„There is plenty of light at the bottom"

KROÓ NORBERT

MTA WIGNER FIZIKAI KUTATÓKÖZPONT

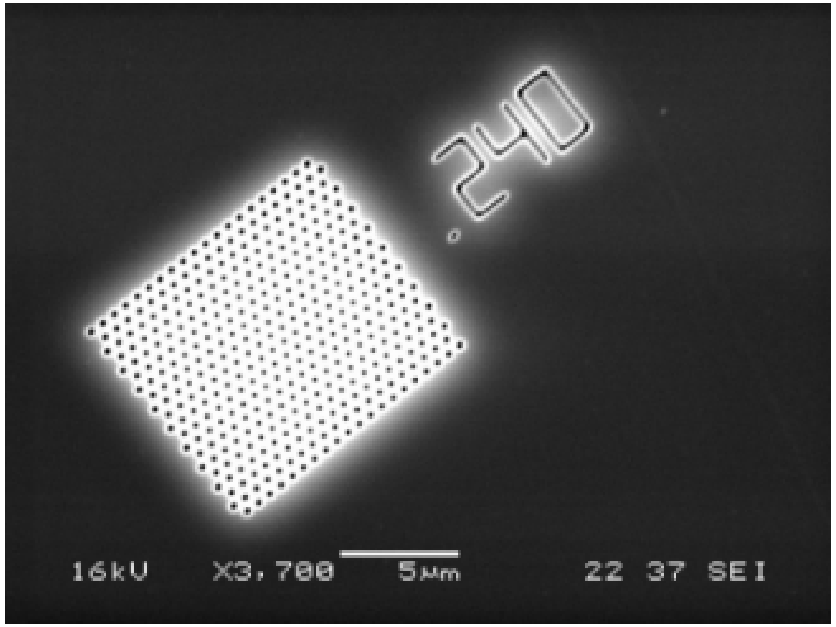
KFKI,28.09.2012

The Lycurgus Cup (glass; British Museum; 4th Century A. D.)

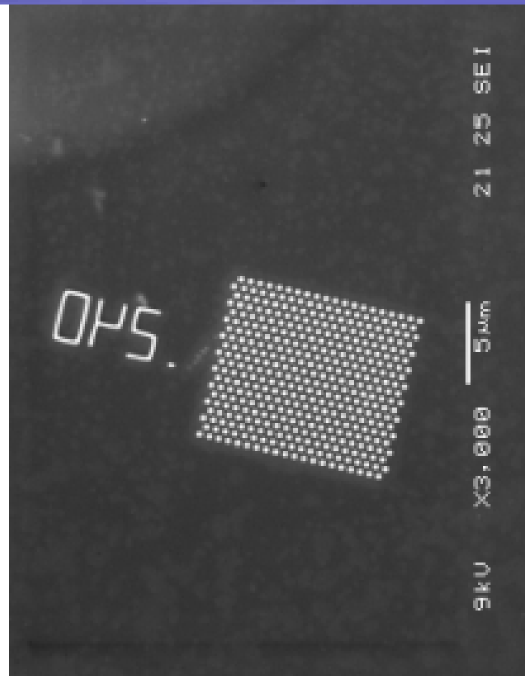
When illuminated from outside, it appears green. However, when illuminated from within the cup, it glows red. Red color is due to very small amounts of gold powder (about 40 parts per million) in glass.

TRANSPARENCY WITH VERY SMALL HOLES

(Electron microscope images)



Reflected biam



Transmitted beam!!!

Holes, smaller than the wavelength of light. Light against the diffraction limit and

$$I > I_{geometry}$$

SURFACE PLASMON POLARITONS are a „NEW TYPE OF LIGHT", they are

1.BOUND TO THE (METAL) SURFACE,

2.HAVE SPECIFIC DISPERSION PROPERTIES,

3.THE DIFFRACTION LIMIT DOES NOT APPLY,

4. MAY BE GUIDED,

5.MAY HAVE A BANDGAP,

6. MAY INTERFERE,

7.REPRESENT VERY HIGH ELECTRIC FIELDS,

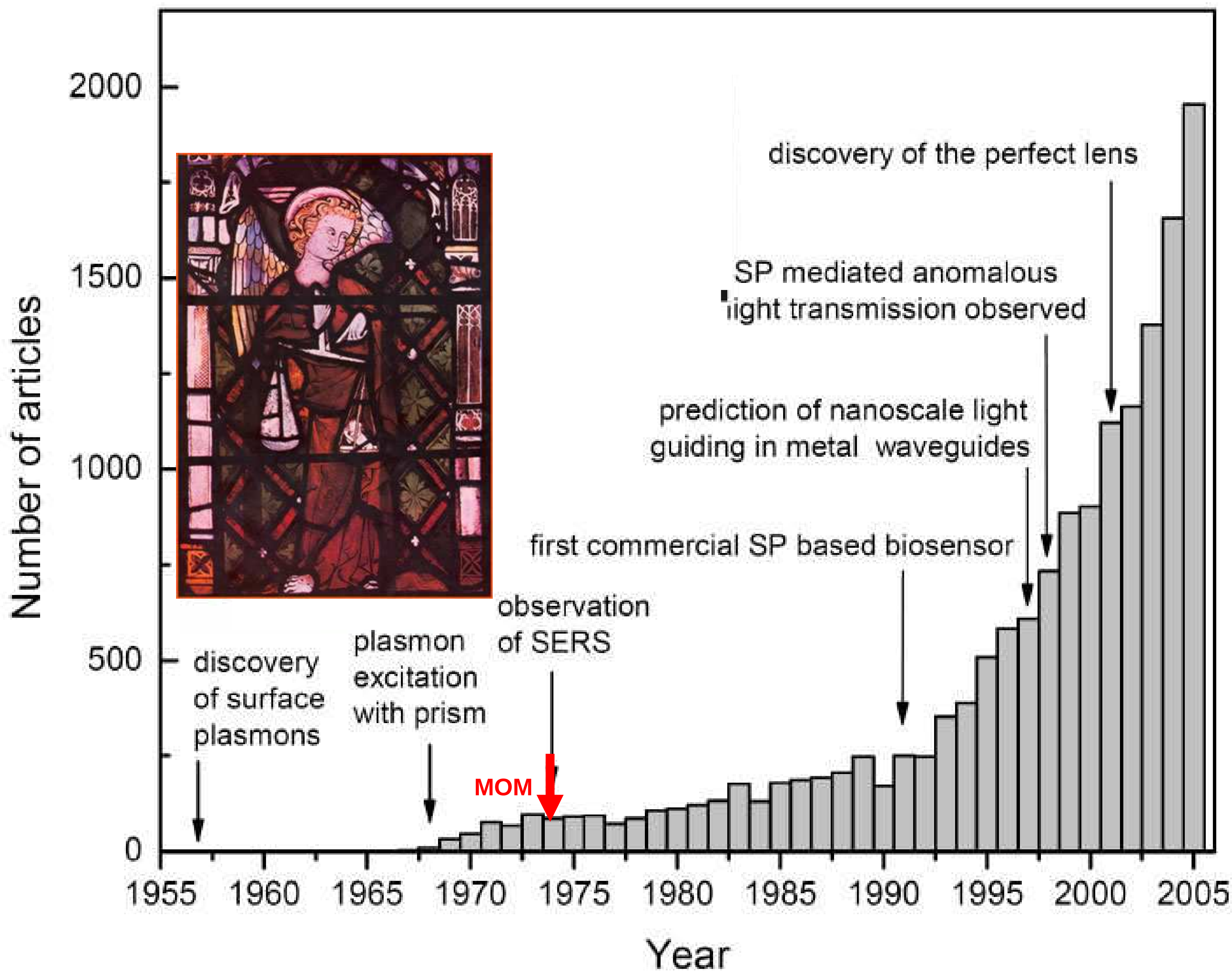
8.MAY BE LOCALIZED (e.g. to nanospheres or nanoshells)

9.MAY BE THE SUBJECT OF NONLINEAR PROCESSES

10.ULTRAFAST PHENOMENA

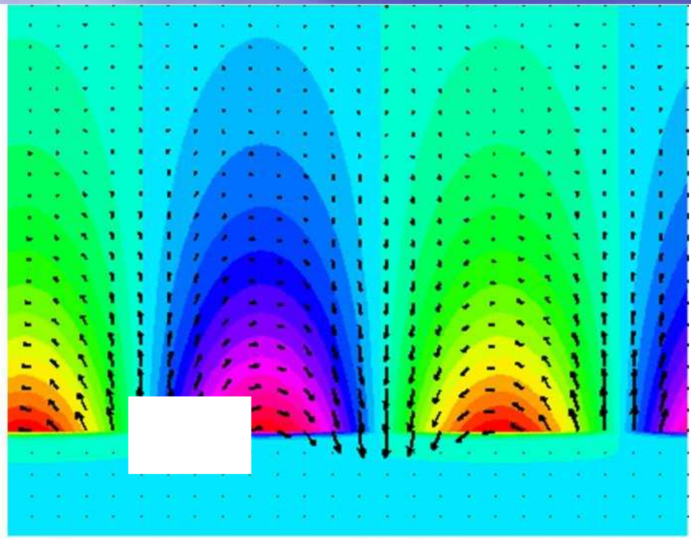
11.SPO „LASER" PHENOMENA

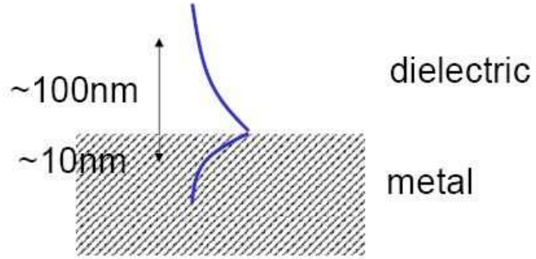
12.SHOW NON-CLASSICAL PROPERTIES



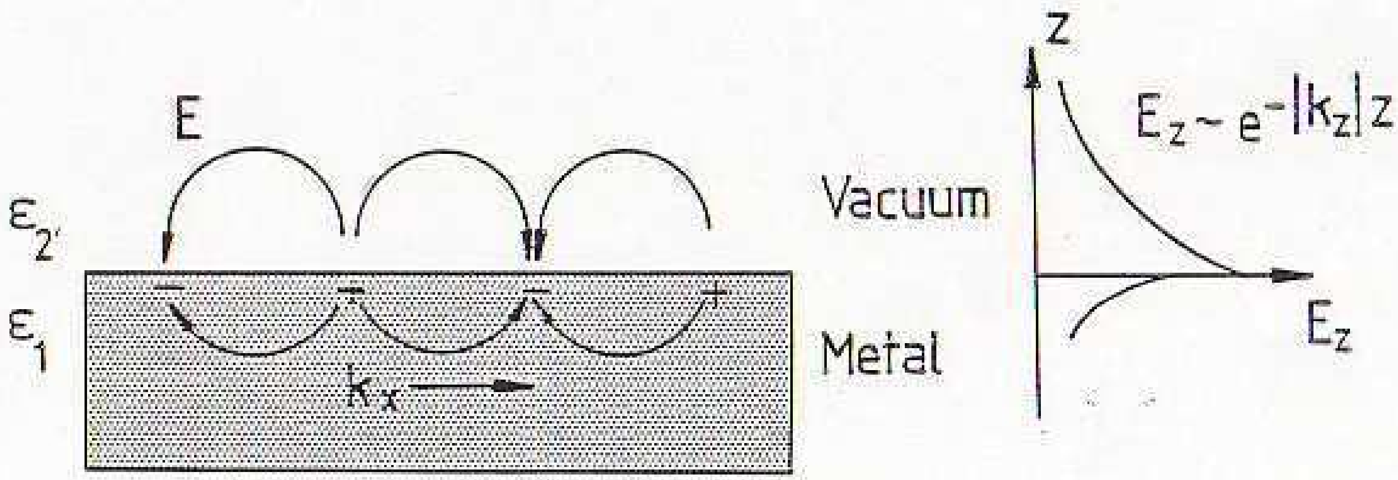
Number of articles
2000
1500
1000
500
0
1955
1960
1965
1970
1975
1980
1985
1990
1995
2000
2005
Year
discovery of the perfect lens
SP mediated anomalous light transmission observed
prediction of nanoscale light guiding in metal waveguides
first commercial SP based biosensor
observation of SERS
plasmon excitation with prism
discovery of surface plasmons
MOM

ELECTRIC FIELD OF SURFACE PLASMONS



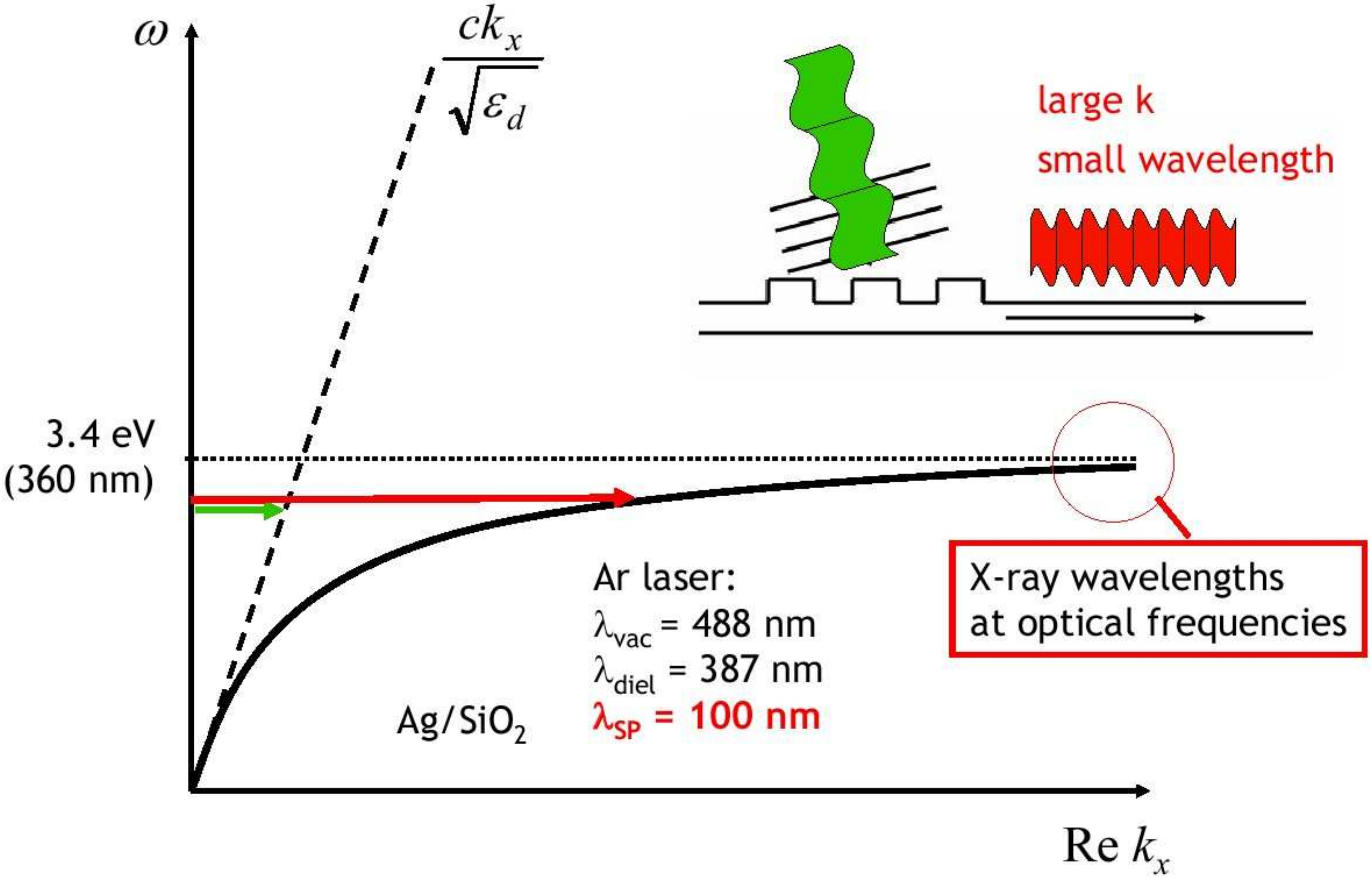


Most of the energy is confined in the dielectric side.

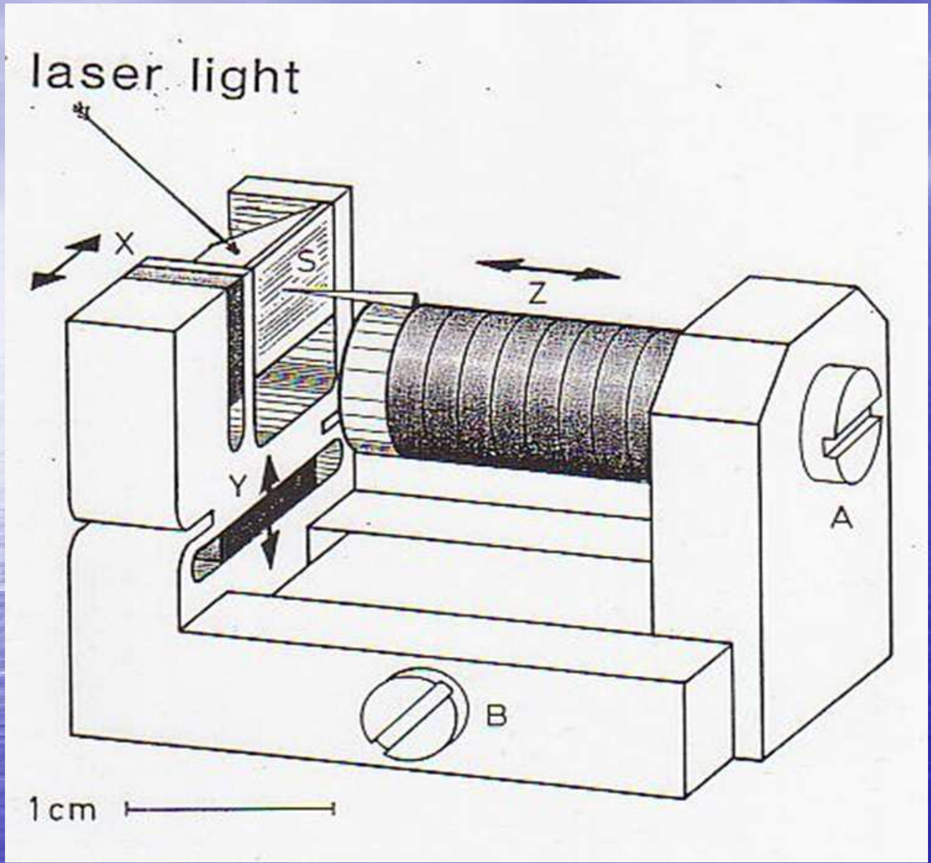


From Maxwell's equations

Surface plasmons dispersion: $k_x = \frac{\omega}{c}\left(\frac{\varepsilon_m \varepsilon_d}{\varepsilon_m + \varepsilon_d}\right)^{1/2}$



