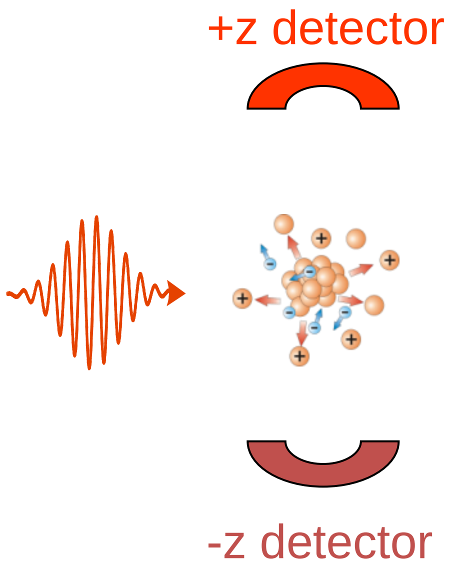
140 GeV/m

Surface-Plasmon Assisted Resonant electron acceleration in Clusters SPARC

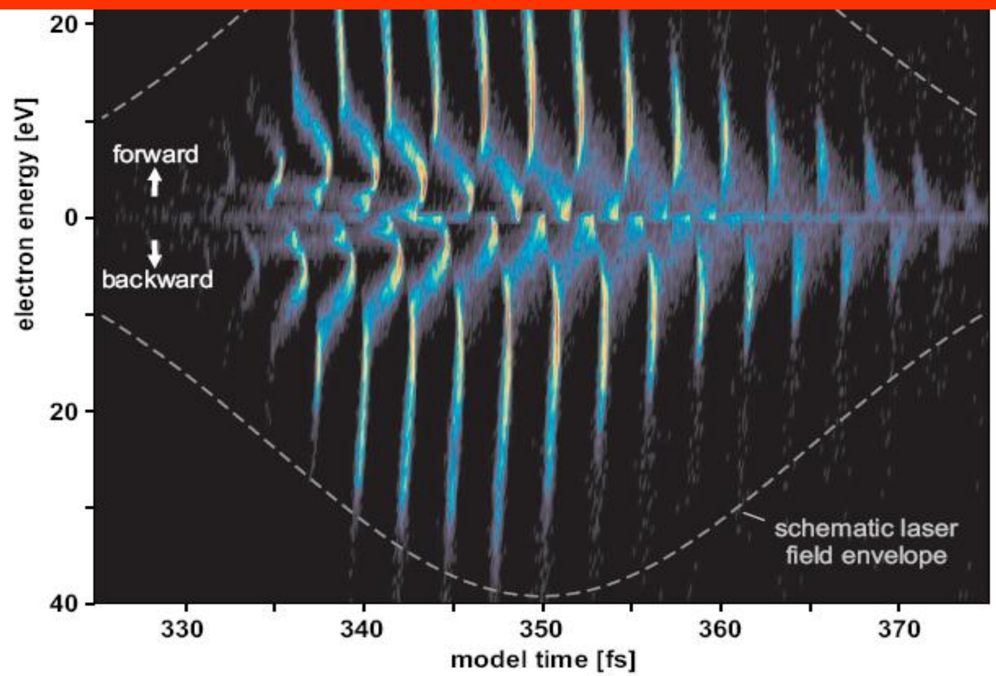
Nano electron accelerator

attosecond electron bursts

numerical experiment

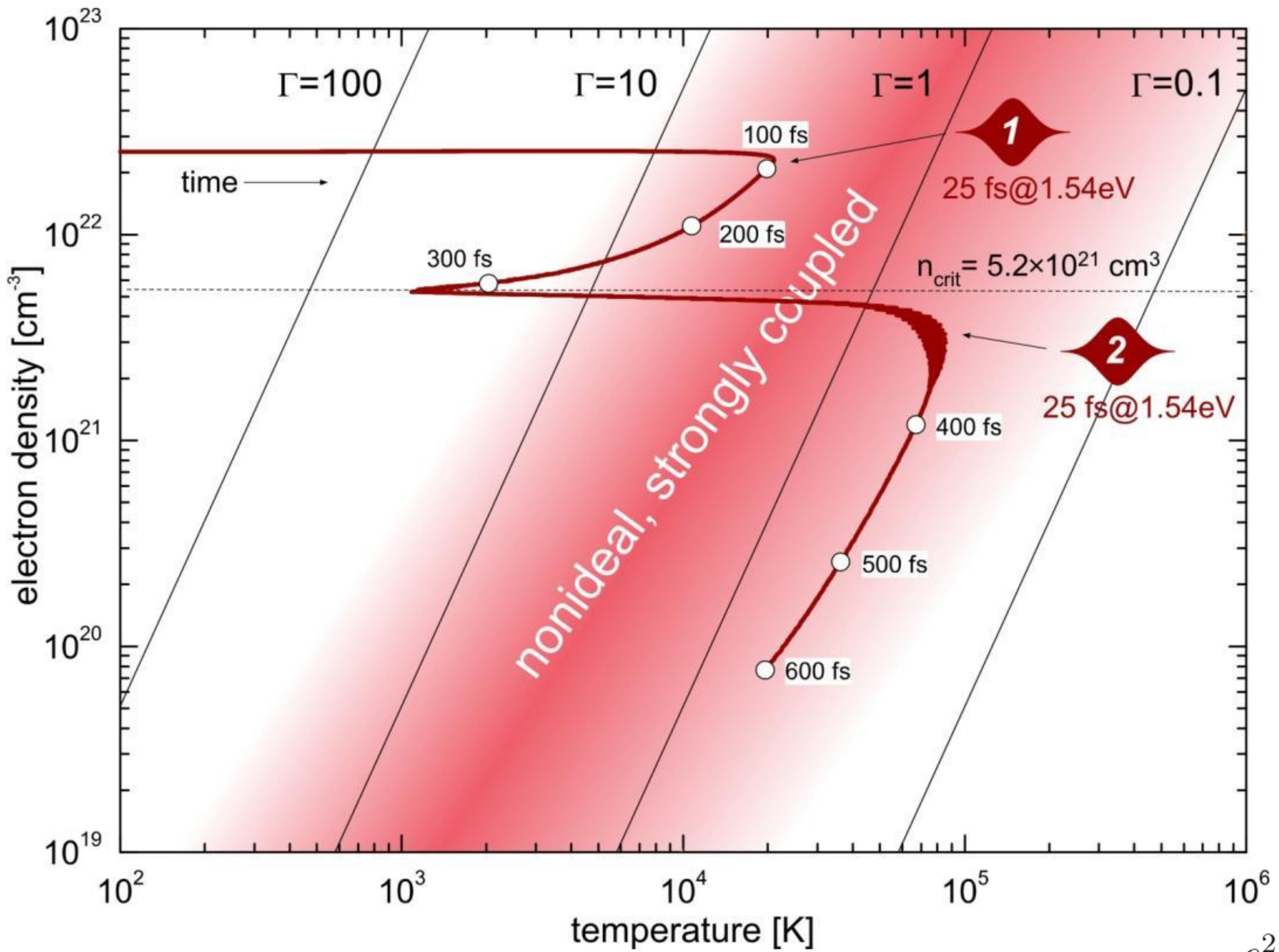


➢interesting playground for future few-cycle and attosecond physics



Fennel et al., PRL 98, 143401 (2007)