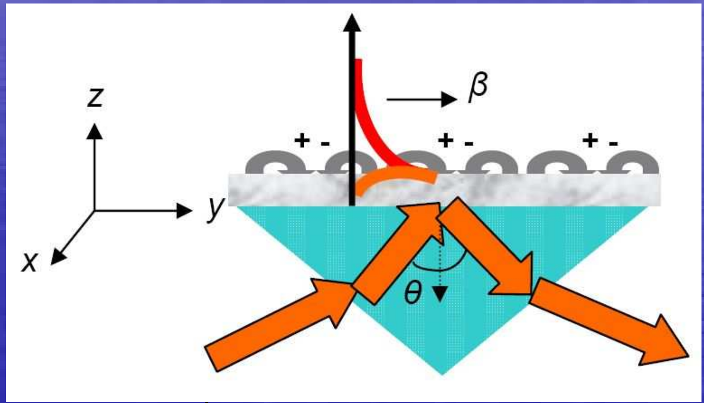
NEAR FIELD STM (against the diffraction limit)

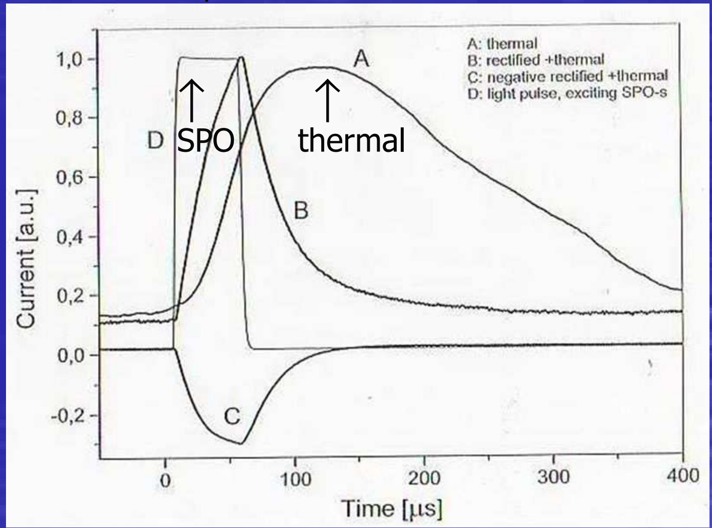


NEAR FIELD:LASER PULSE EXCITED

SURFACE PLASMONS

(Kretschmann geometry)

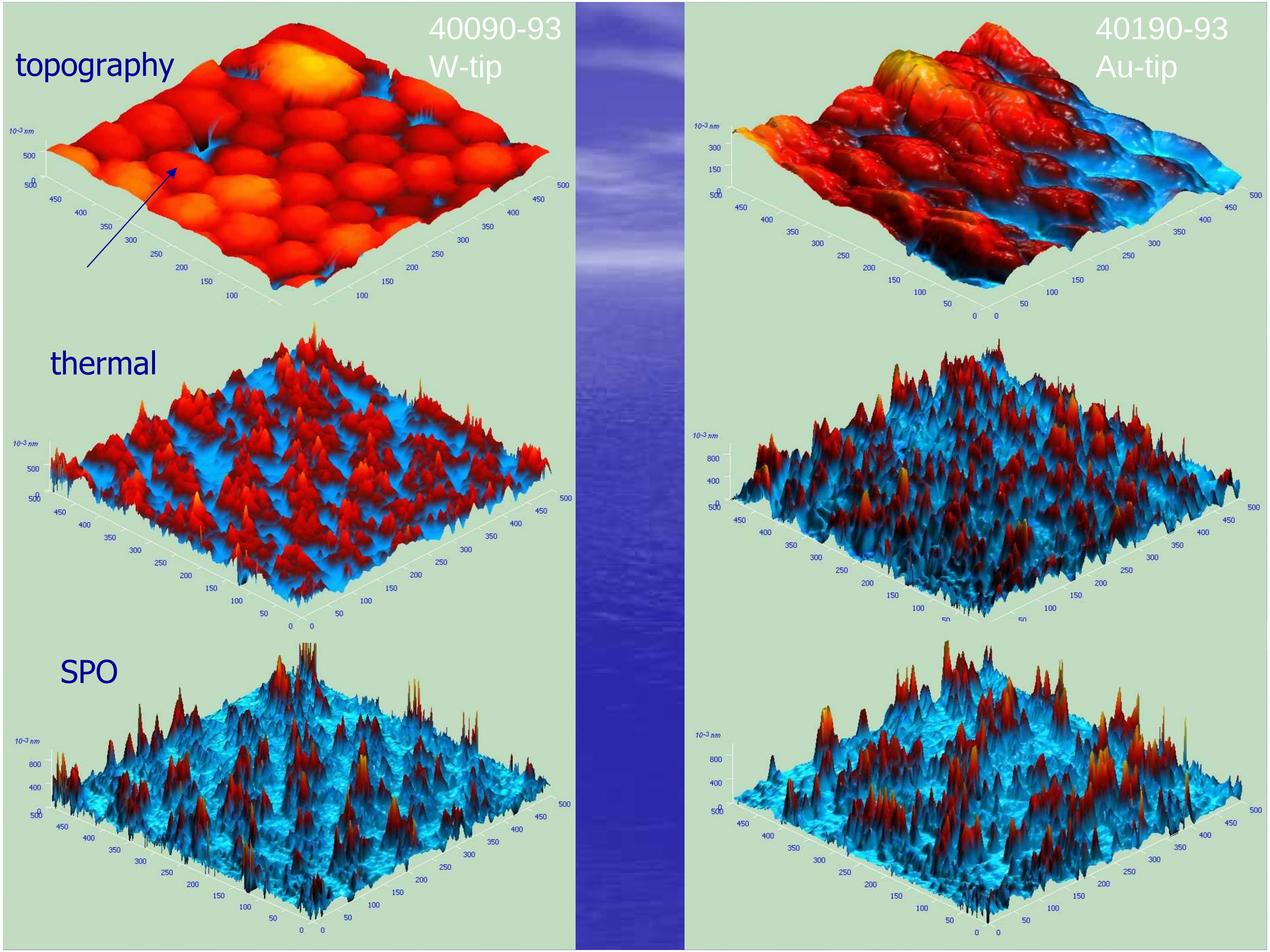




TYPICAL RESPONSE

SIGNALS AND THEIR PART

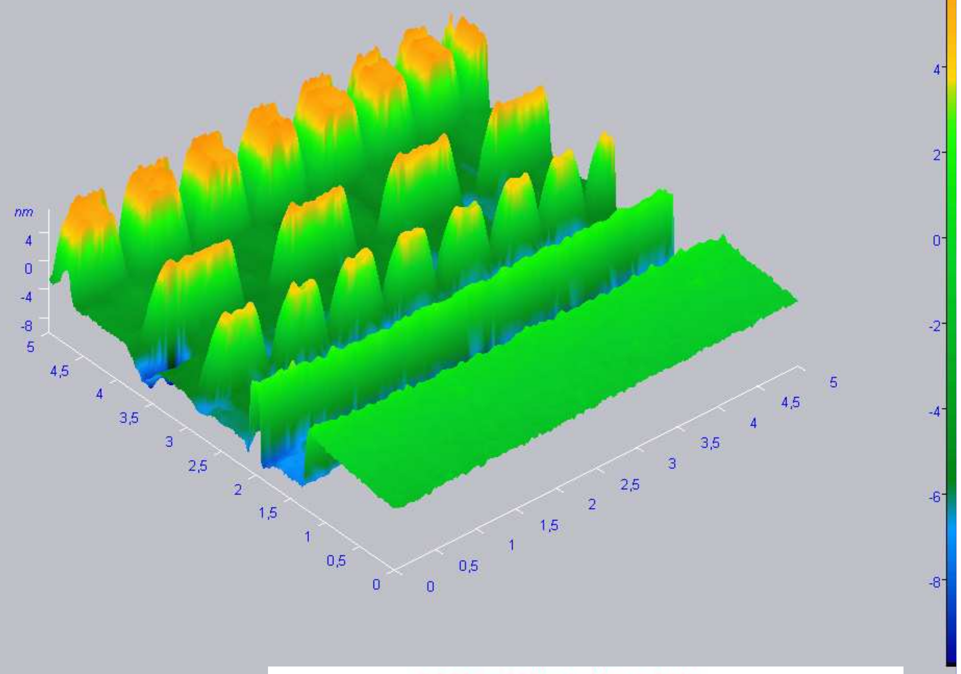
USED FOR IMAGING


40090-93
W-tip
40190-93
Au-tip
topography
thermal
SPO

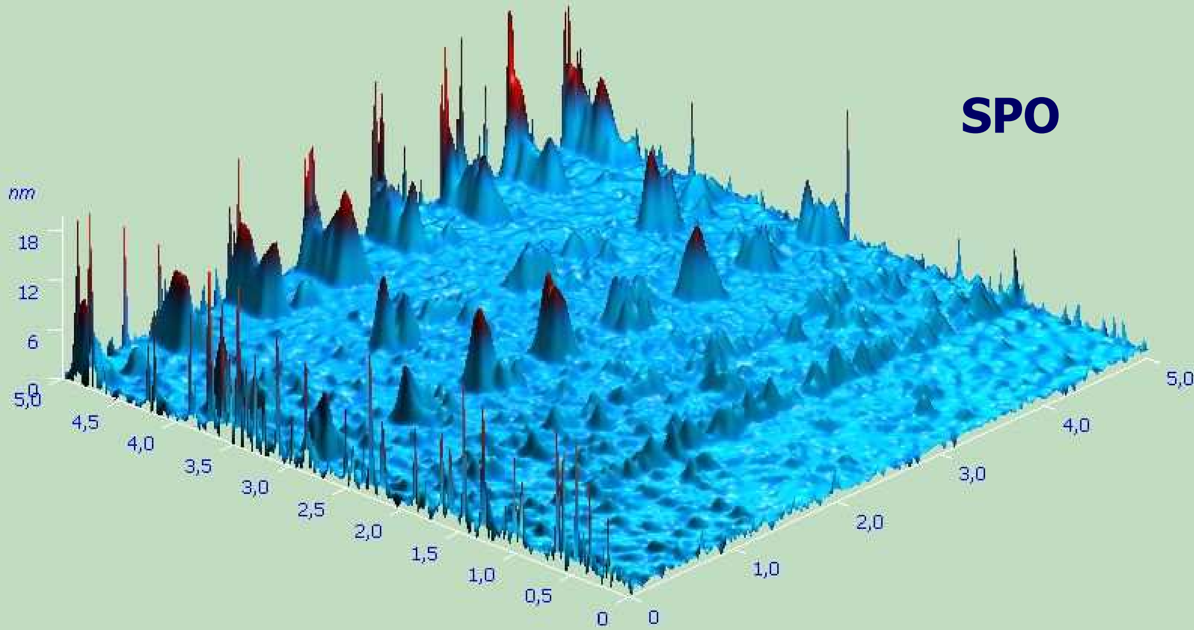
SURFACE PLASMON GUIDE

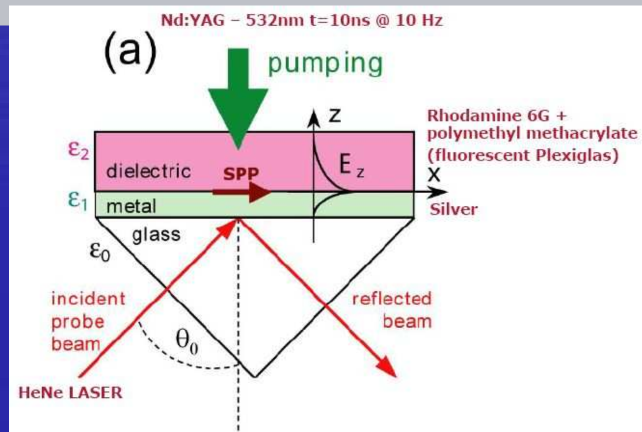
topography

4



SPO



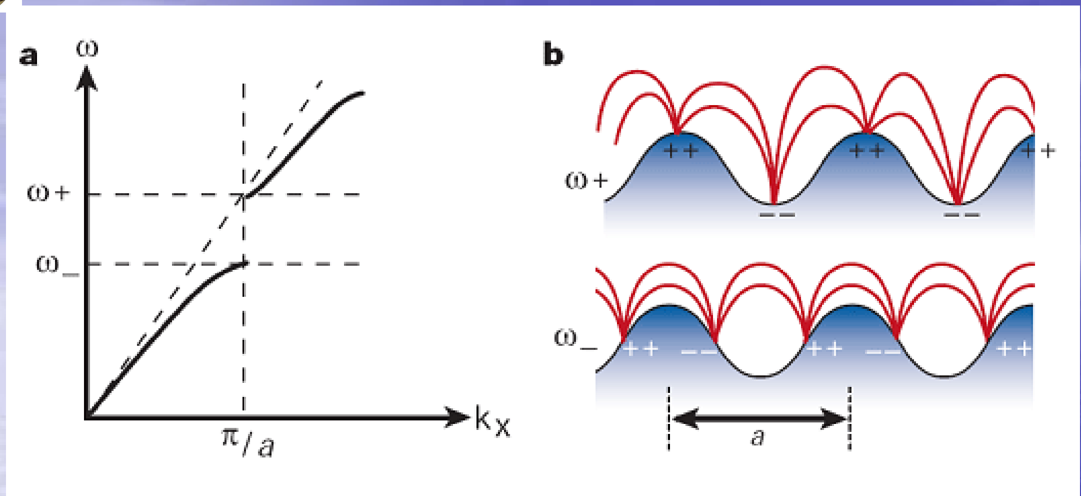


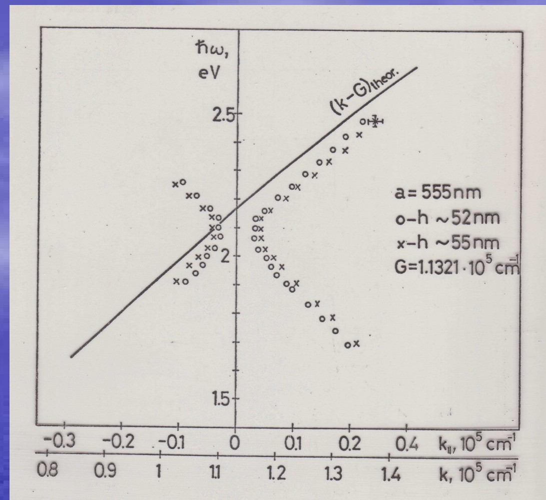
38590+92, 4000x4000nm

Gold images,Cut from

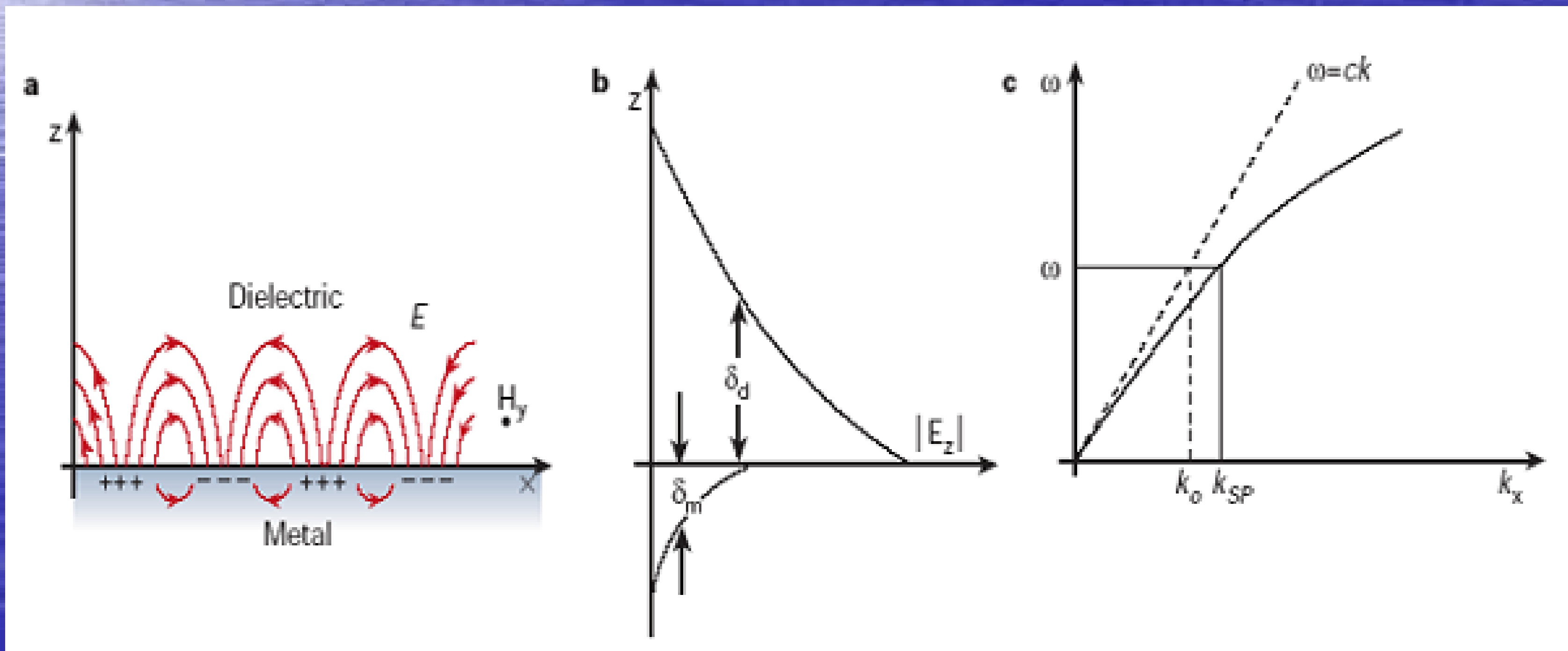
5000x5000nm images

SURFACE PLASMON ENERGY, OR MOMENTUM „GAPS”

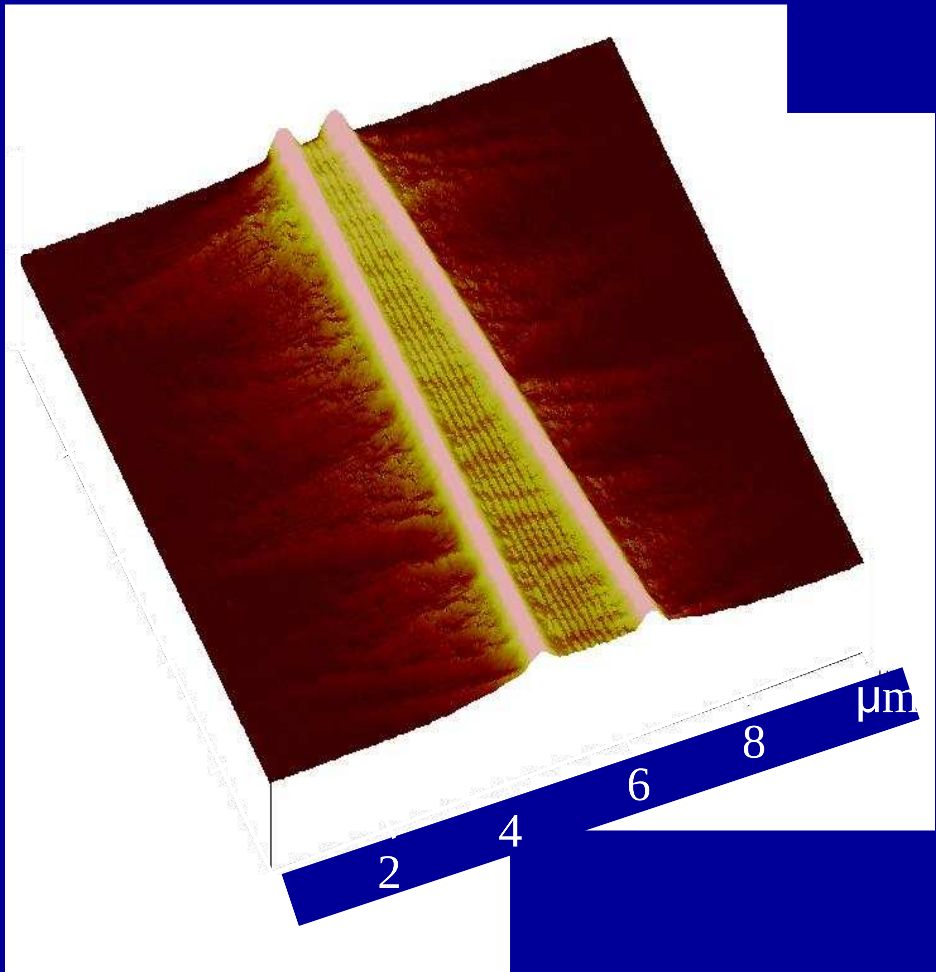


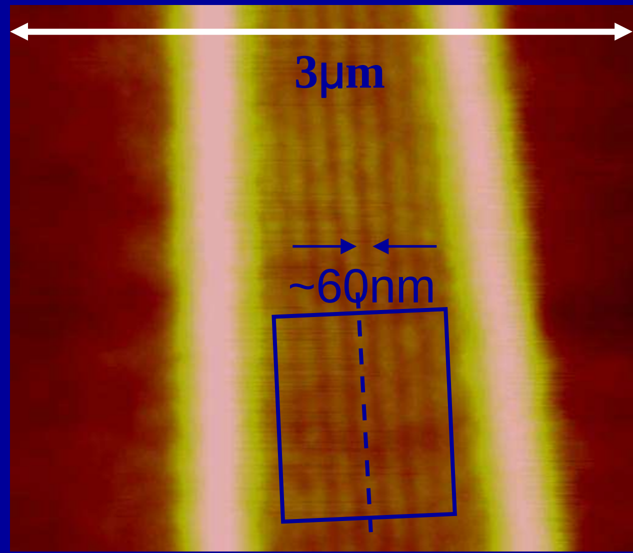


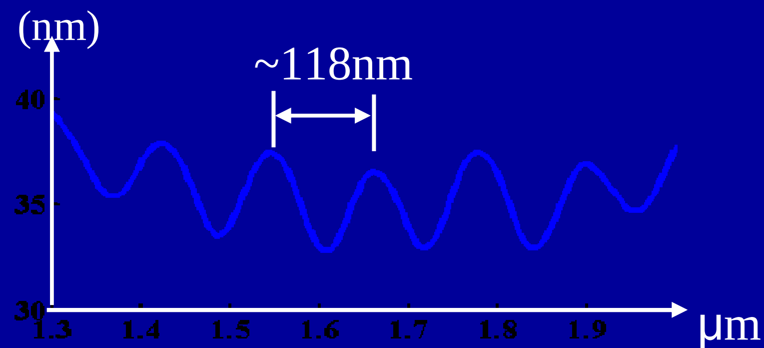
Donor (chromophore), Acceptor (fluorophore)



Litography





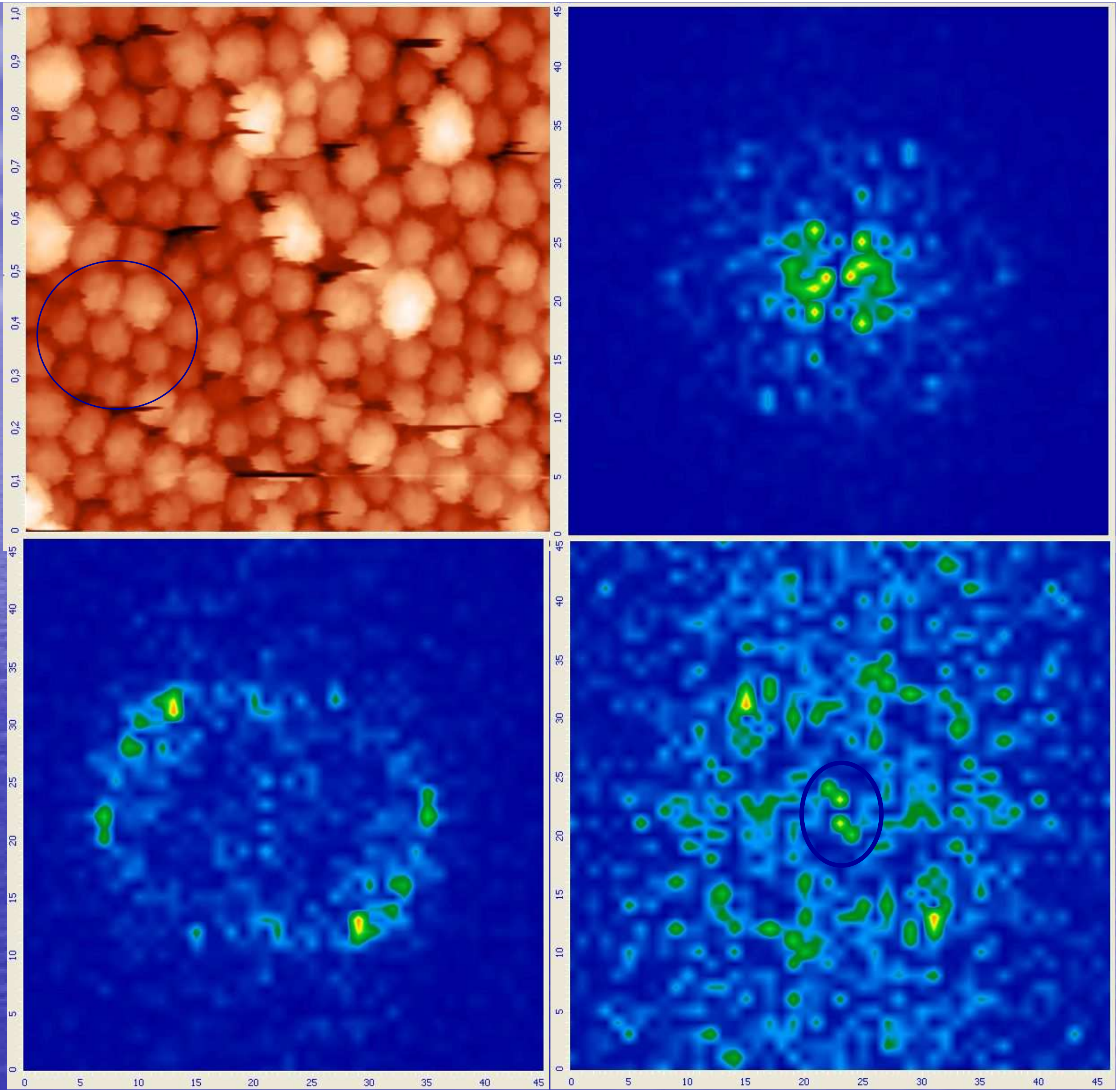


40160
topography
and FFT

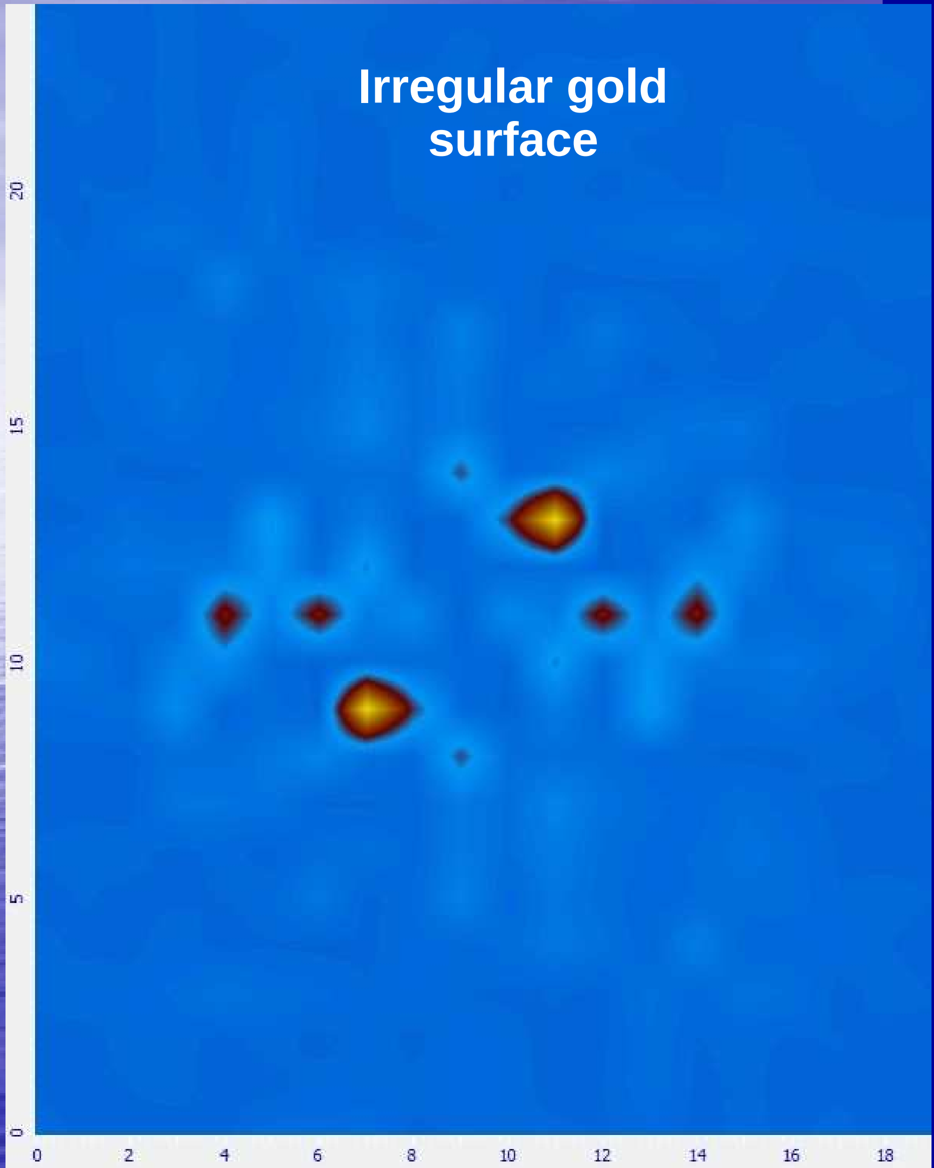
Au surface and tip,
100nm spheres
1x1µ image

40162 (thermal)
and 40163 (SPO)
FFT

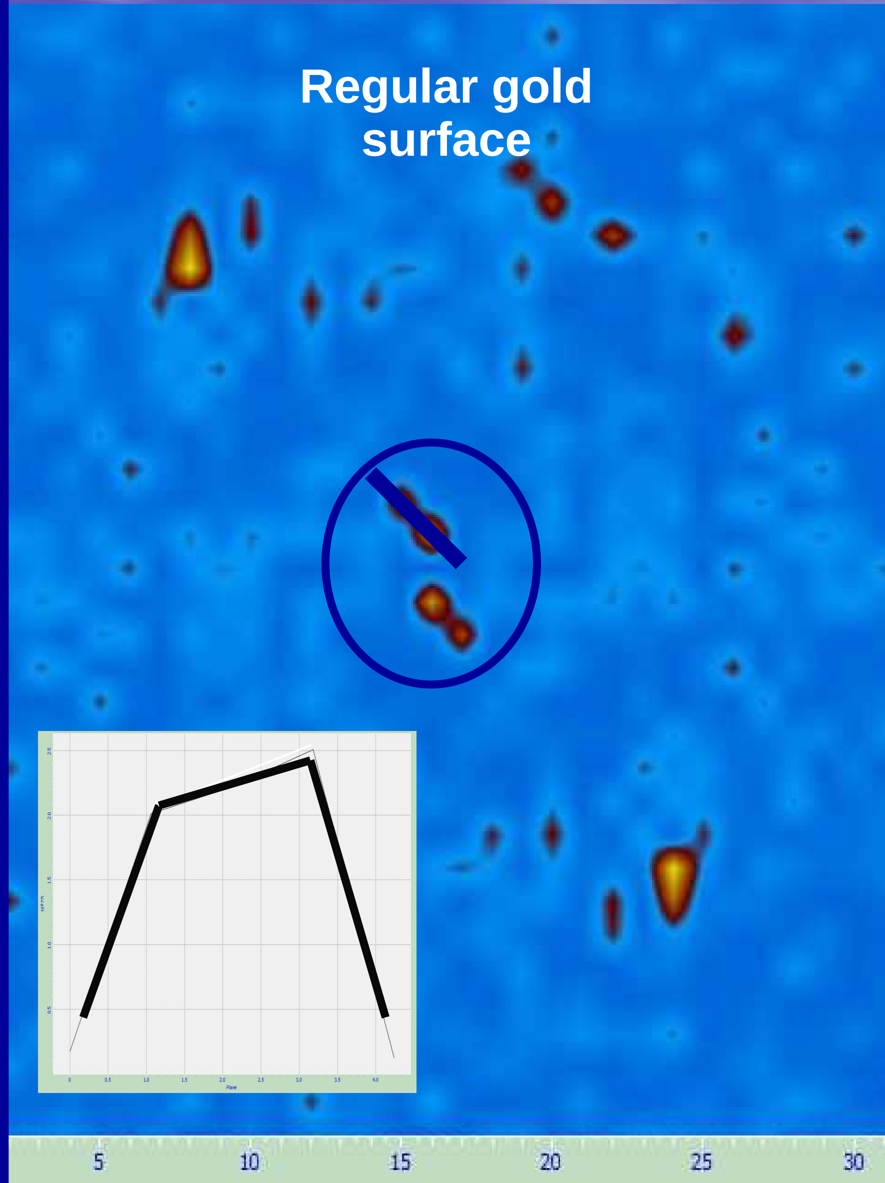
**Fano-
resonance!**



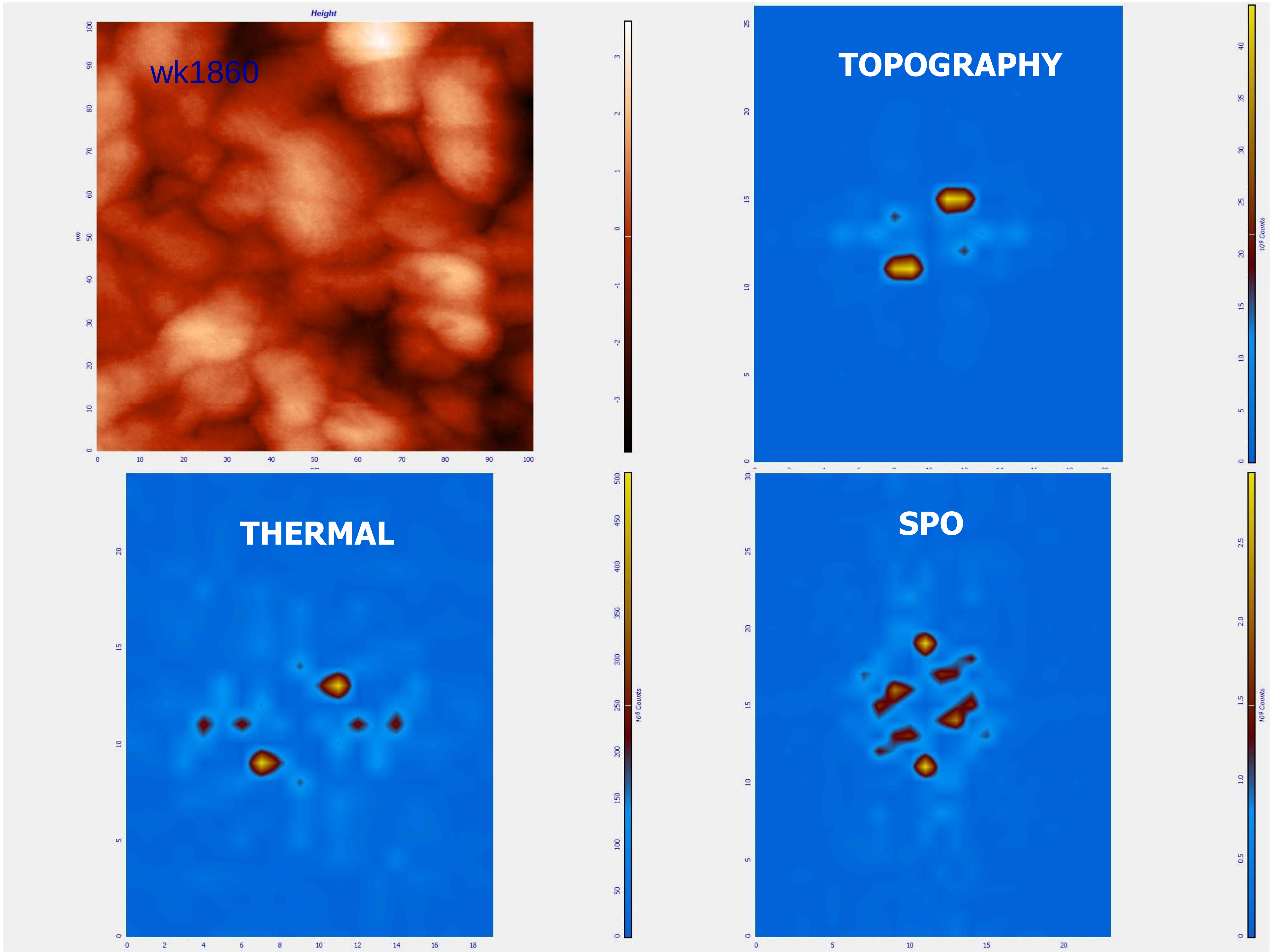
SPO IMAGES



wk01862 FFT



wk40163 FFT

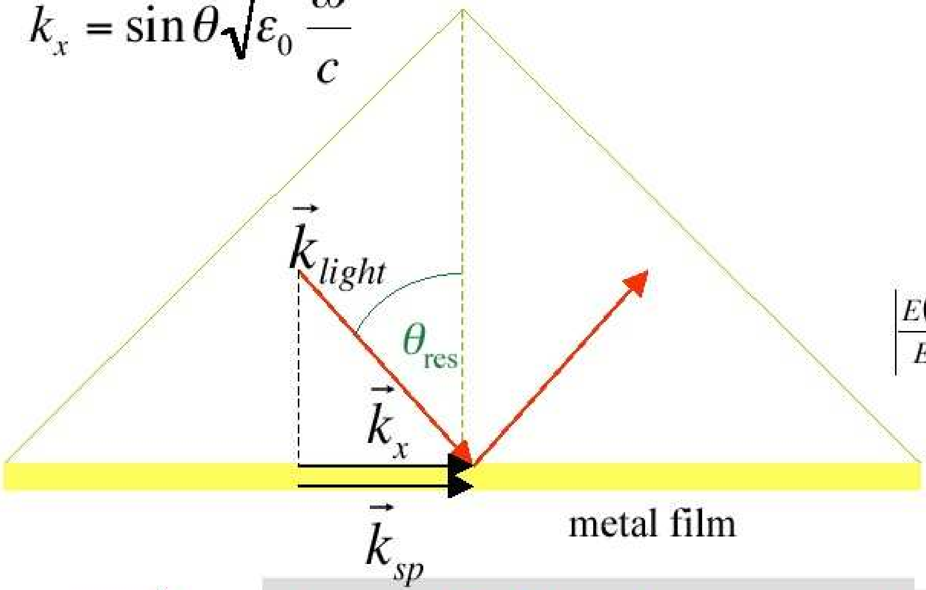

Height
wk1860
TOPOGRAPHY
THERMAL
SPO

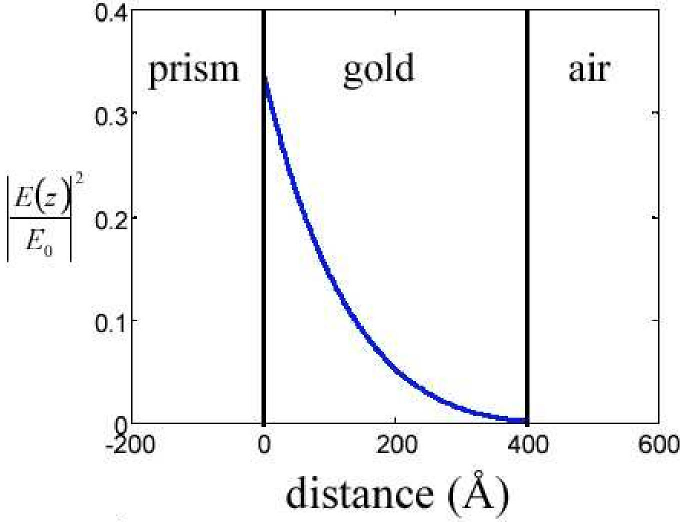
GIANT ELECTRIC FIELDS!