density-temperature plane



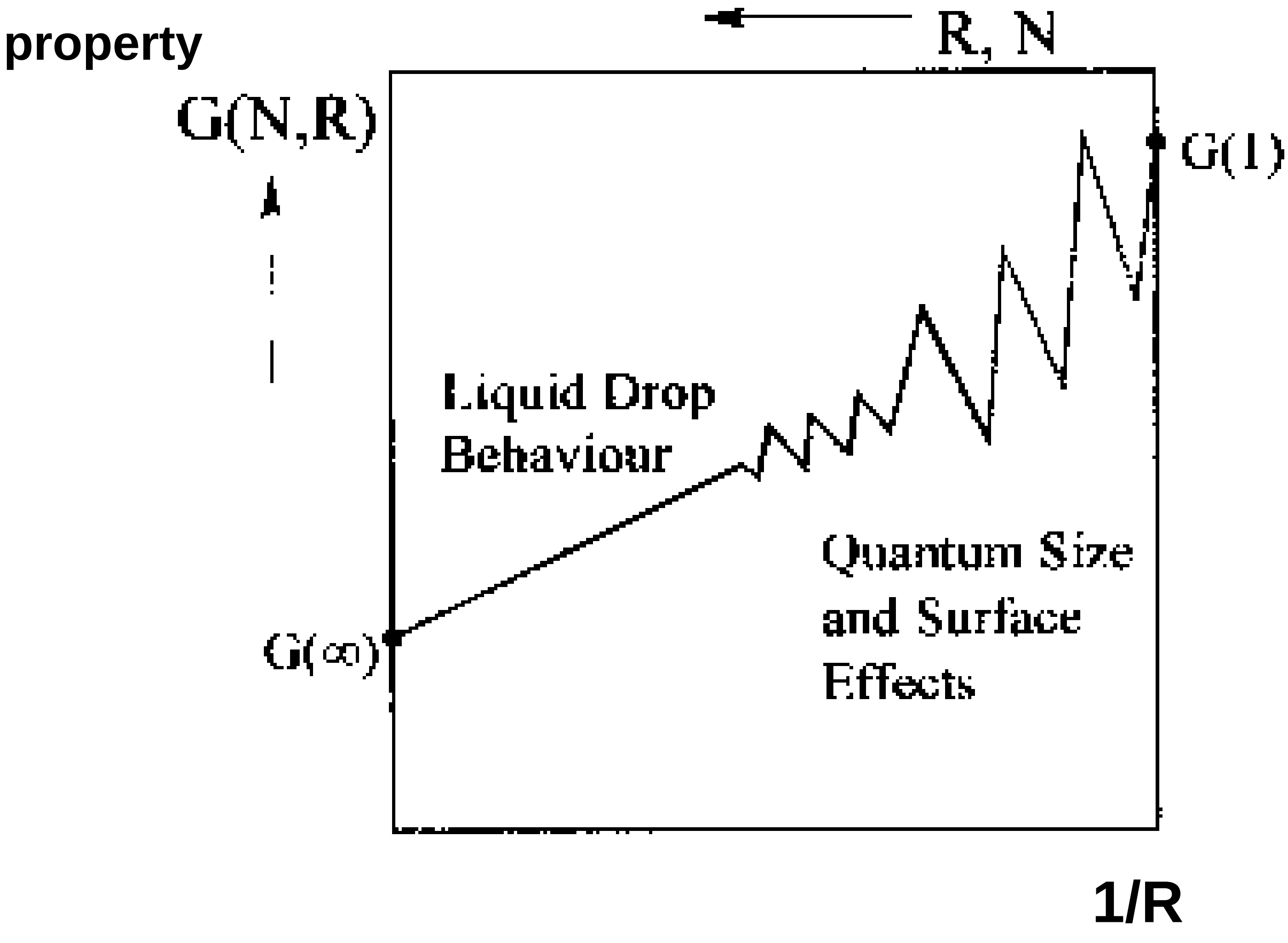
coupling parameter

$$\Gamma = \frac{e^2}{4\pi\varepsilon_0 k_B T}(\frac{4}{3}\pi n_e)^{1/3}$$

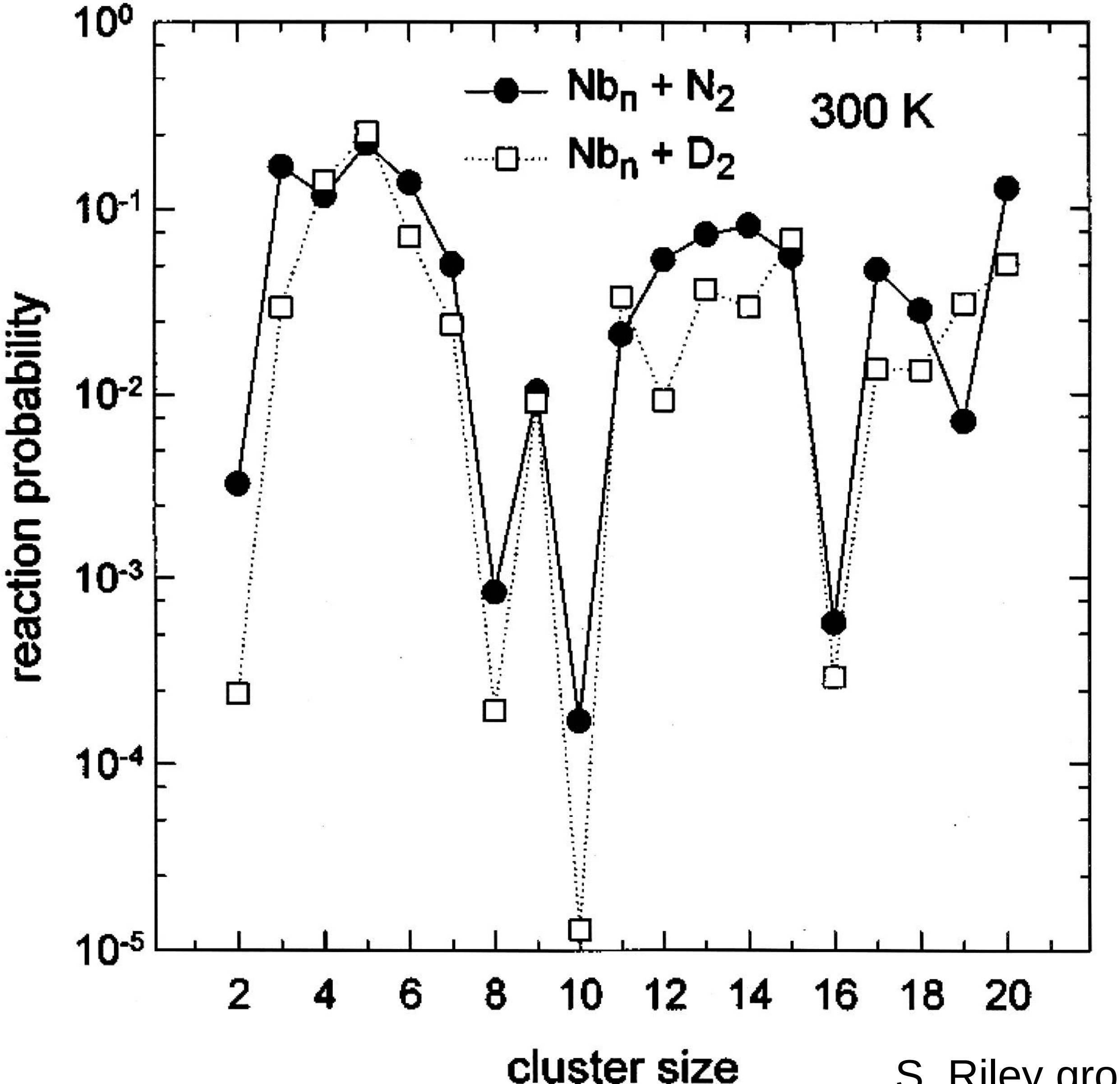
plasma physics on the nm scale

summarizing statements and few more features

scaleable *vs.* non-scaleable regime

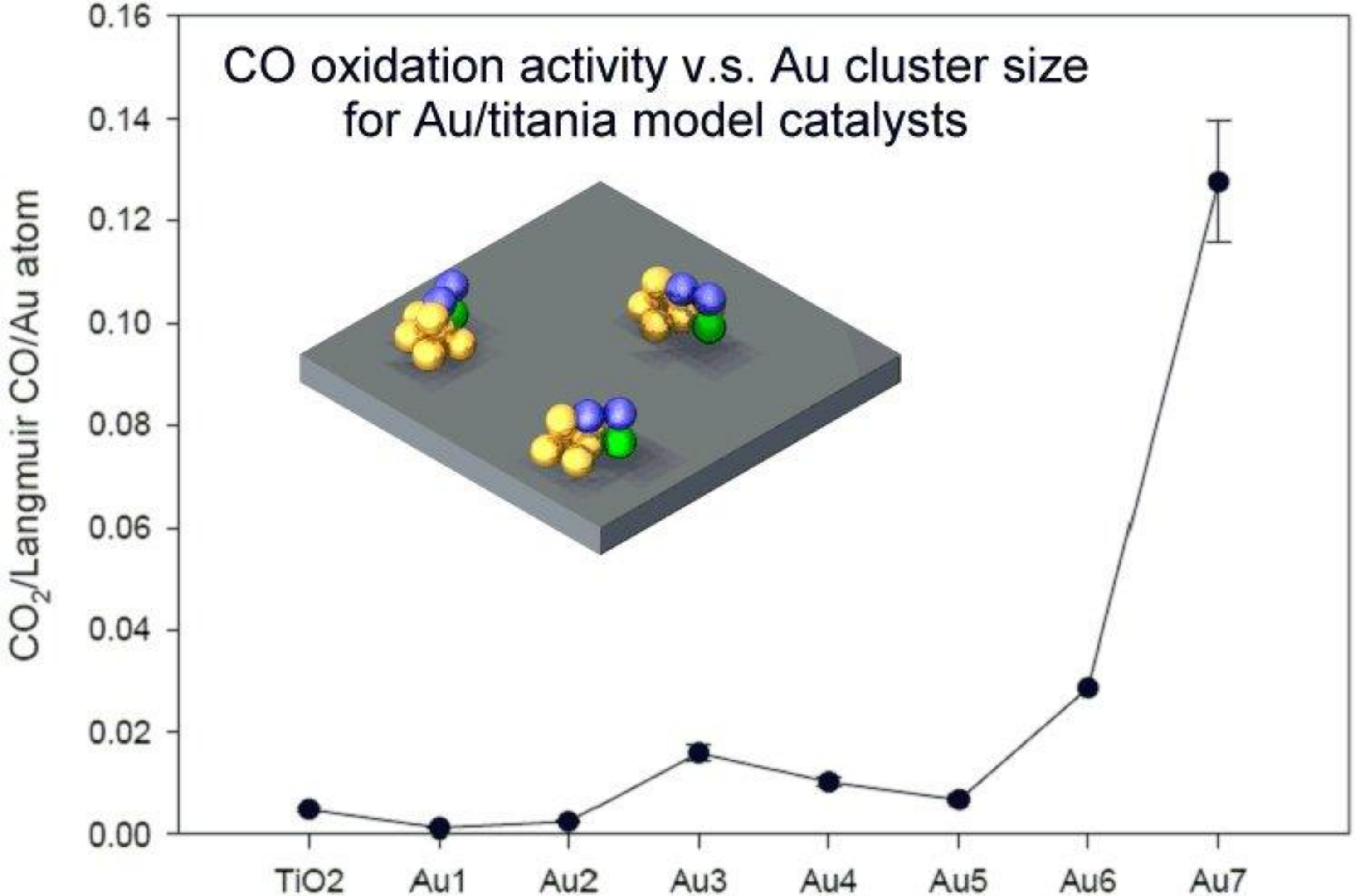


chemical reactivity of clusters



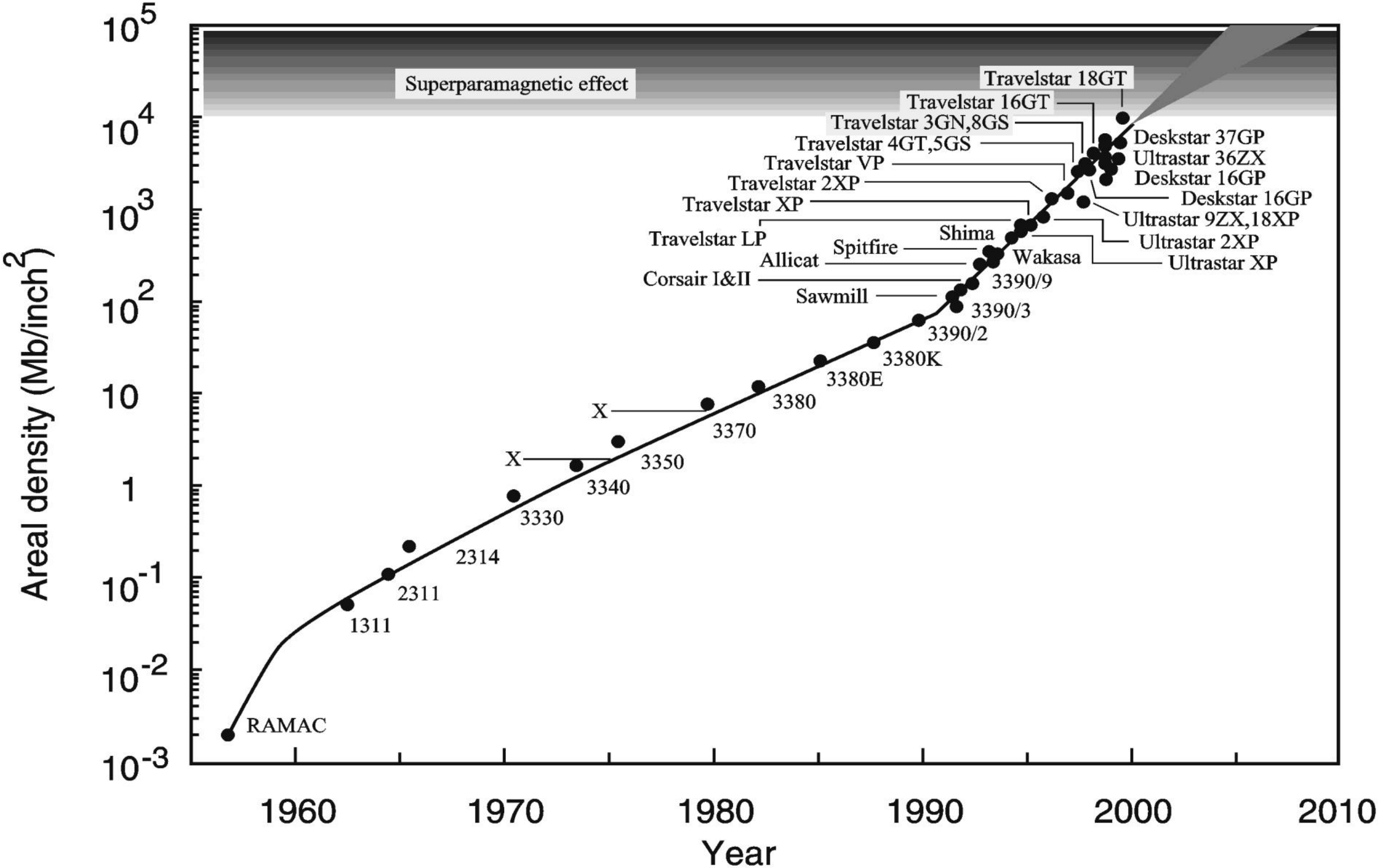
S. Riley group

clusters in chemistry: Au_N catalysis



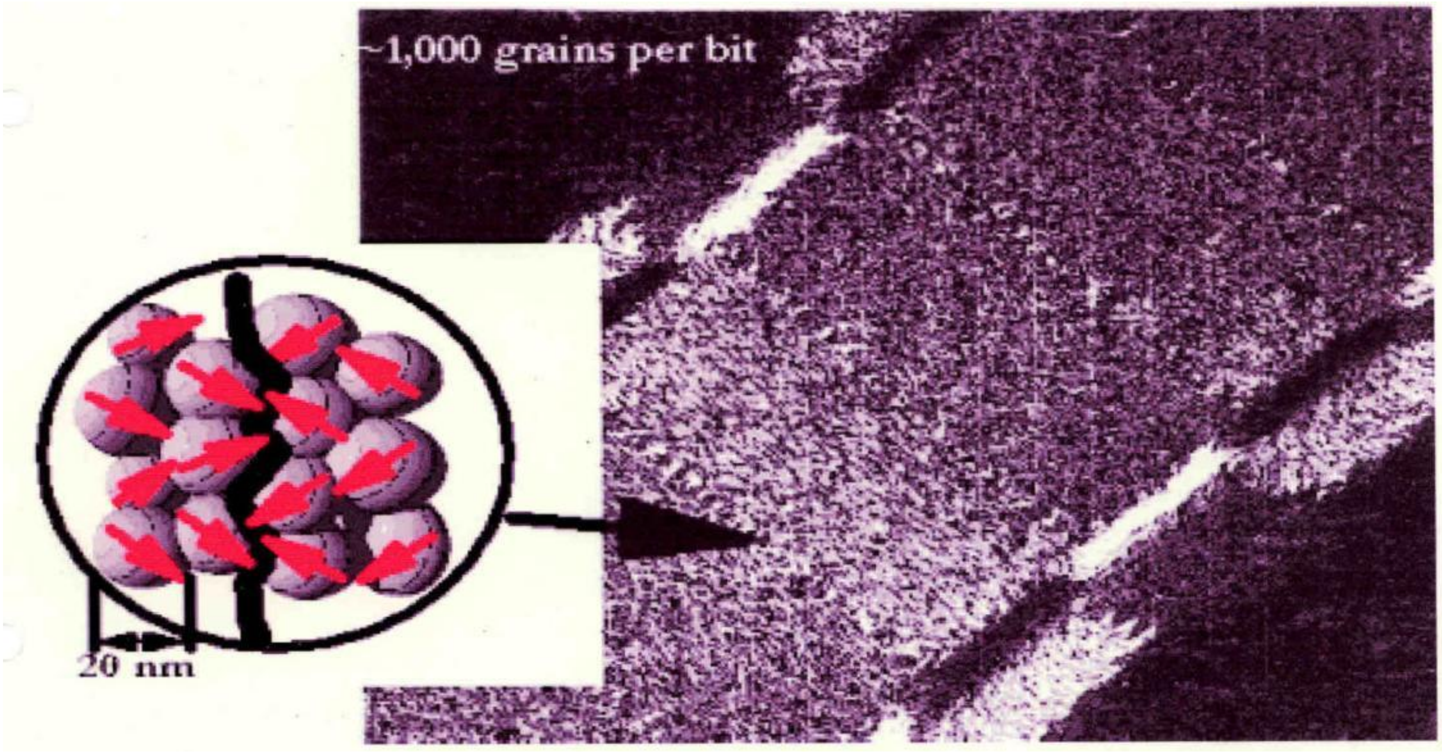
Scott Anderson

magnetic data storage



See: Thompson et. al., IBM J. Res. Develop. 44, 311 (2000)