- Attenuated total reflection

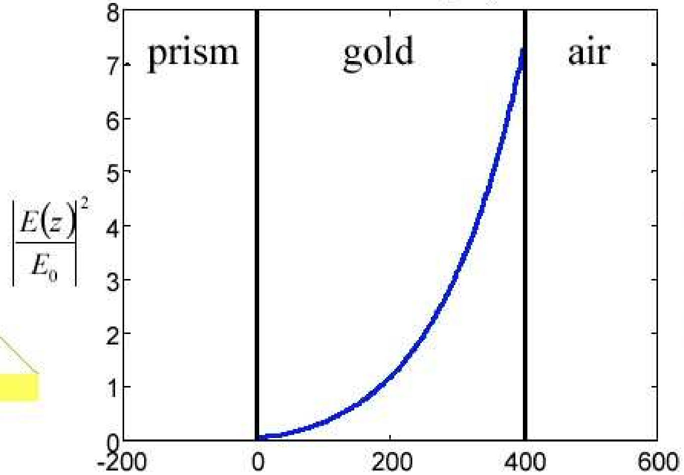
$$k_{sp} = \frac{\omega}{c}\sqrt{\frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2}} < \frac{\omega}{c}$$

$$k_x = \sin\theta \sqrt{\varepsilon_0}\,\frac{\omega}{c}$$





normal incidence

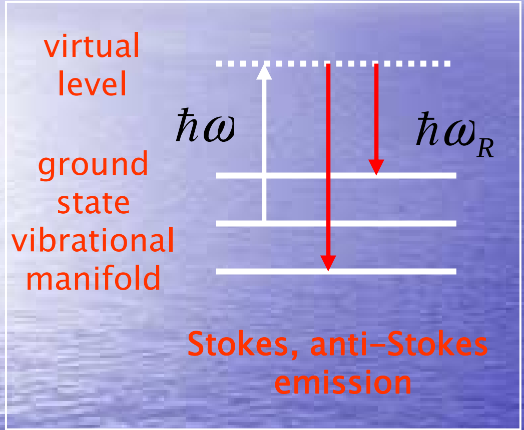


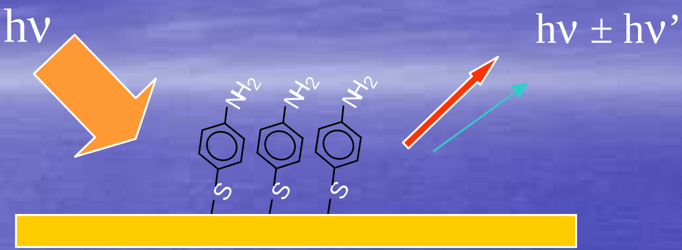
surface plasmon resonance angle

strong field enhancement effect due to surface plasmons

Surface Enhanced Raman Spectroscopy (SERS)

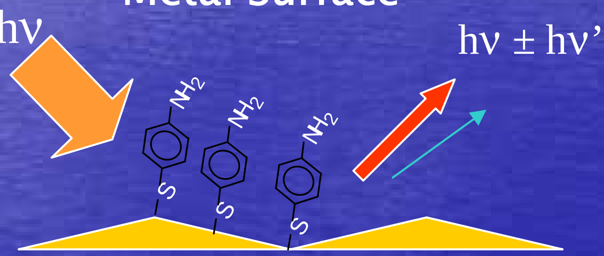
(Jeanmarie and Van Duyne, 1974)





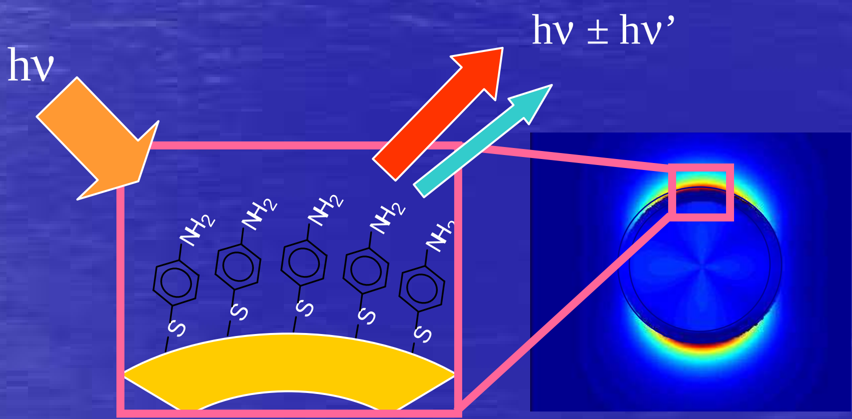
Metal Surface

Spectrum enhanced by $>10^6$ when molecule in vicinity of a metal surface



Roughened Metal Surface

Random roughness allows direct excitation of surface plasmons, greater SERS enhancements



Plasmonic SERS Substrates

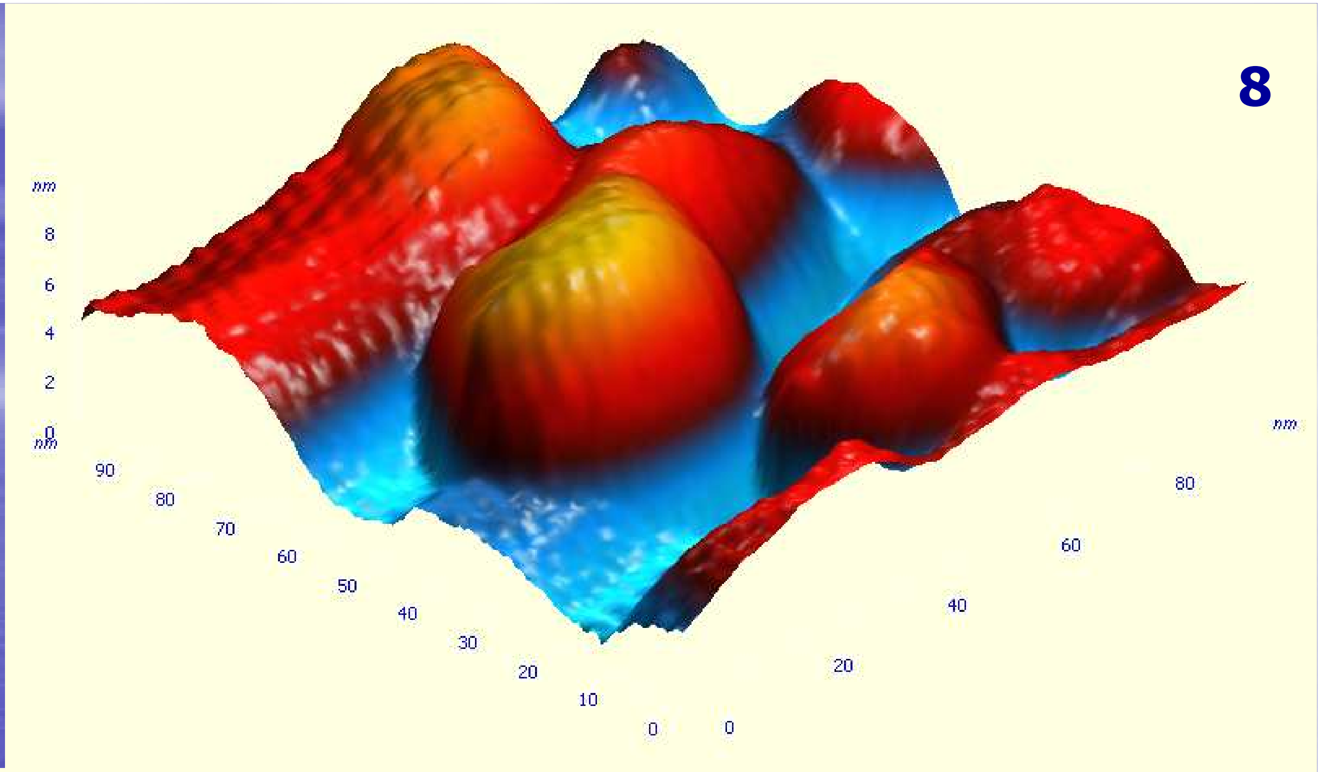
Rational design and optimization of plasmonic substrates for SERS:

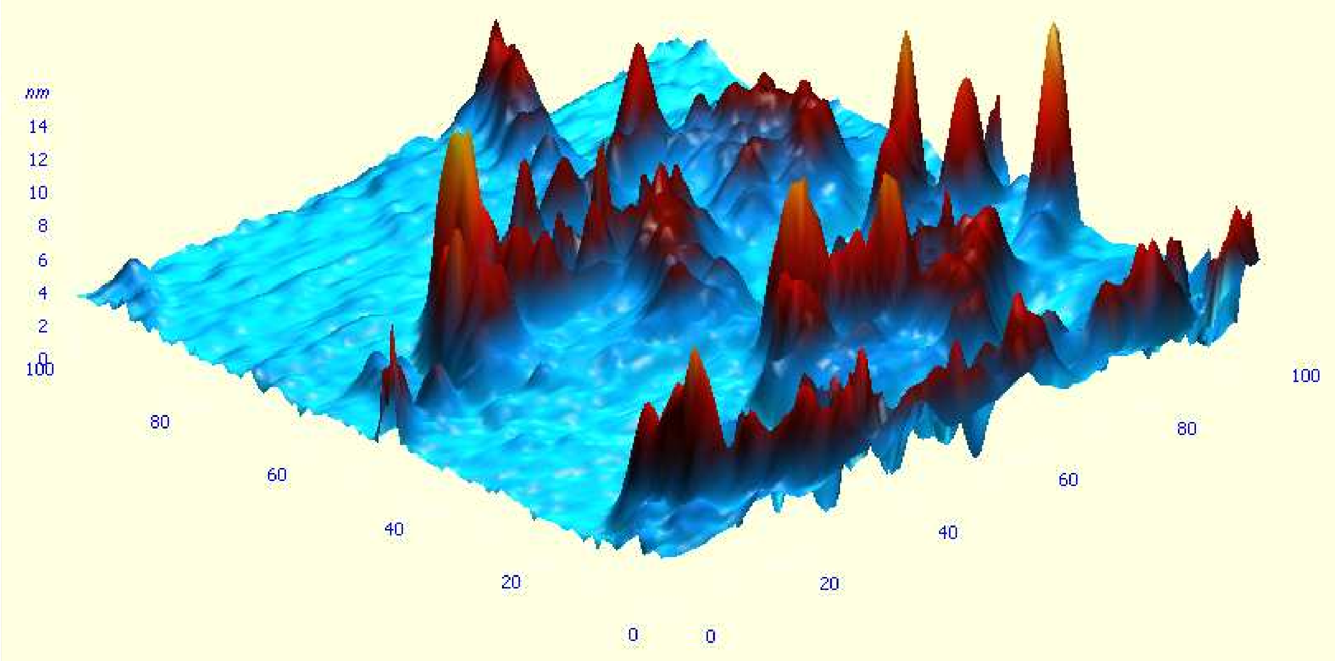
Surface Plasmon-assisted Spectroscopy

Technique	*Largest enhancement factor*
Surface enhanced raman **SERS**	10^{14} Nie and Emery, *Science*, **1997**, *275*, 1102.
Surface enhanced IR **SEIRA**	10^{4} Tsang, et.al., *Phys. Rev. Lett.*, **1980**, *45*, 201.
Sum frequency generation **SESFG**	10^{4} Baldelli, et.al., *J. Chem.Phys.*, **2000**, *113*, 5432.
Second harmonic generation **SESHG**	10^{4} Chen, et.al., *Phys. Rev. Lett.*, **1981**, *46*, 145.
Surface enhanced fluorescence **SEF**	~100