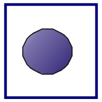
already today:
granular magnetic media for magnetic recording

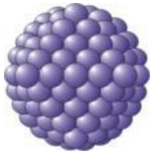


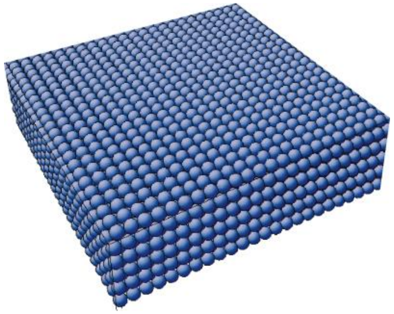
IBM

Magnetism between atom and bulk

Eisen (Fe) :

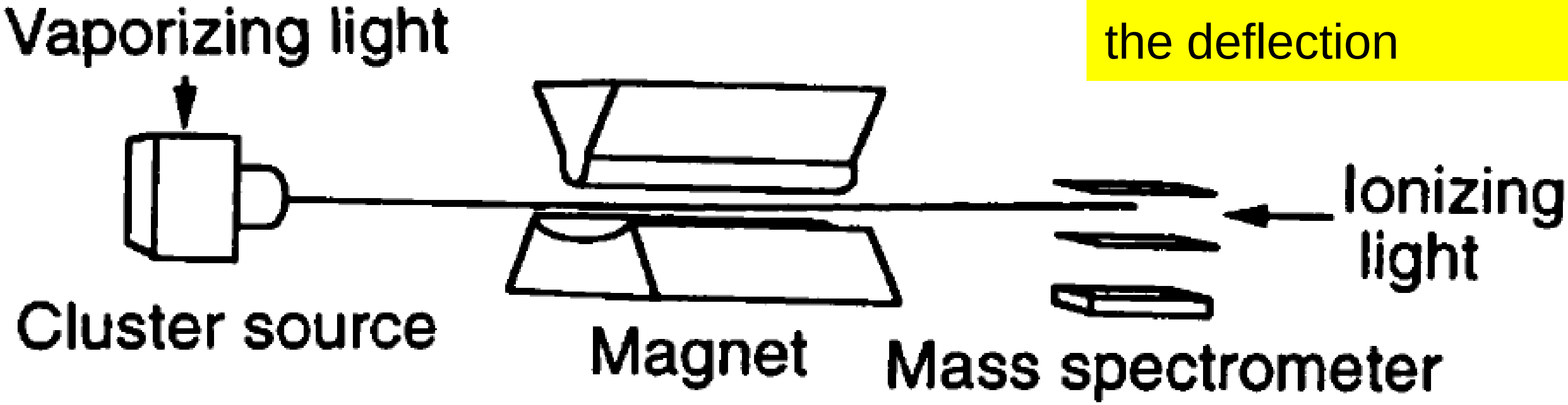




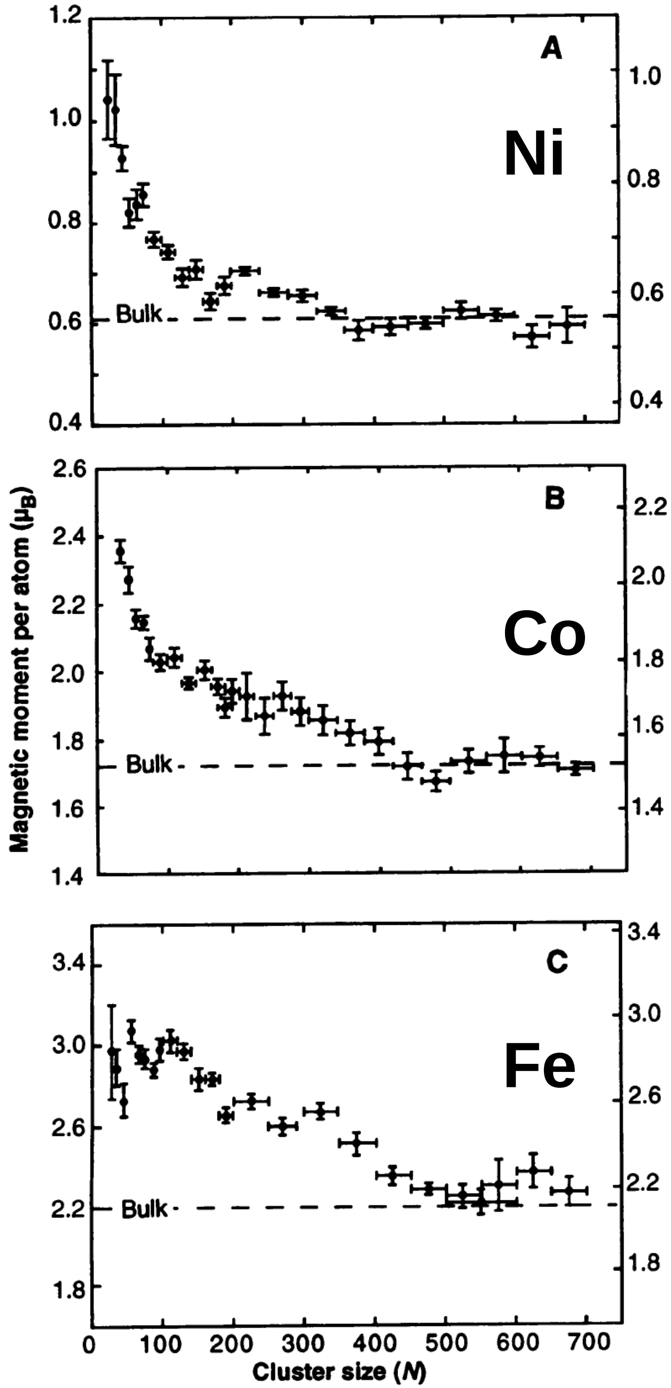


	Atom:	Cluster:	Festkörper:
Magn. Moment	$M = 6\ \mu_B$	?	$M = 2.2\ \mu_B$