100x100nm GOLD

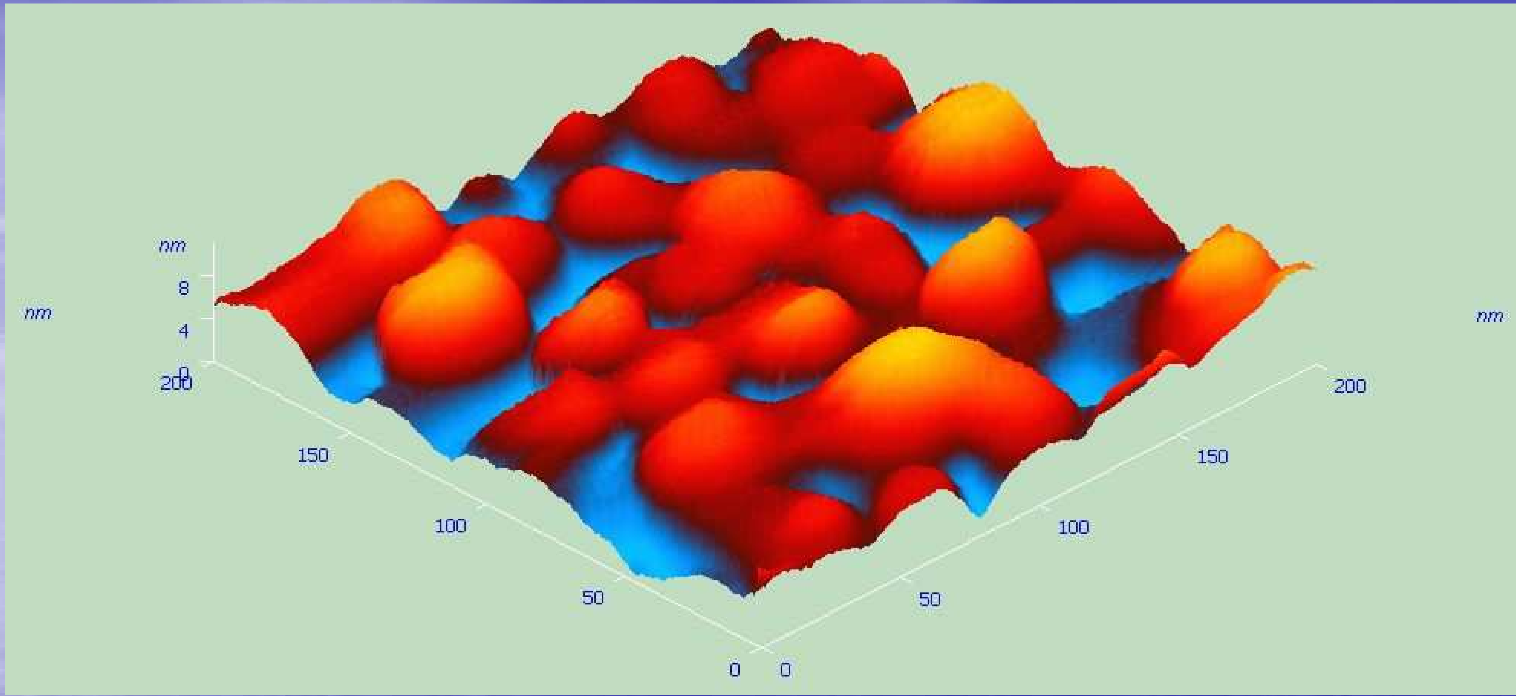
Topography and SPO near field STM image



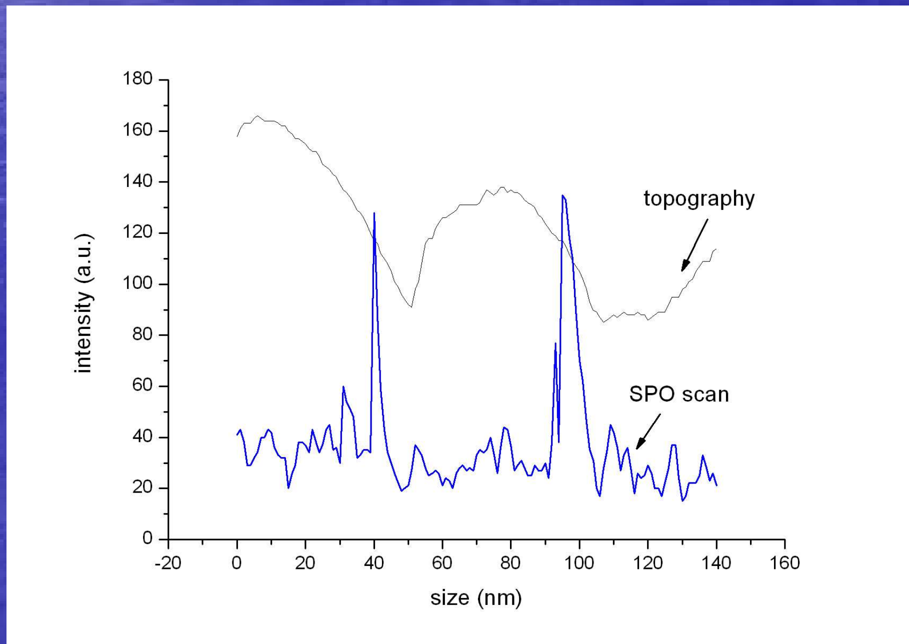


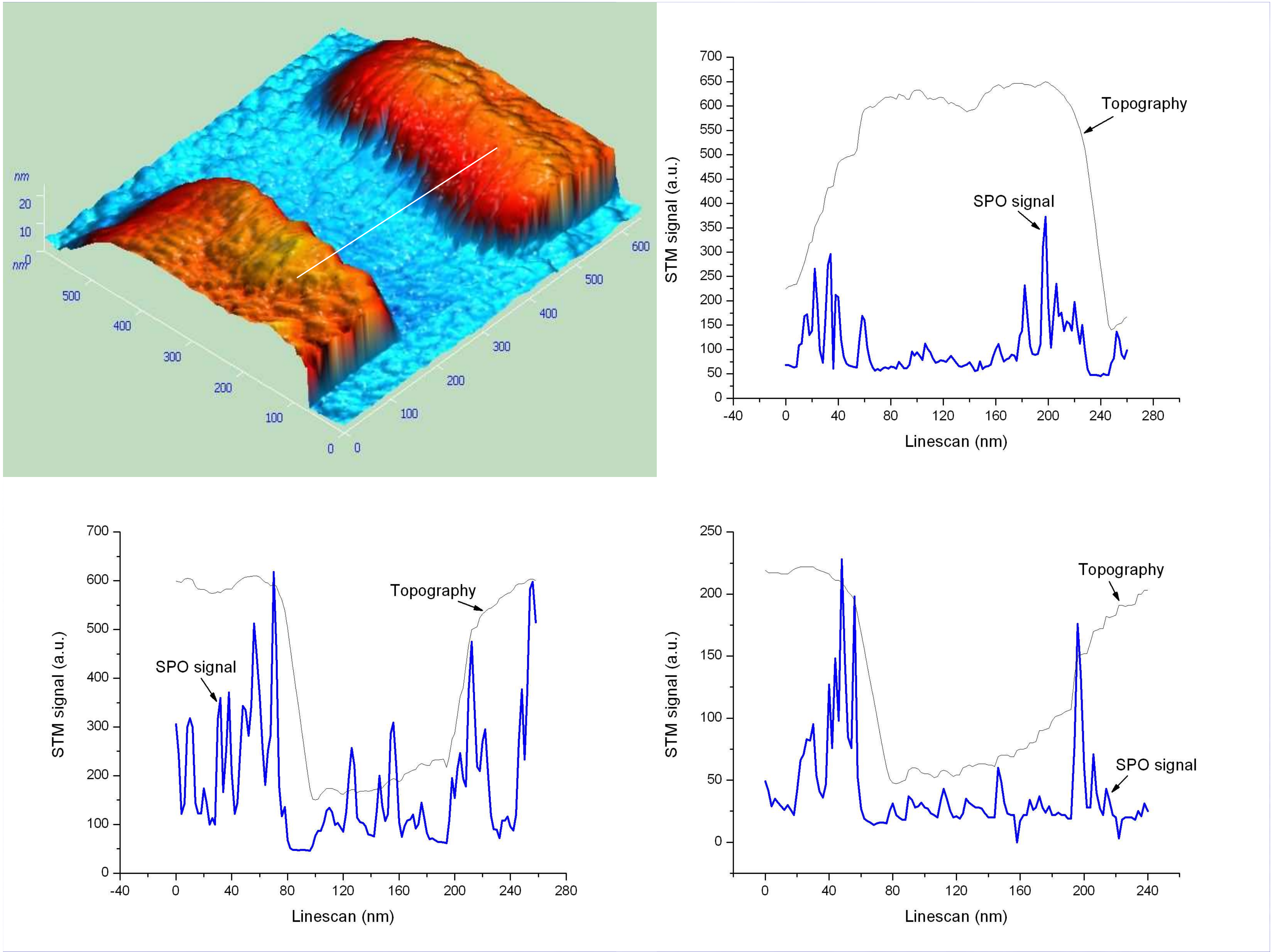
34300+02 Au

Cut from 200x200nm images



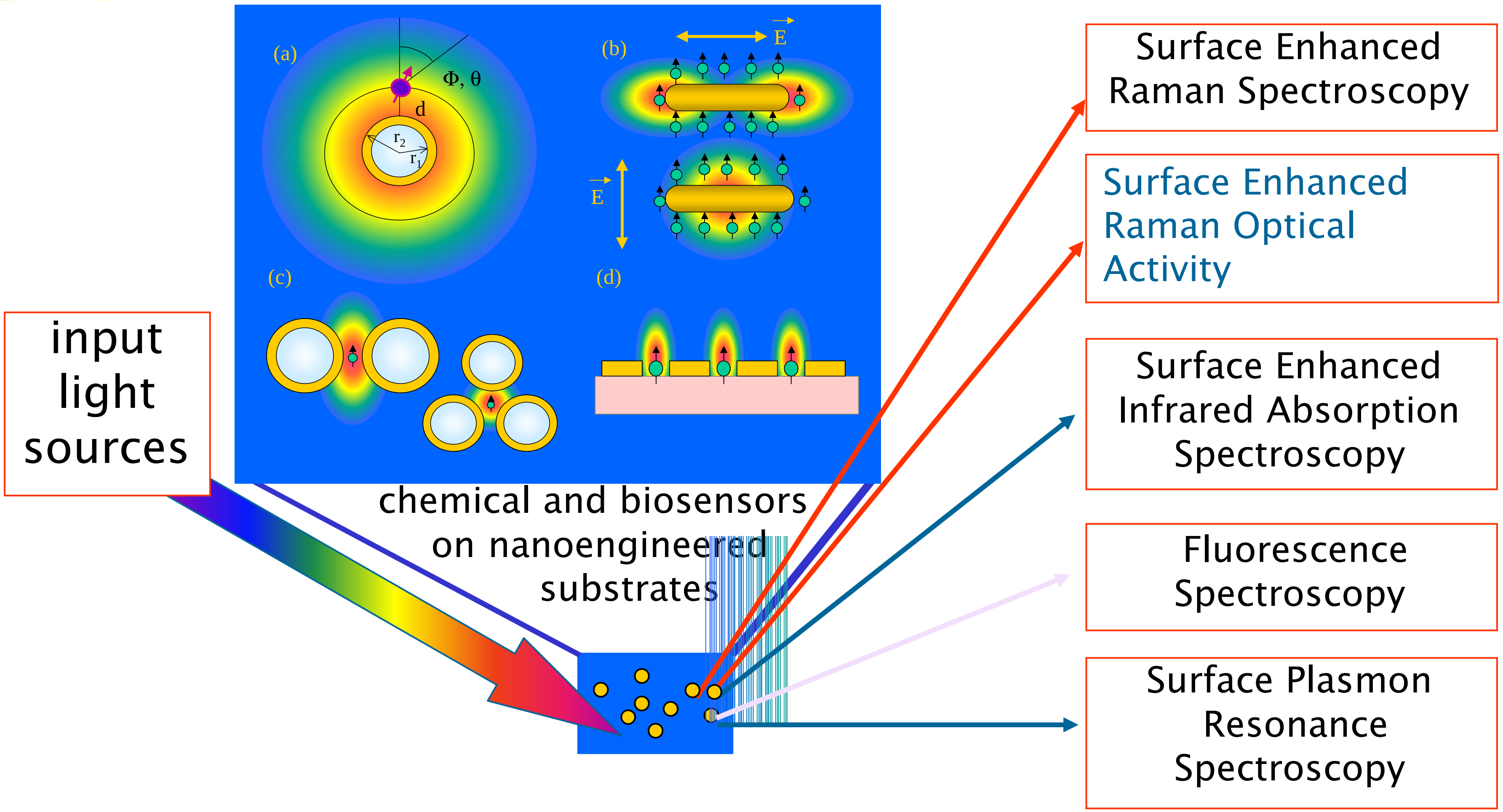
34300
Pt/Ir-tip



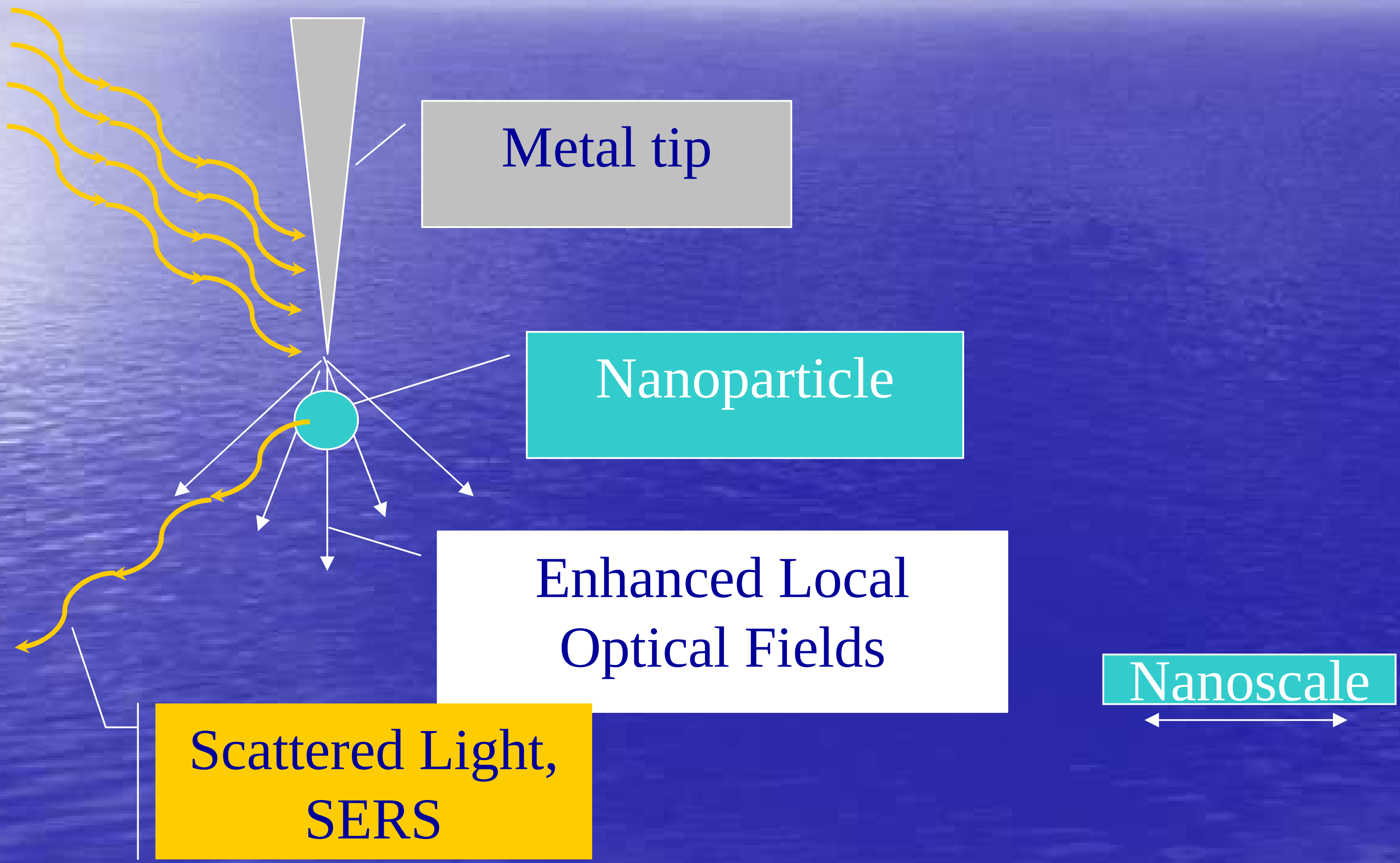

nm
20
10
0
nm
500
400
300
200
100
0
0
100
200
300
400
500
600
STM signal (a.u.)
700
650
600
550
500
450
400
350
300
250
200
150
100
50
0
-40
0
40
80
120
160
200
240
280
Linescan (nm)
Topography
SPO signal
STM signal (a.u.)
700
600
500
400
300
200
100
0
-40
0
40
80
120
160
200
240
280
Linescan (nm)
SPO signal
Topography
STM signal (a.u.)
250
200
150
100
50
0
0
40
80
120
160
200
240
Linescan (nm)
Topography
SPO signal

Chemical and biological sensing

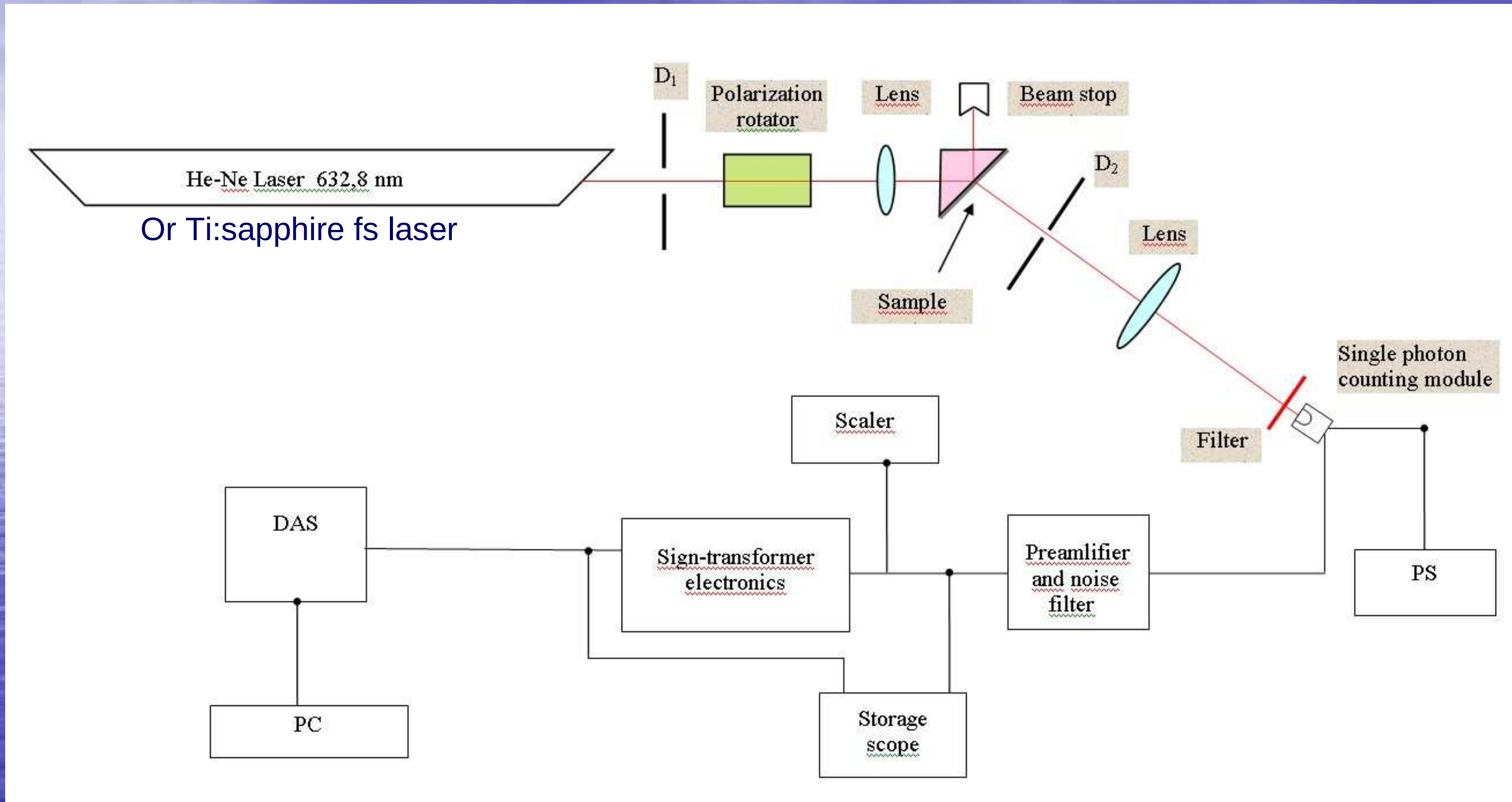
The strong focusing of E&M waves into nanosize spots can lead to large local electric fields and enormously enhanced cross sections for nonlinear spectroscopies.



THE USE OF ENHANCED LOCAL FIELDS FOR NANO-MICROSCOPY

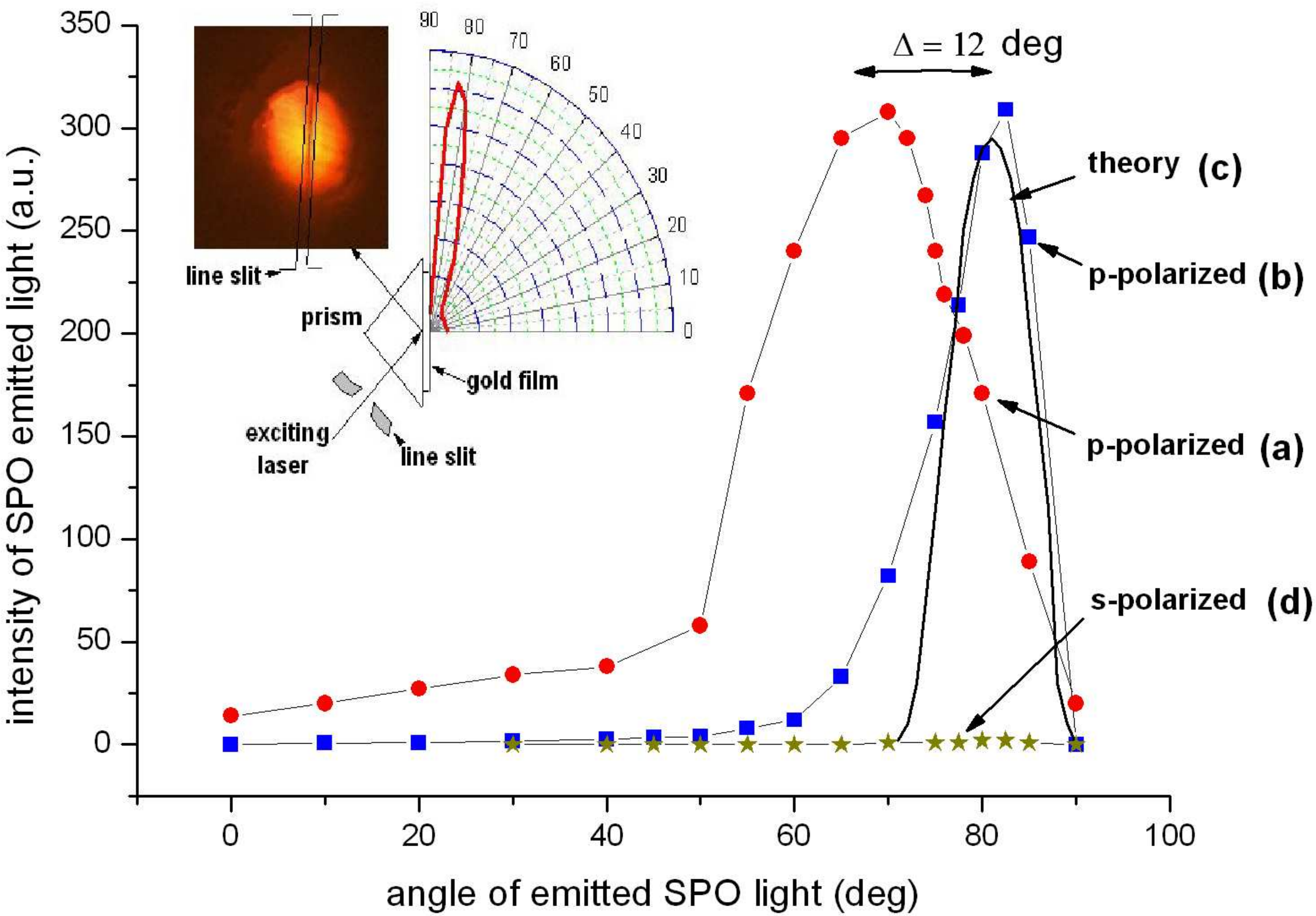


TO EXPLORE THE FIELD ENHANCEMENT EFFECT OF SURFACE PLASMONS

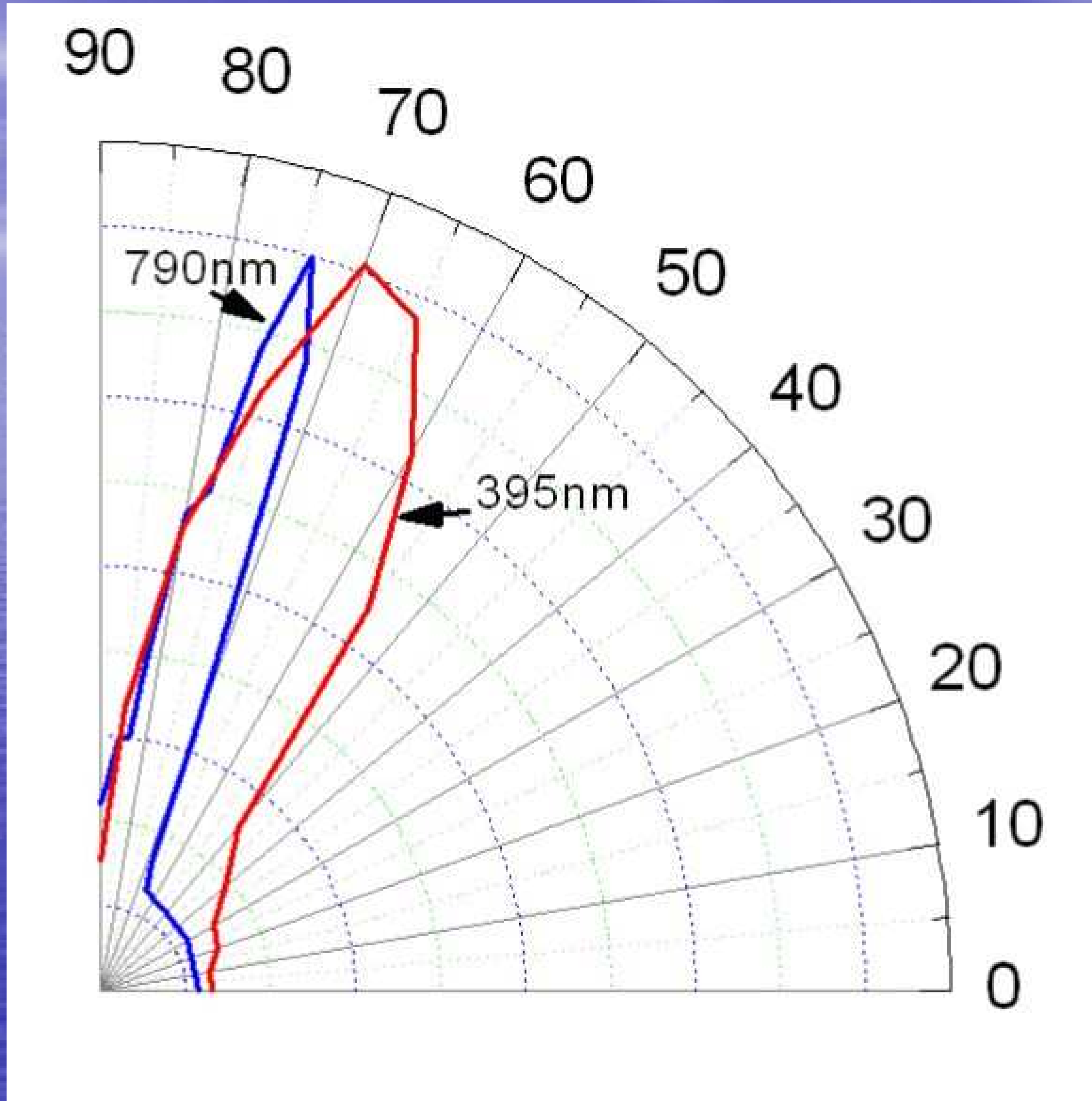


Simple layout, but sophisticated fast electronics and detector (at 0° and 70°)

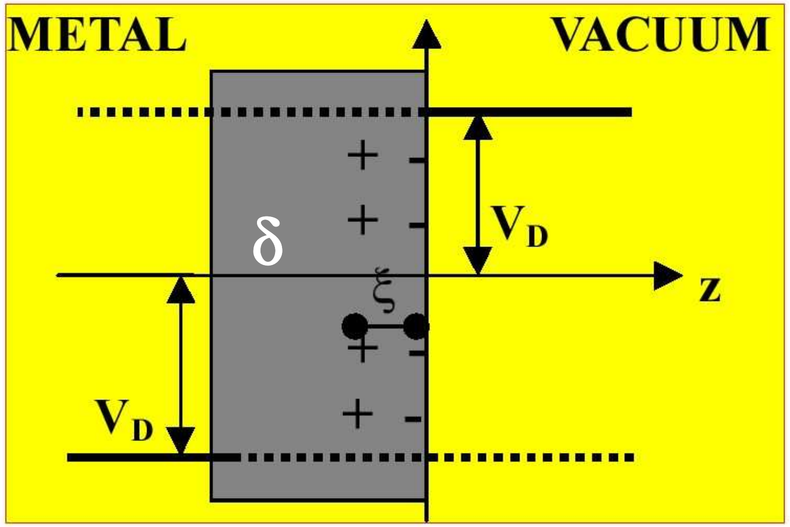
Opt.Expr. Vol16, No26, 21656-61 (2008)



intensity of SPO emitted light (a.u.)
angle of emitted SPO light (deg)
Δ = 12 deg
theory (c)
p-polarized (b)
p-polarized (a)
s-polarized (d)
line slit
prism
gold film
exciting laser
line slit


90
80
70
60
50
40
30
20
10
0
790nm
395nm

MODELLING SURFACE PLASMONS



Electric field inside : $F = F_0 \exp(z'/\delta) \sin\omega t$, where δ = skin depth
Displacement inside : $\xi(z',t) = a_0 \exp(z'/\delta) \sin\omega t$, $a_0 = eF_0/m\omega^2$

Oscillating Double Layer Potential :

$V_D = (\omega_p/4\omega)\ m\ (2mc^2)$

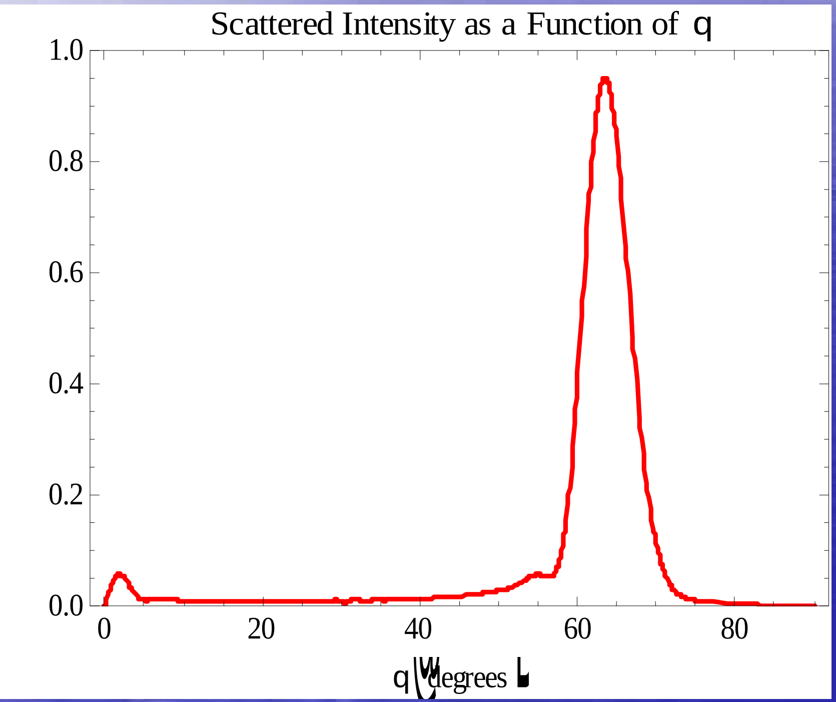
$\mu = eF_0/m\omega c = v_{osc}/c$ = intensity parameter = $10^{-9}\ I^{1/2} / E_{ph}$

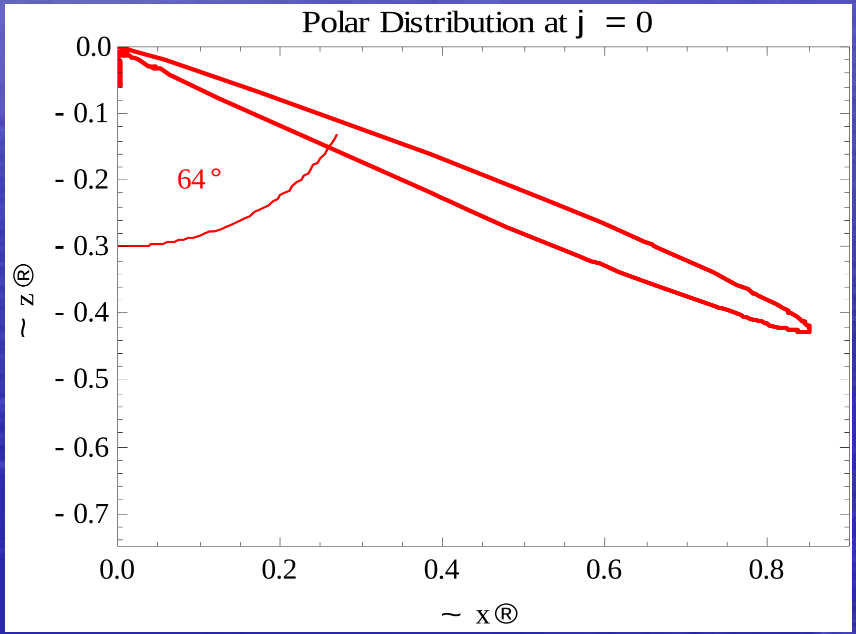
-$U_d(x,z,t) = U_d^{(1)}(x,z,t) + U_d^{(2)}(x,z,t)$

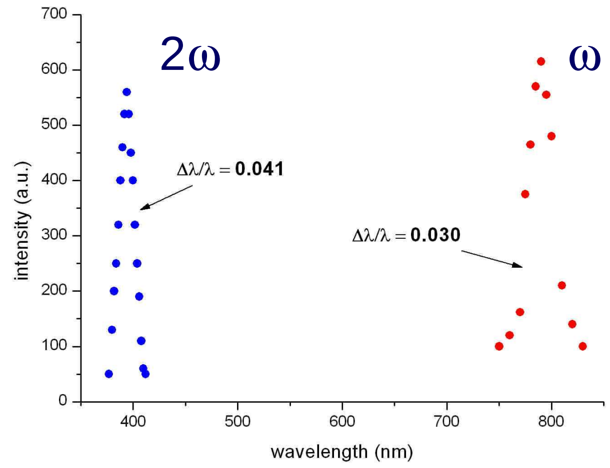
-ENHANCEMENT ~ $(\omega_p/\omega_0)^2$ (ADDITIONAL: ROUGHNESS + TIP ENHANCEMENT!)

-THE QUADRATIC TERM CAN RESULT IN SECOND HARMONIC GENERATION AND EVEN SQUEEZING

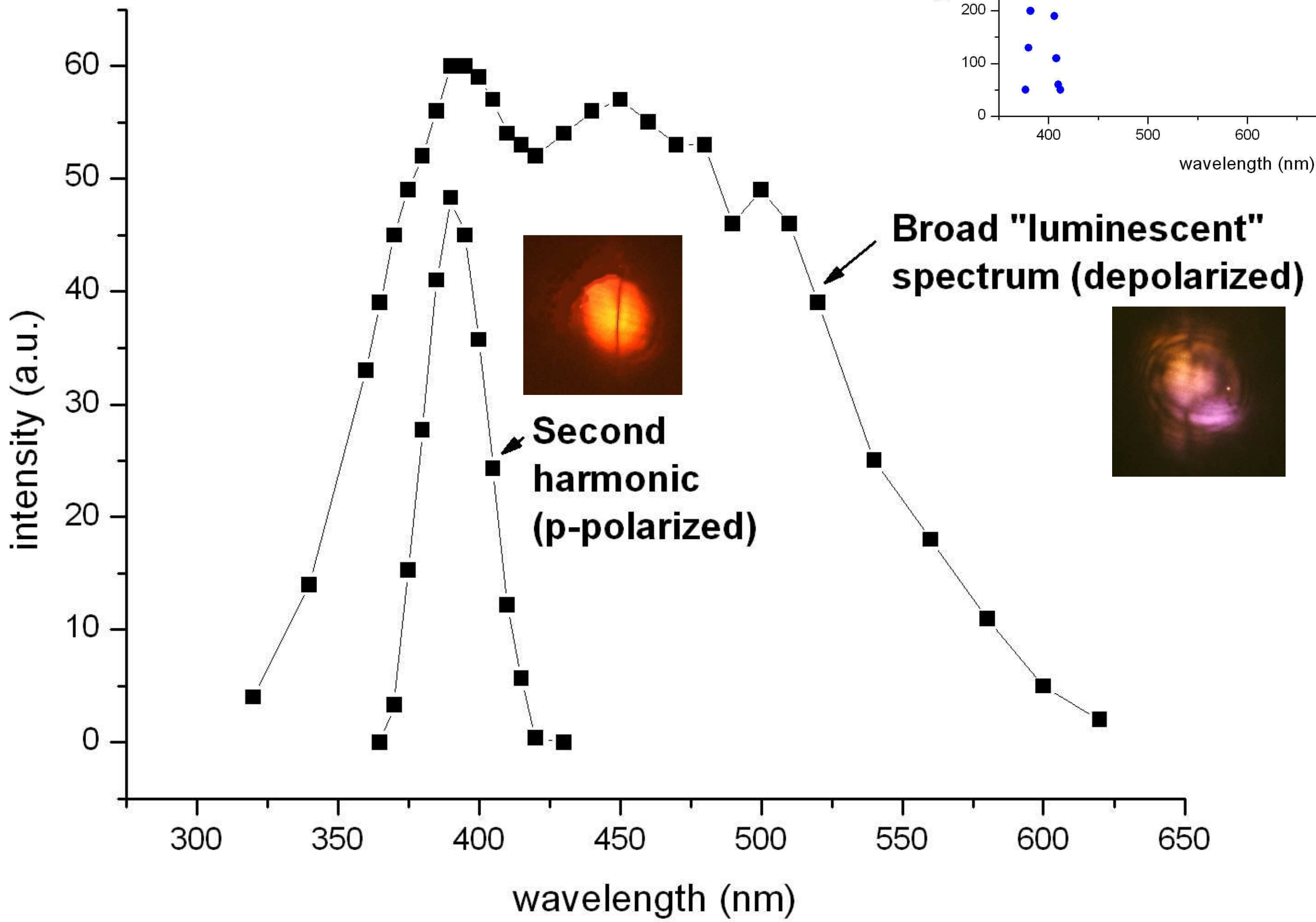
Polar distribution of the scattered fundamental radiation at φ = 0



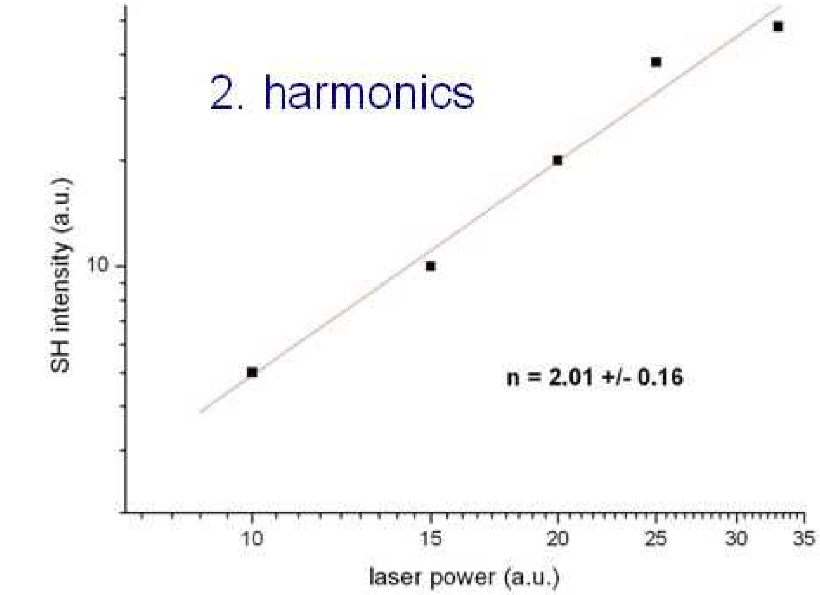


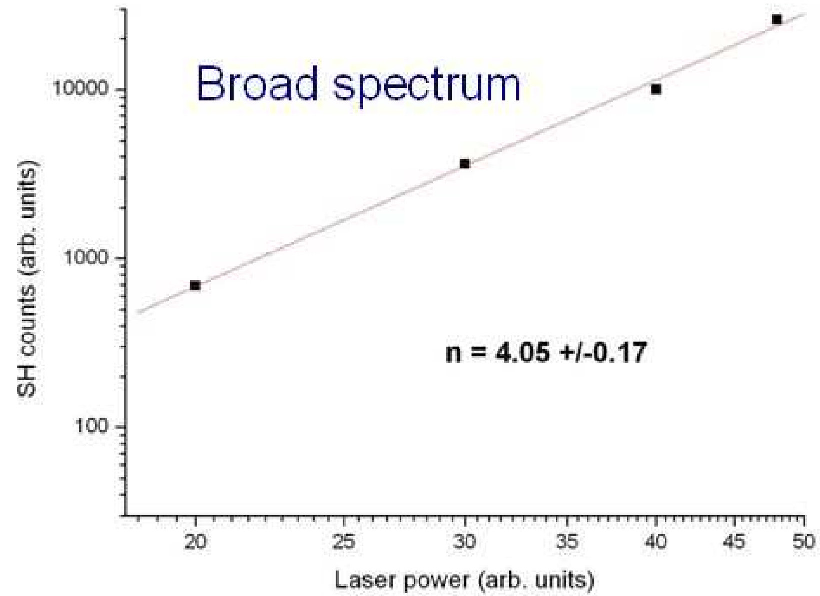


Δλ/λ = 0.041
Δλ/λ = 0.030
intensity (a.u.)
wavelength (nm)



Broad "luminescent" spectrum (depolarized)
Second harmonic (p-polarized)
intensity (a.u.)
wavelength (nm)


2. harmonics
SH intensity (a.u.)
10
n = 2.01 +/- 0.16
10 15 20 25 30 35
laser power (a.u.)