measure the magnetic moments by deflection in a Stern-Gerlach experiment



position sensitive ionization to measure the deflection



magnetic moments per atom of clusters in a beam

Billas, de Heer, Science 265, 1682, 1994

cluster magnetism with non-magnetic elements

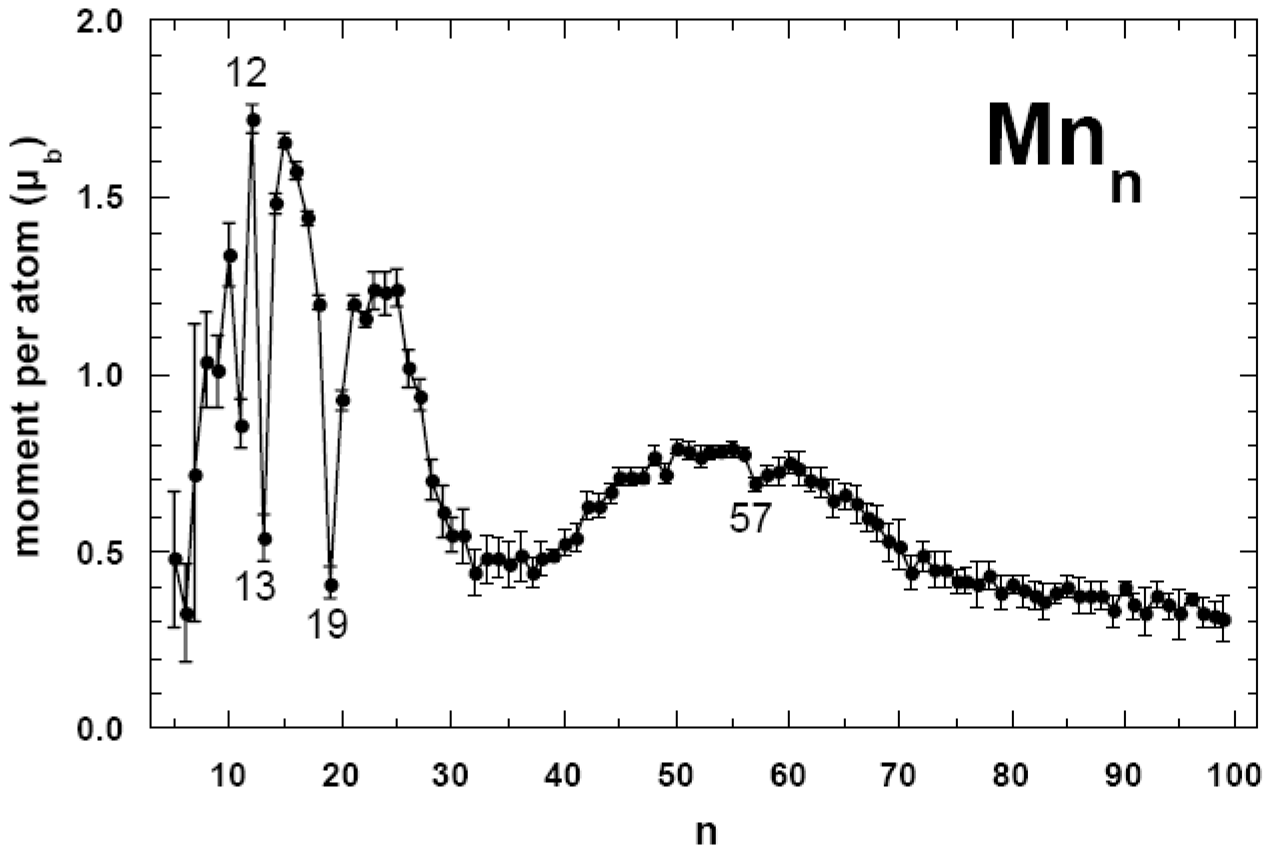
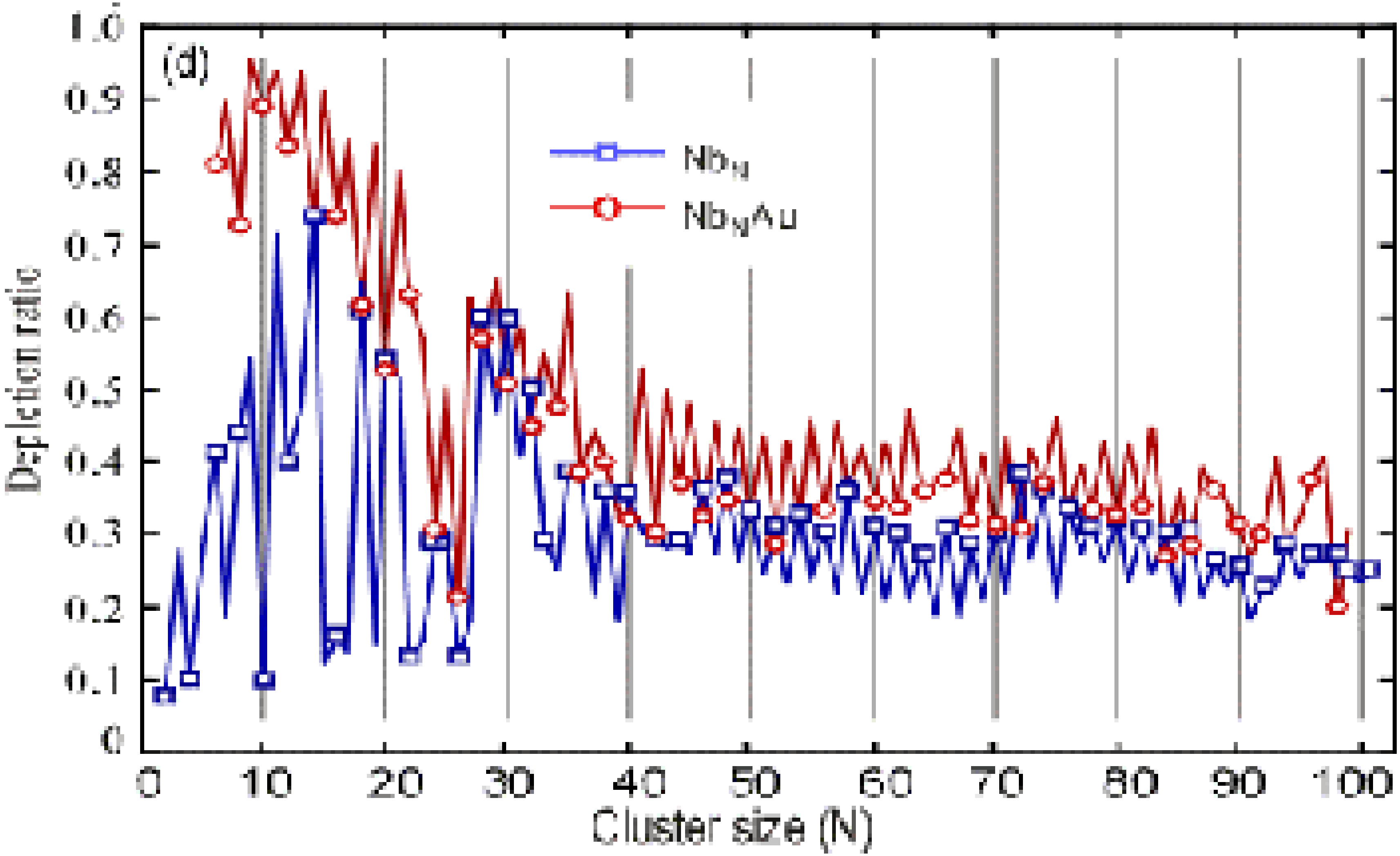


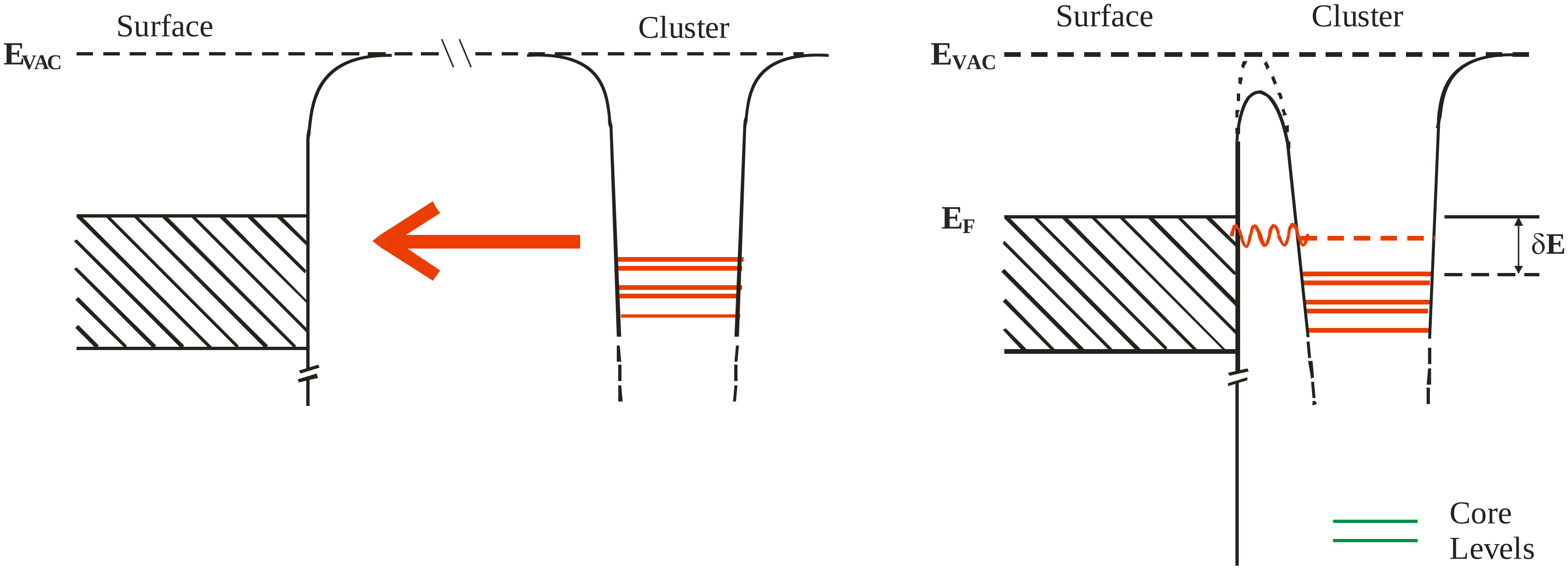
Figure 1. Magnetic moments per atom for manganese clusters produced at 68K.

Knickelbein

magnetic enhancement by gold doping



clusters at surfaces

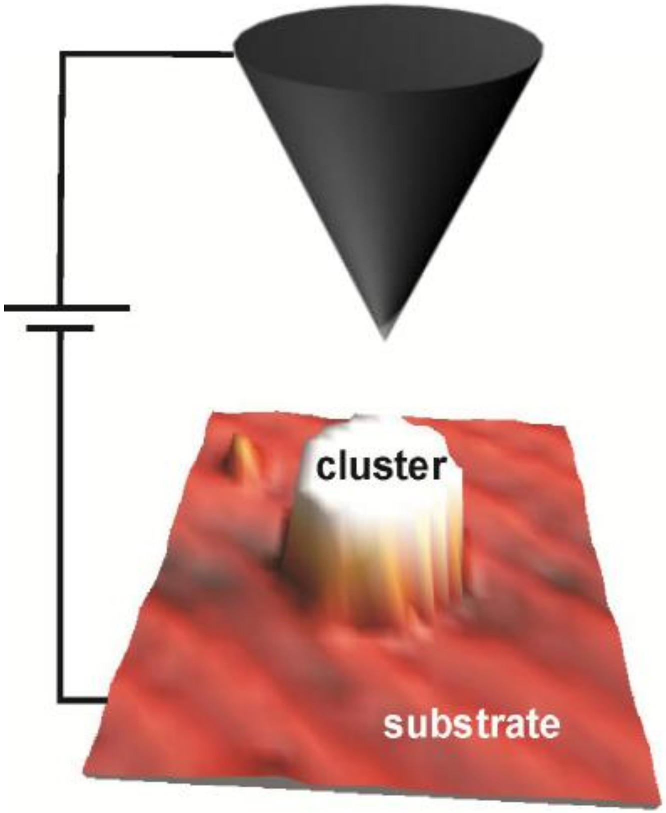


Cluster Physics *and* Surface Physics

metallic clusters on metal surface

Electronic properties of metallic nanoparticles
as a function of size and shape

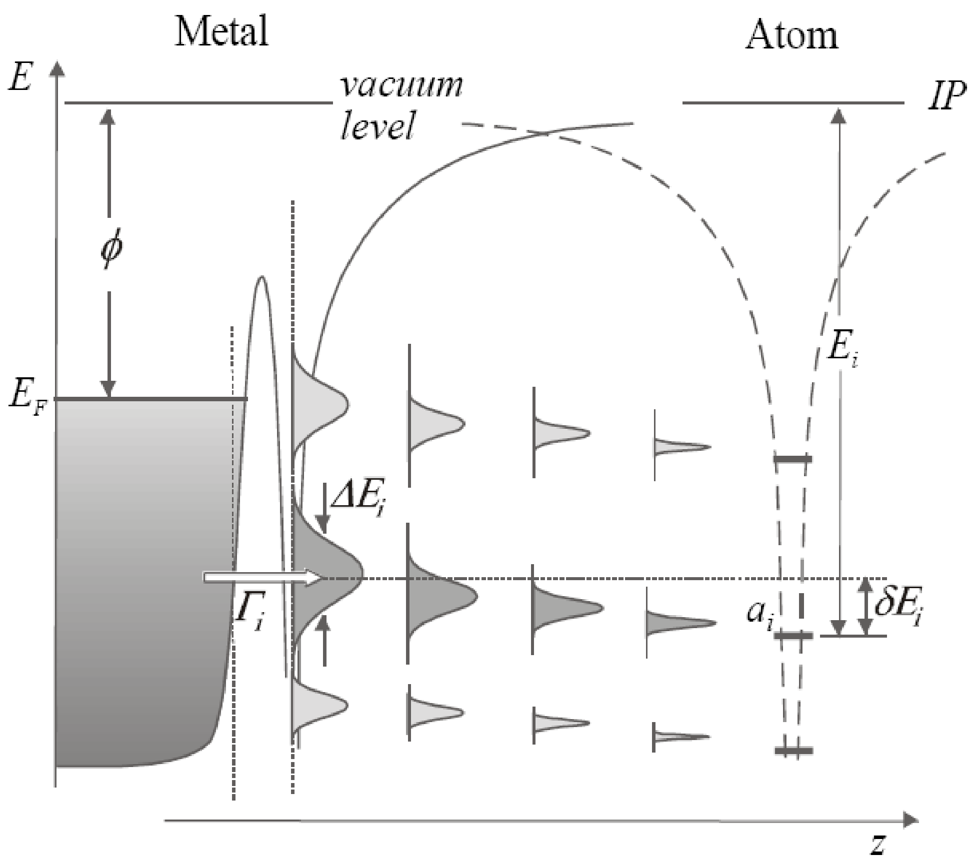
Tool: Scanning Tunneling Microscope



Quantum mechanical confinement phenomena in individual nanoparticles on a metallic substrate:

- Strong lateral confinement
- Interaction with substrate

atoms approaching surfaces: interactions

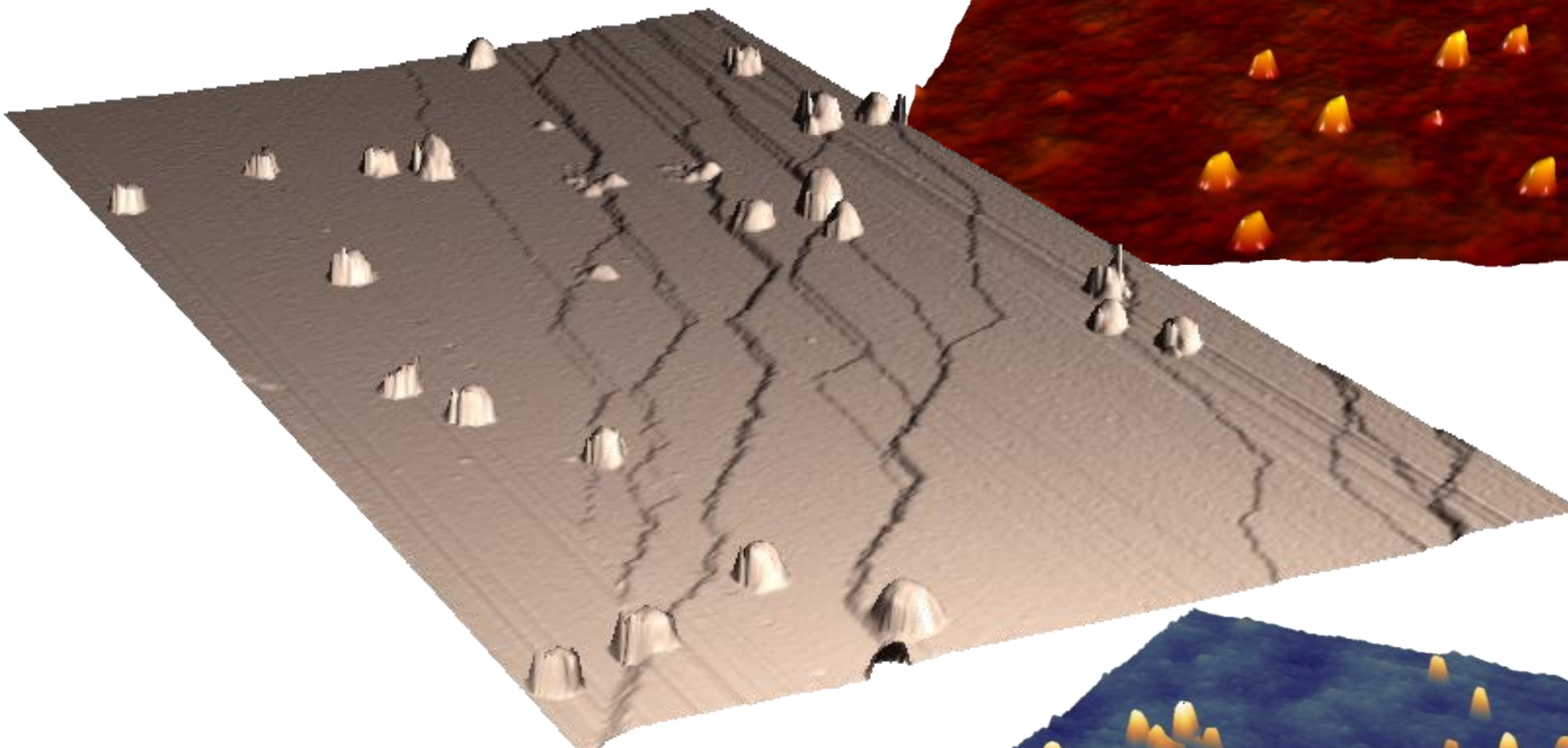


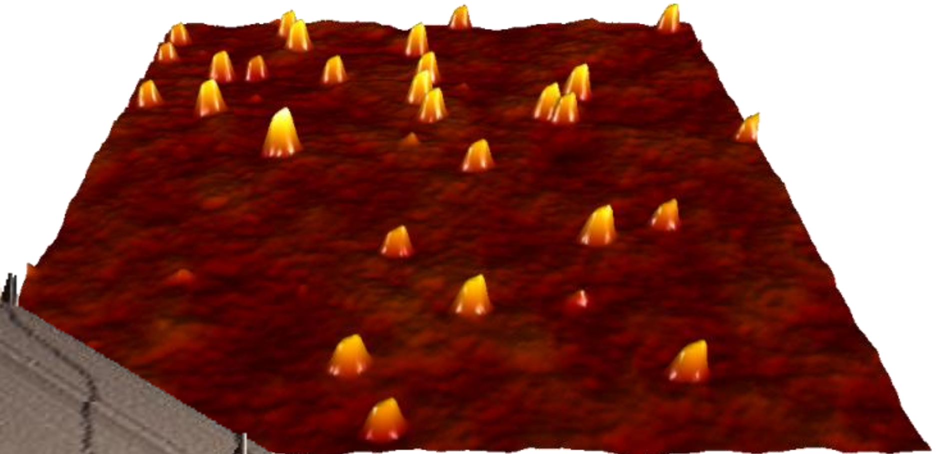
Newns-Anderson model

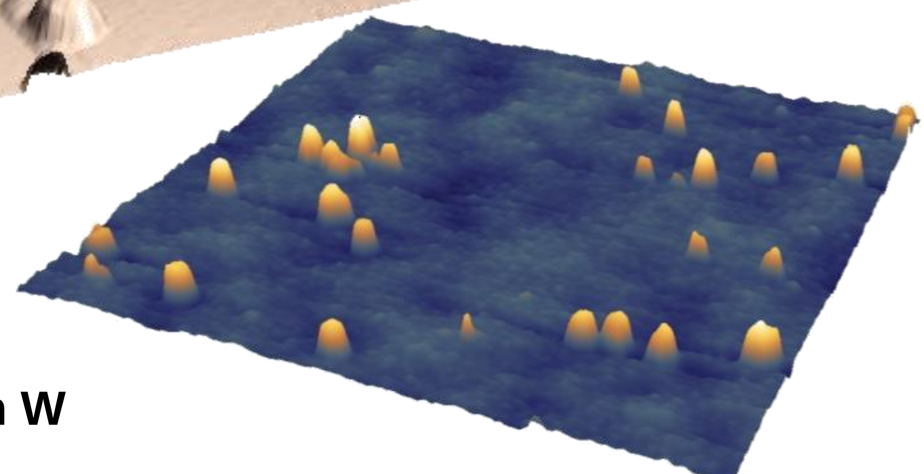
- *coupling strength*
  atomic and
  metal valence-band states
- *spatial overlap*
  atomic and metallic
  electron wave functions

cfr. J. Los *et al.*, Phys. Rep.**190**(1990)133.
P. Lievens *et al.*, NIMB**135**(1998) 471.

clusters at crysatalline surfaces: own work





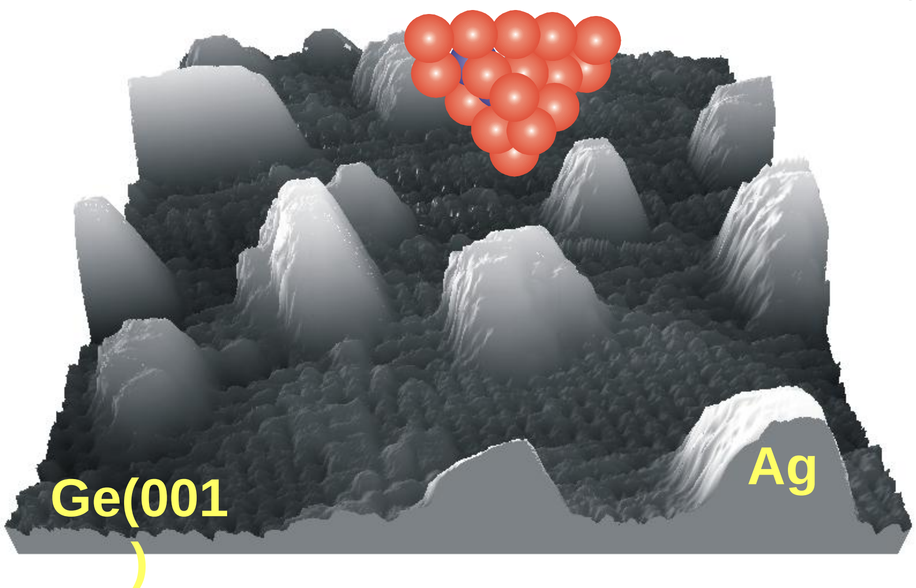


3 – 10 nm Cobalt particles on W

charge transport in the STM

$$I \propto \int_0^{eU} \rho_P(E_F - eU + E)\cdot \rho_S(E_F + E)\cdot T(E,eU)\,dE$$

DOS sample **DOS tip** **transmission**

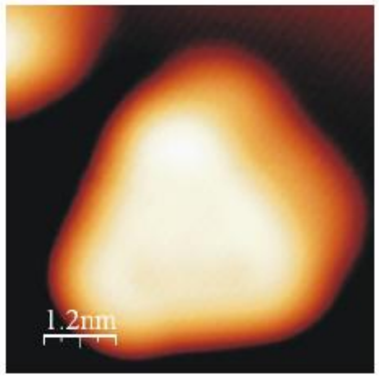


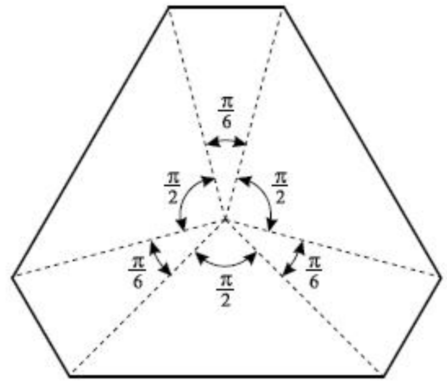
differential conductance

see lecture of Ingo Barke

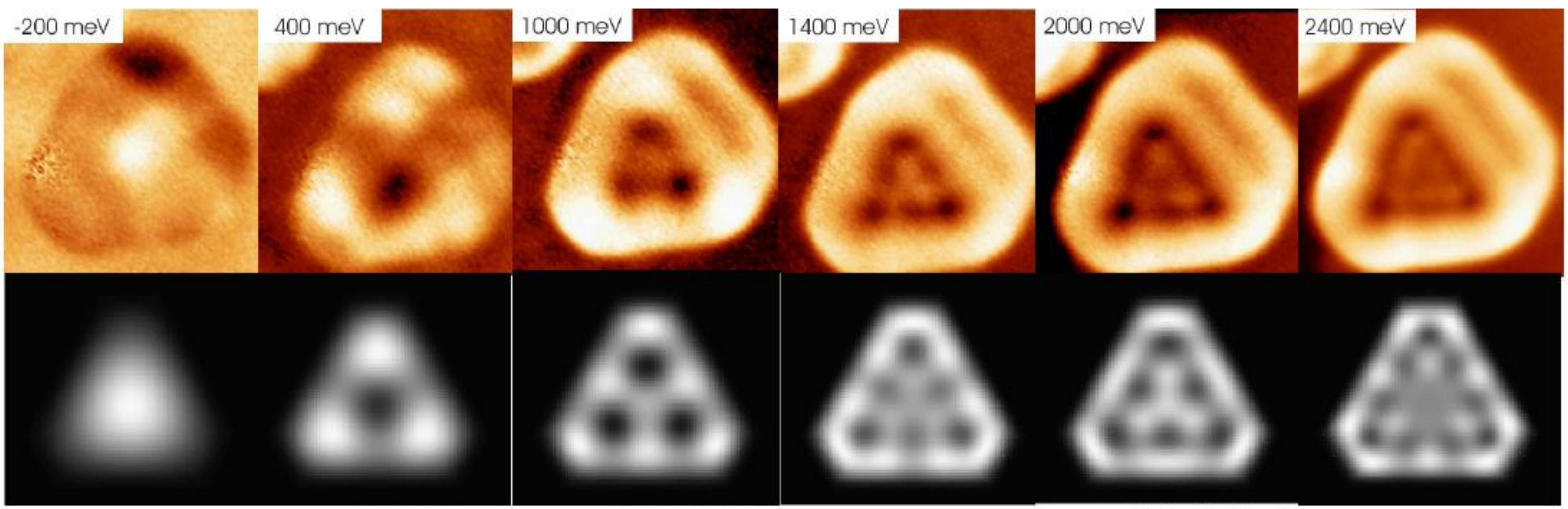
electron confinement to nanoscale Co islands

LDOS mapping & standing waves





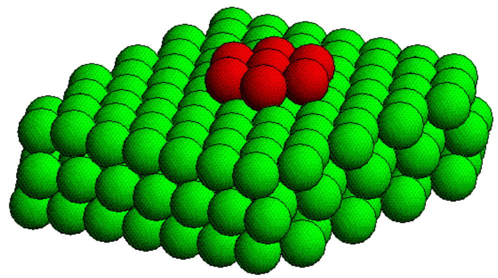
Particle-in-a-box
$S = 6.5\ nm^2$
$m^* = 0.40\ m_e$



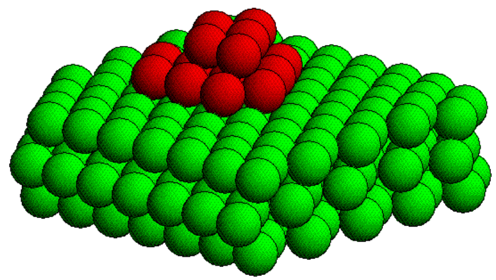
$\lambda=1$ $E_n - E_0 = 30meV$ $\lambda=20$ $E_n - E_0 = 540meV$ $\lambda=45$ $E_n - E_0 = 1200meV$ $\lambda=60$ $E_n - E_0 = 1600meV$ $\lambda=80$ $E_n - E_0 = 2140meV$ $\lambda=95$ $E_n - E_0 = 2540meV$

theoretically demanding systems

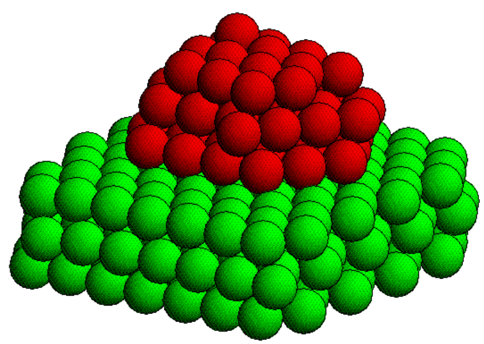
Au_N on gold



Au_7



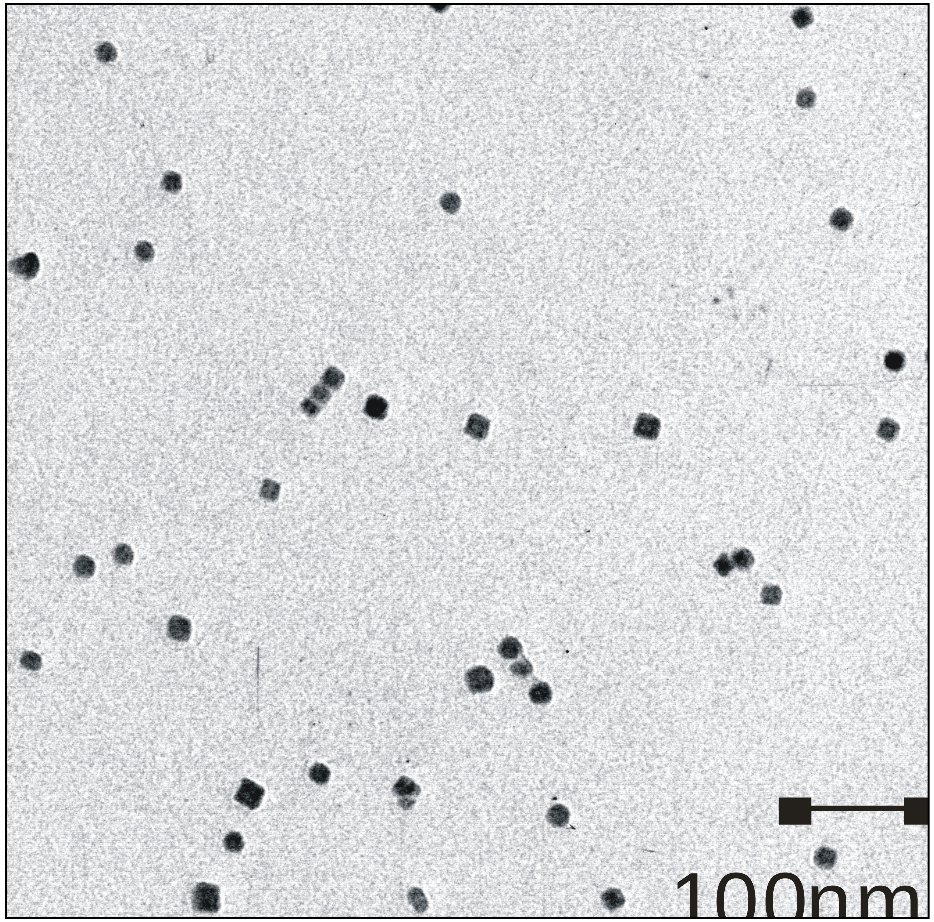
Au_{17}



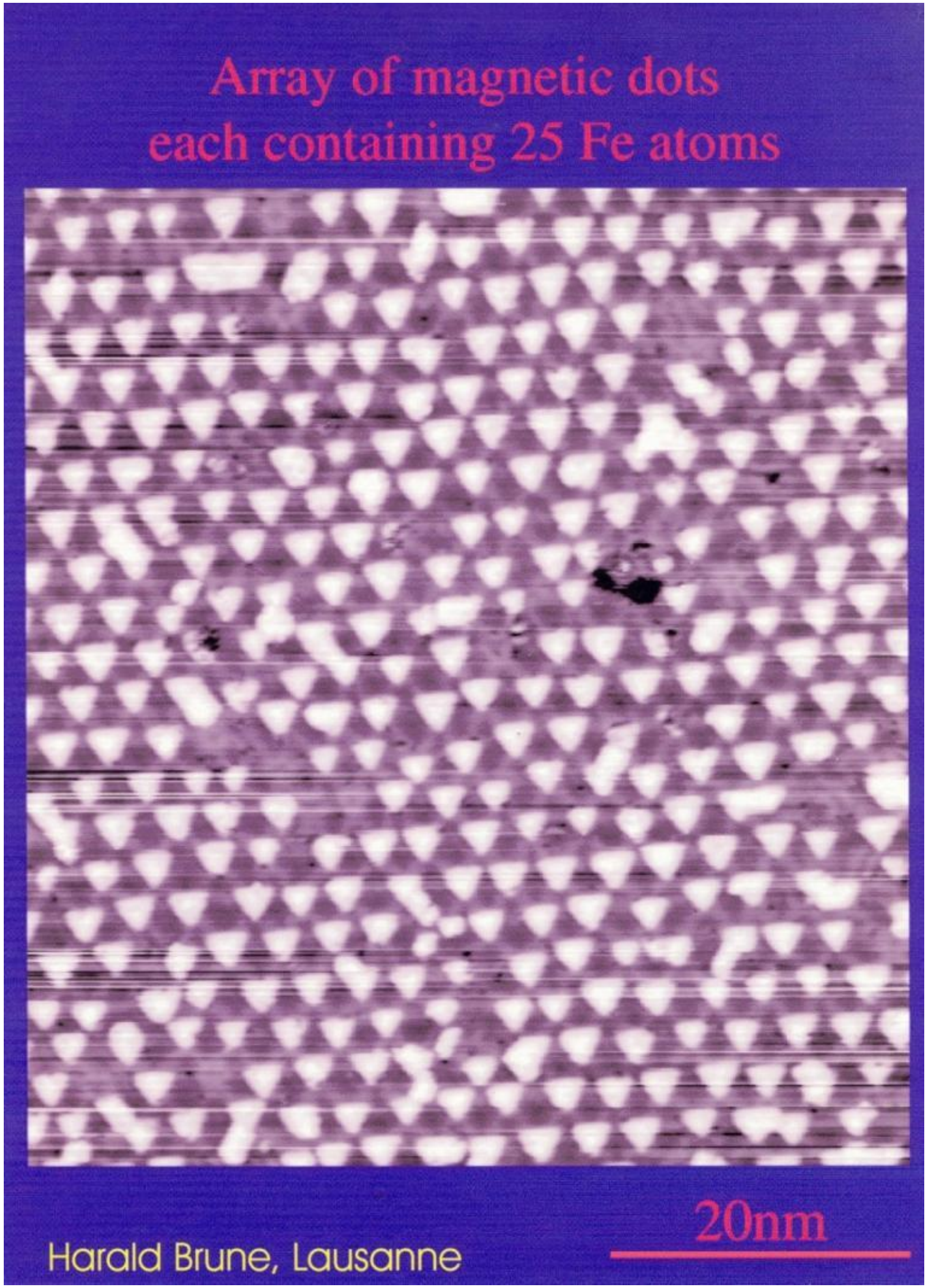
Au_{55}

Springborg, Saarbrücken

example of deposited iron clusters
mean diameter 9 nm



R.-P. Methling, V. Senz, E.-D. Klinkenberg, Th. Diederich, J. Tiggesbäumker, G. Holzhüter, J. Bansmann und K.H. Meiwes-Broer
Eur. Phys. J. D **16**:173-176, 2001



clusters growth by self organization

subject of surface physics