Broad spectrum
SH counts (arb. units)
10000
1000
100
n = 4.05 +/-0.17
20 25 30 35 40 45 50
Laser power (arb. units)

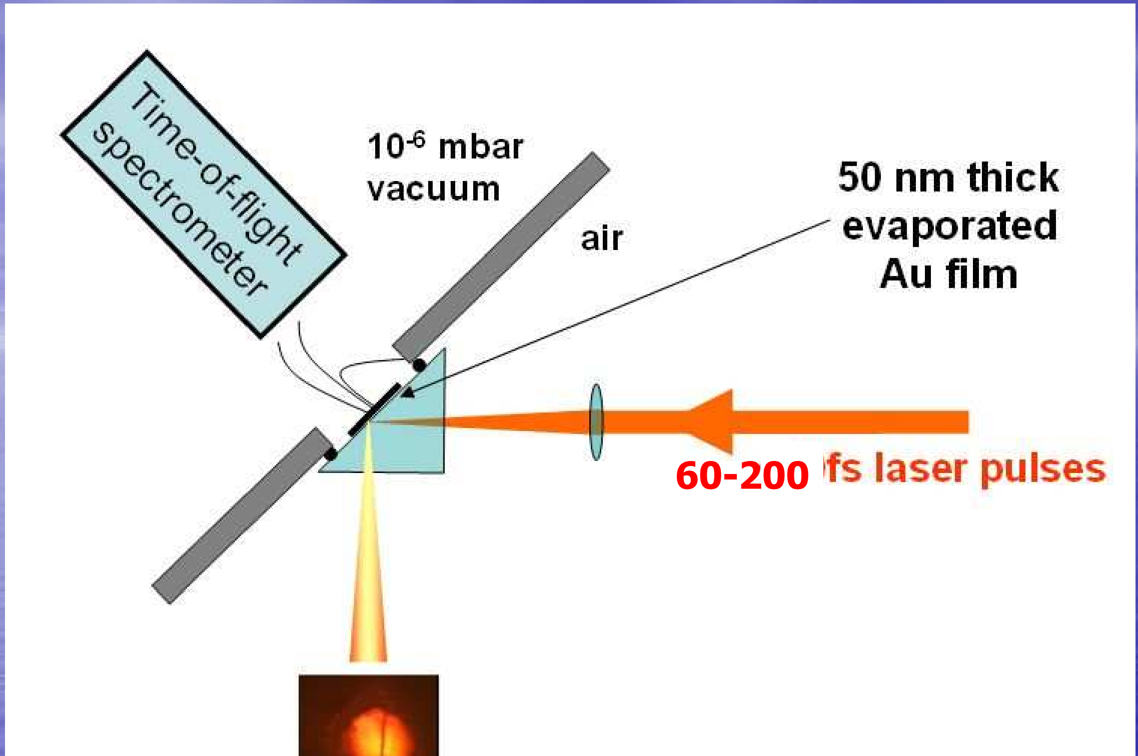
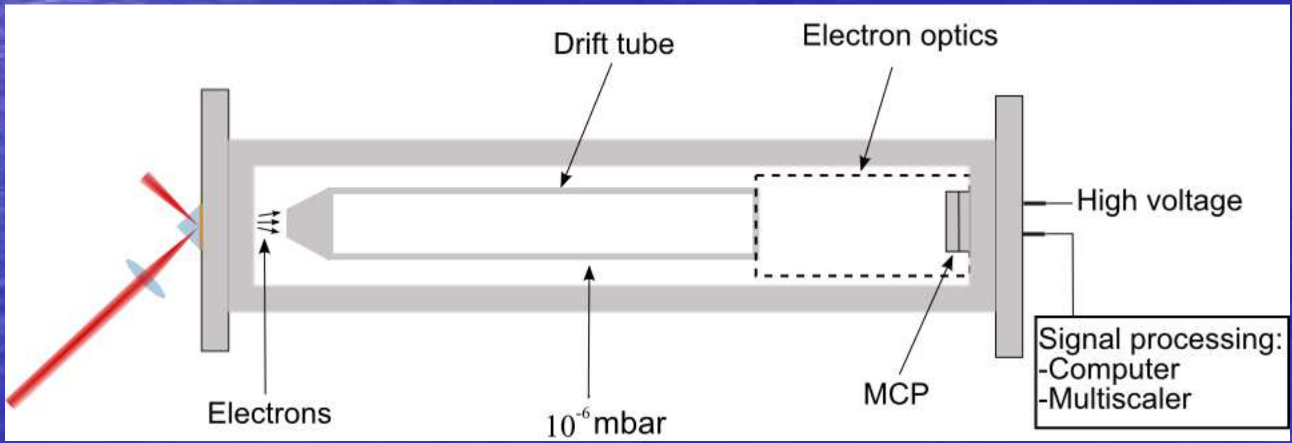
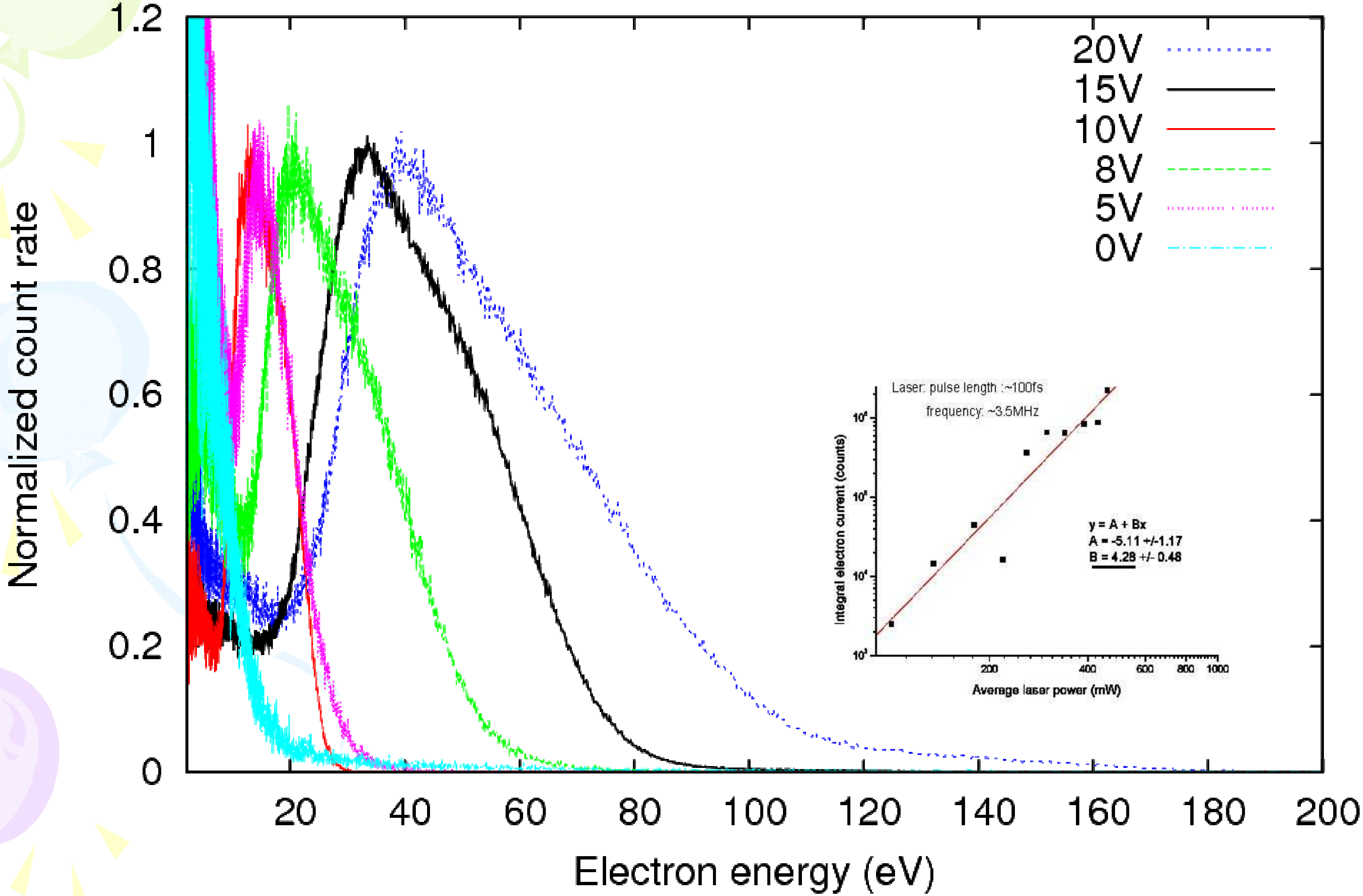
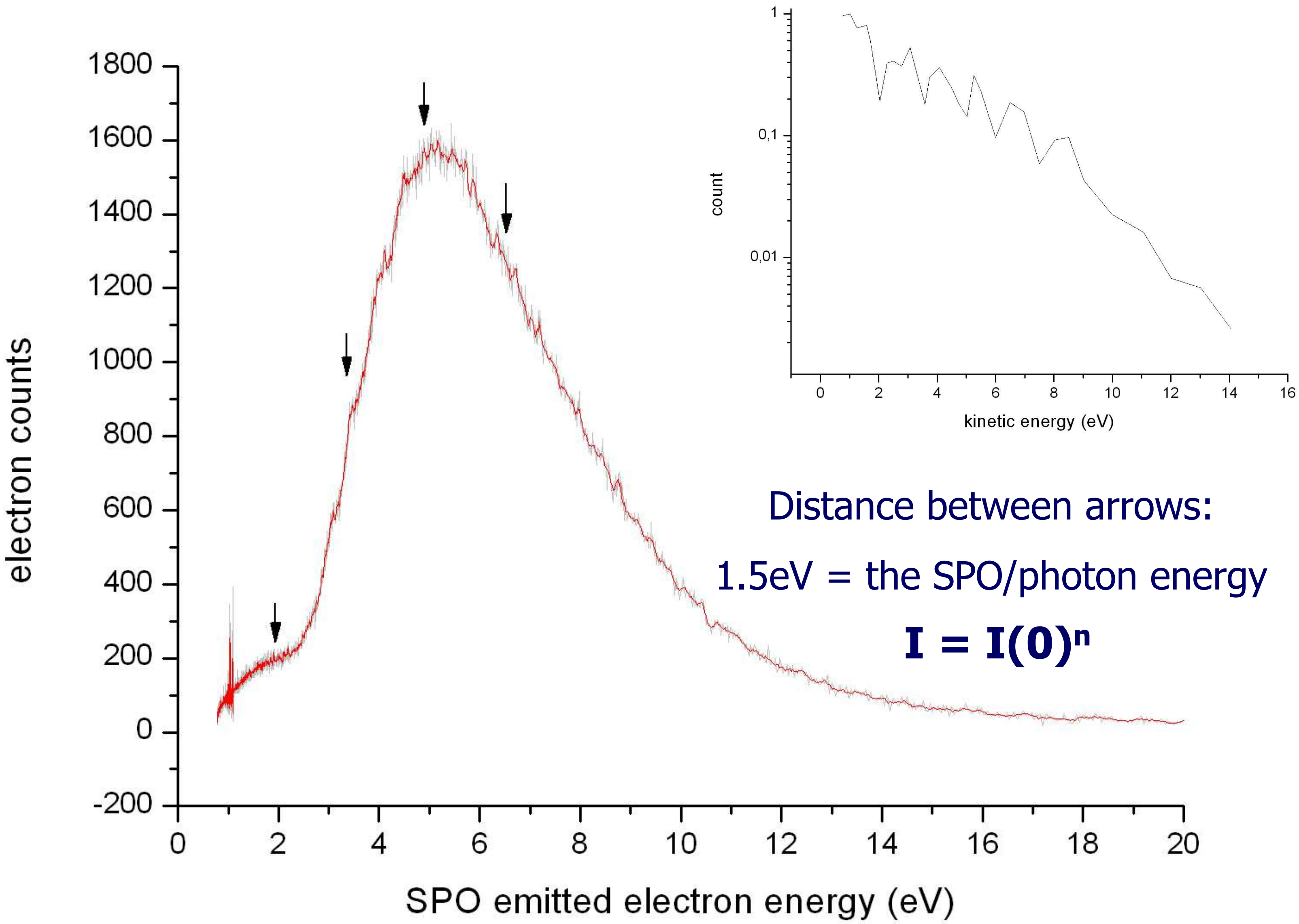

Time-of-flight spectrometer
10^{-6} mbar vacuum
air
50 nm thick evaporated Au film
60-200 fs laser pulses

Figure 1a


Drift tube
Electron optics
High voltage
Electrons
10^{-6}mbar
MCP
Signal processing:
-Computer
-Multiscaler

Laser: 400mW, 1.2kHz; Sample 2





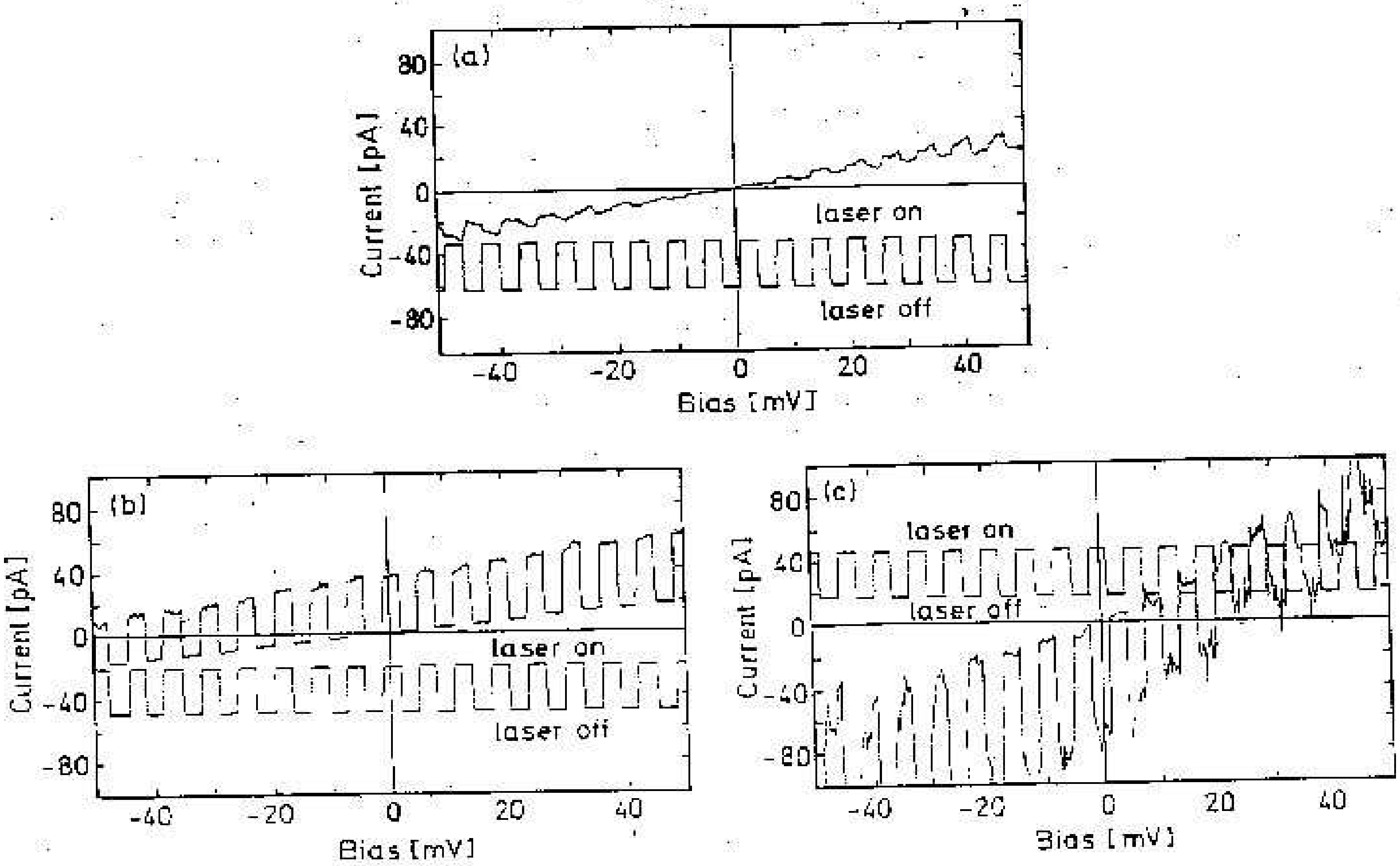
Distance between arrows:

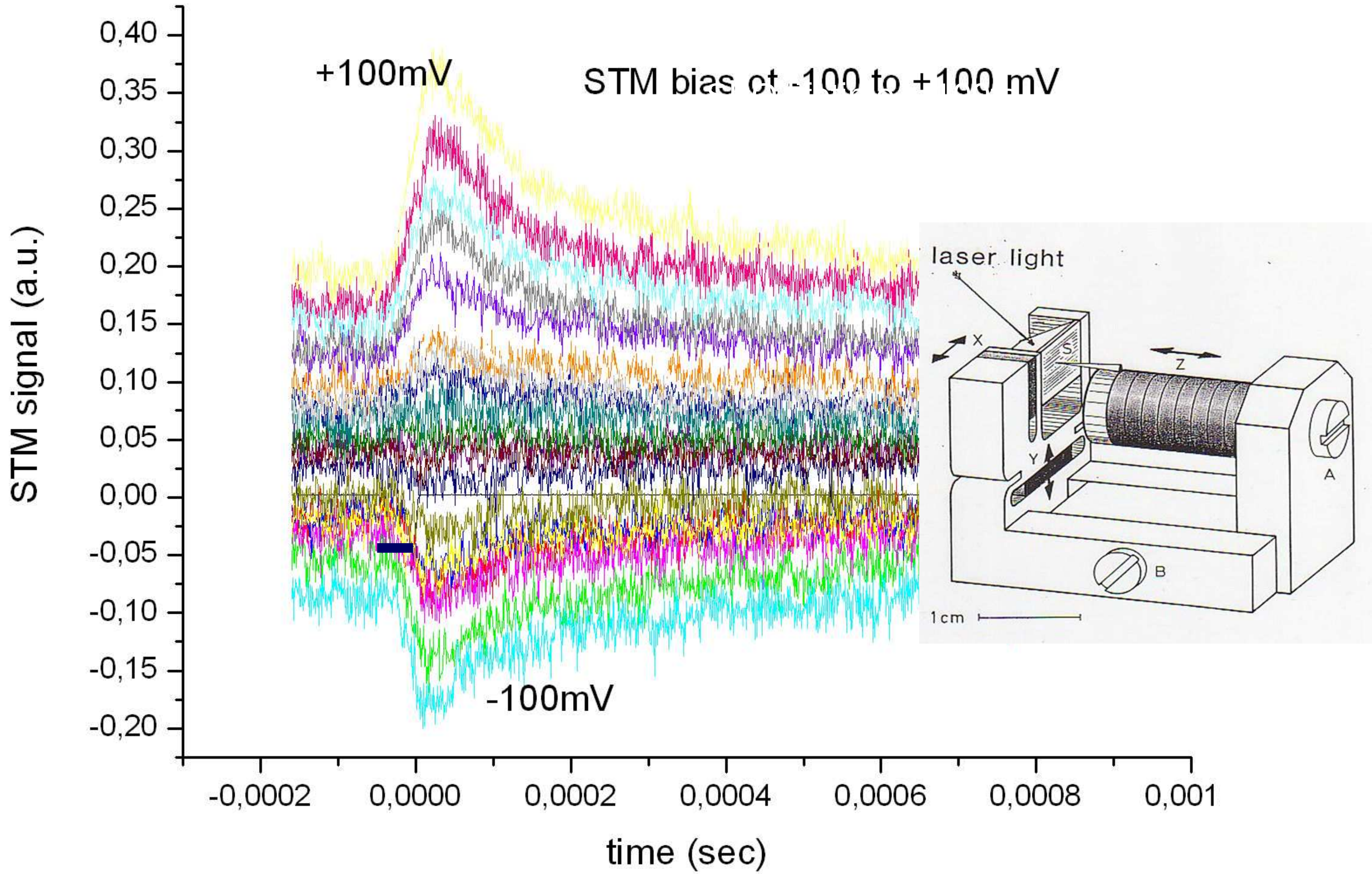
1.5eV = the SPO/photon energy

$$\mathbf{I = I(0)^n}$$

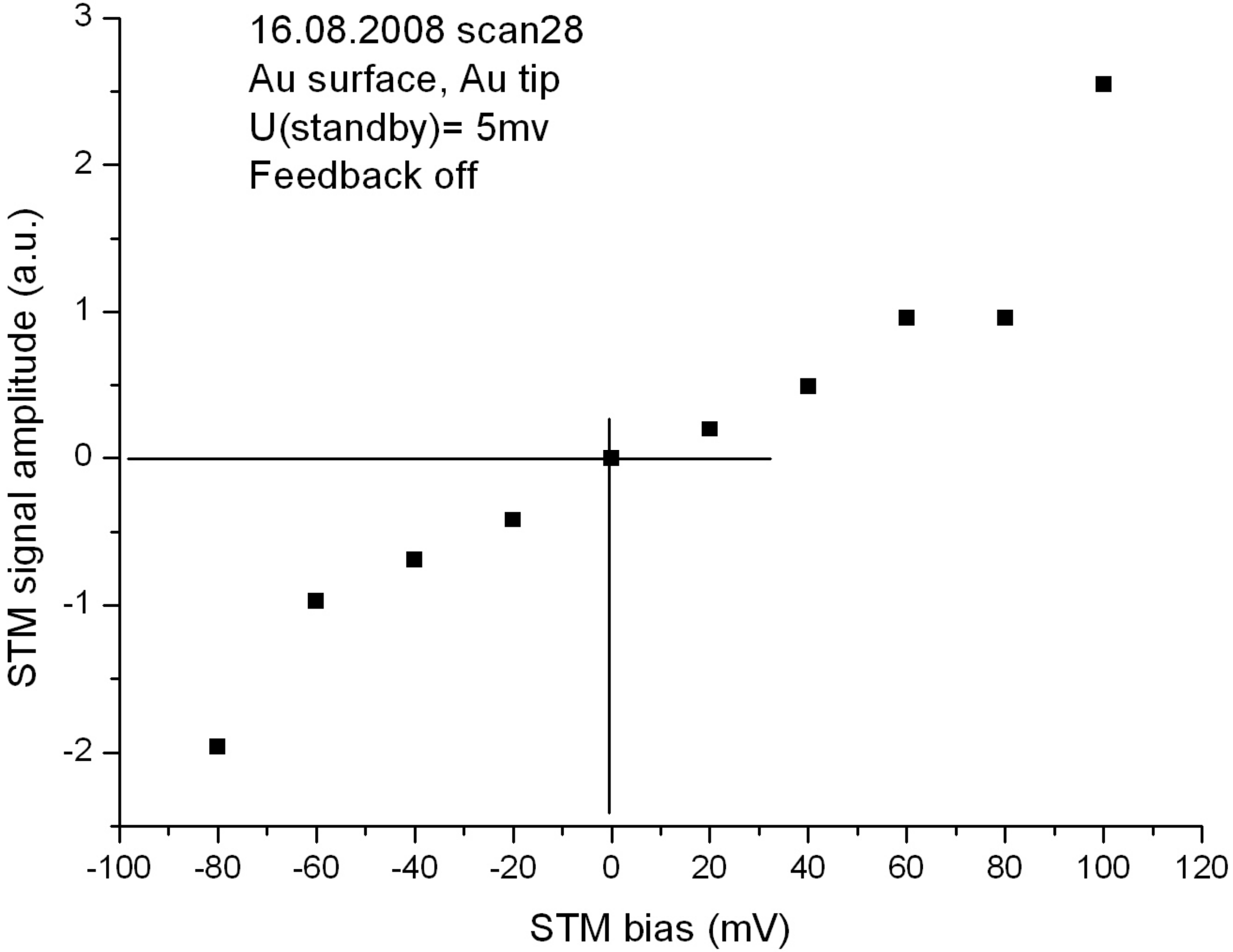
Data from STM measurements (rectification in some surface spots)

Europhys.Lett.15, 289 (1991)

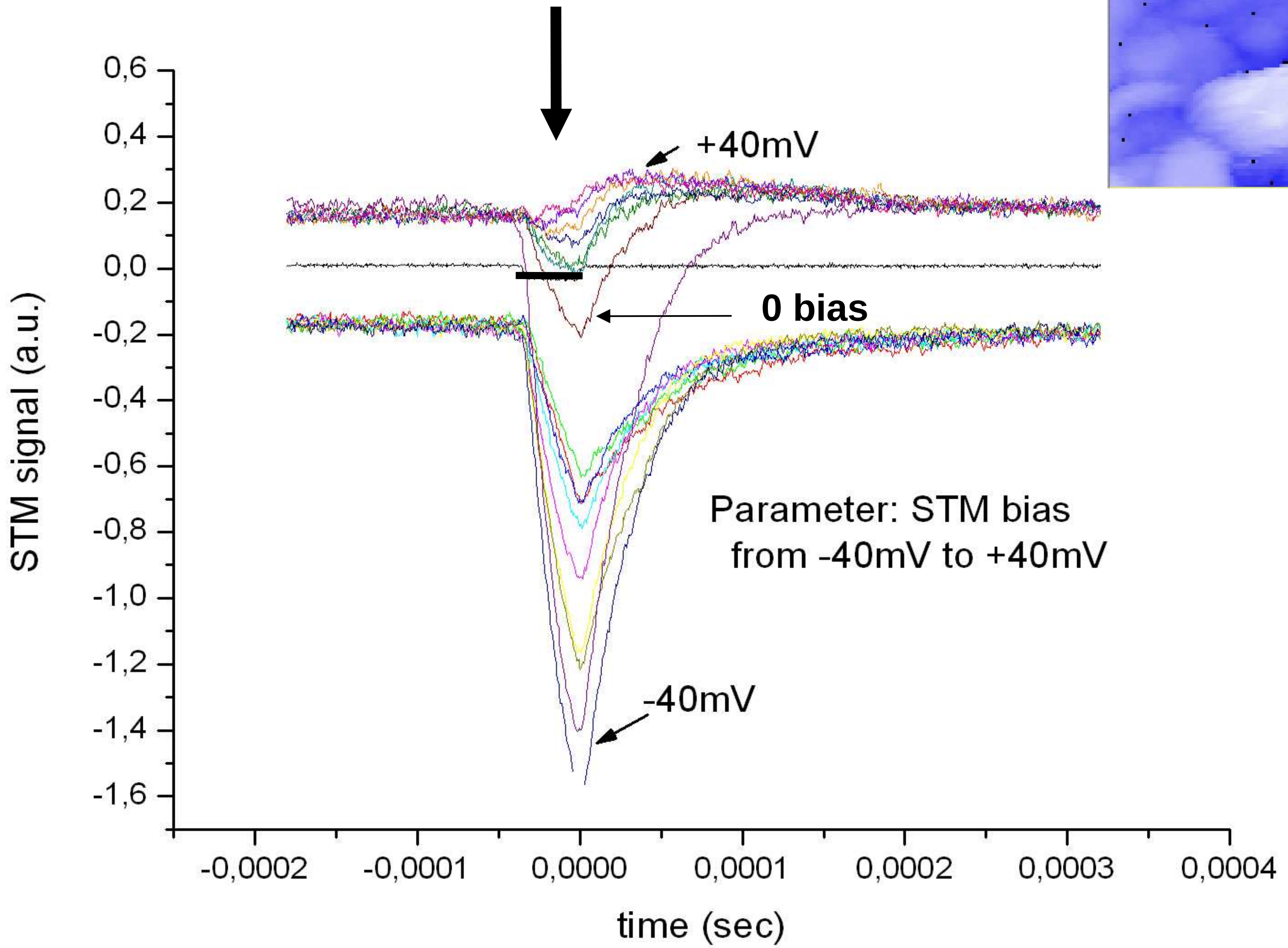


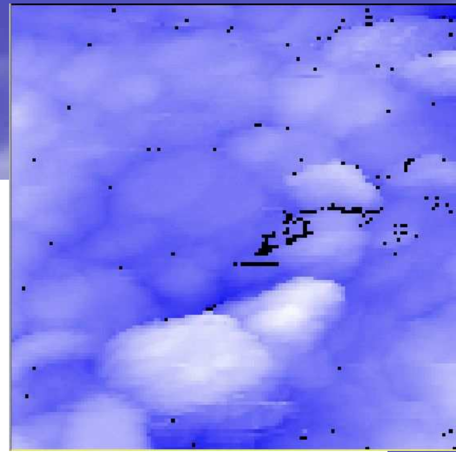


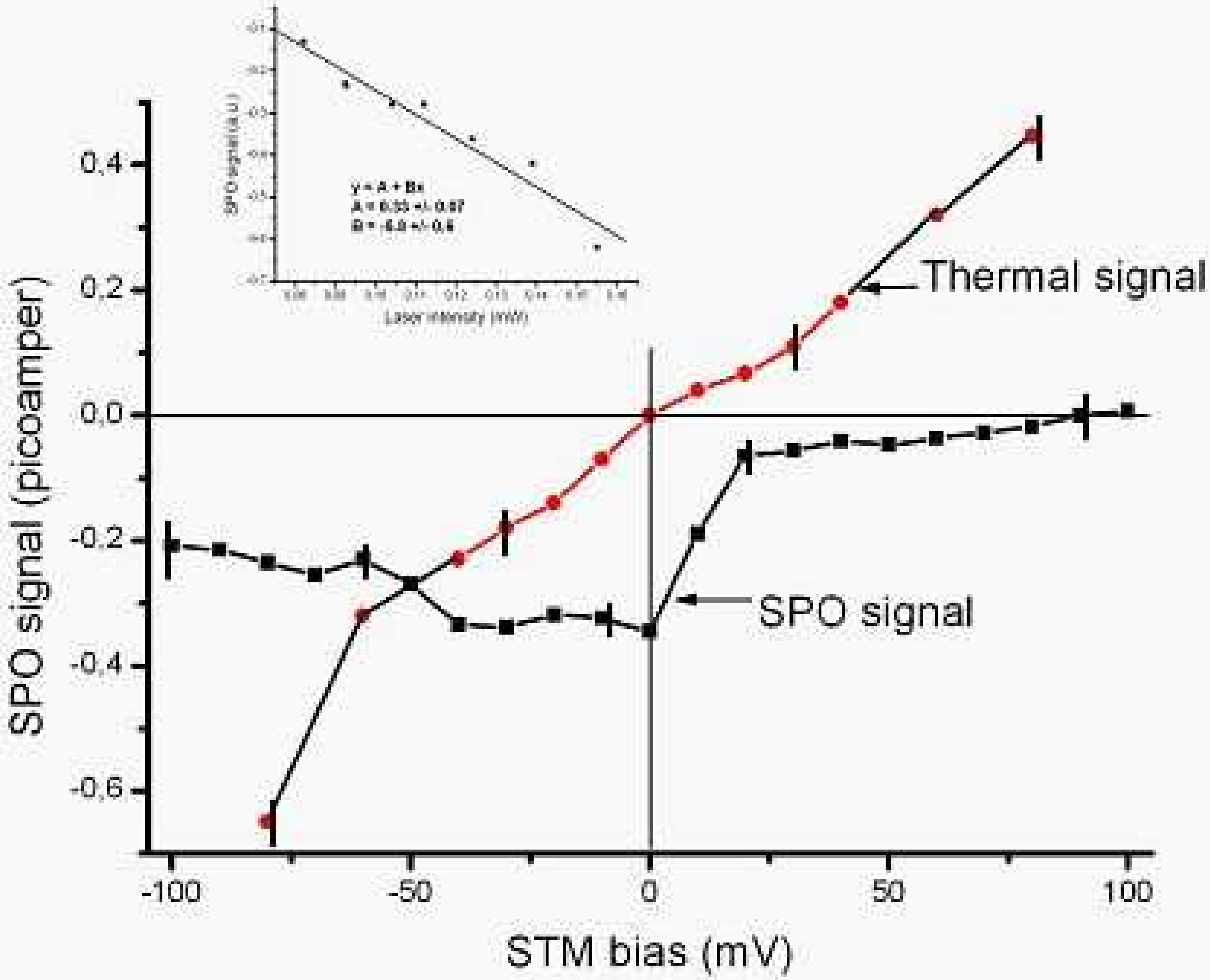
+100mV
STM bias at -100 to +100 mV
-100mV
STM signal (a.u.)
0,40
0,35
0,30
0,25
0,20
0,15
0,10
0,05
0,00
-0,05
-0,10
-0,15
-0,20
-0,0002
0,0000
0,0002
0,0004
0,0006
0,0008
0,001
time (sec)
laser light
X
S
Z
Y
A
B
1cm

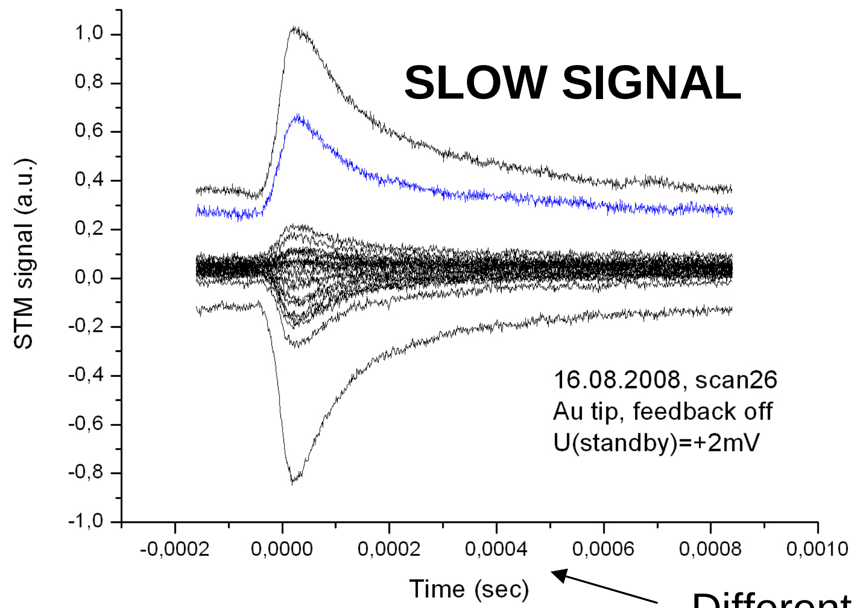

16.08.2008 scan28
Au surface, Au tip
U(standby)= 5mv
Feedback off
STM signal amplitude (a.u.)
STM bias (mV)
3
2
1
0
-1
-2
-100
-80
-60
-40
-20
0
20
40
60
80
100
120

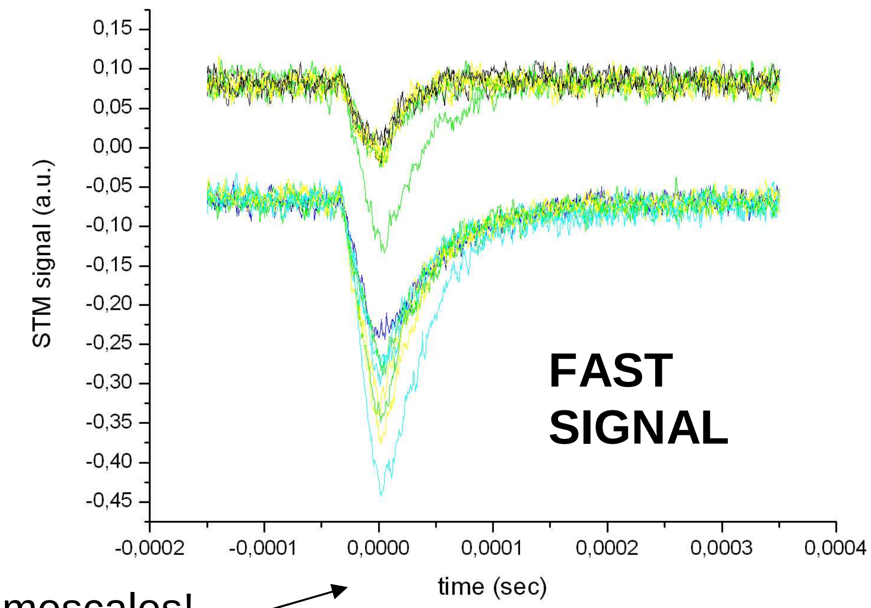
Phys Rev B 83, 205428 (2011)



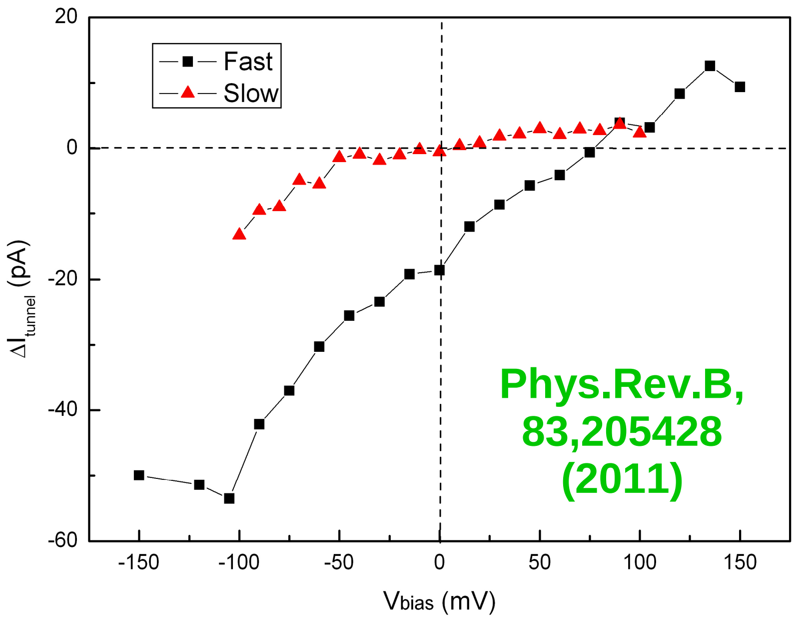


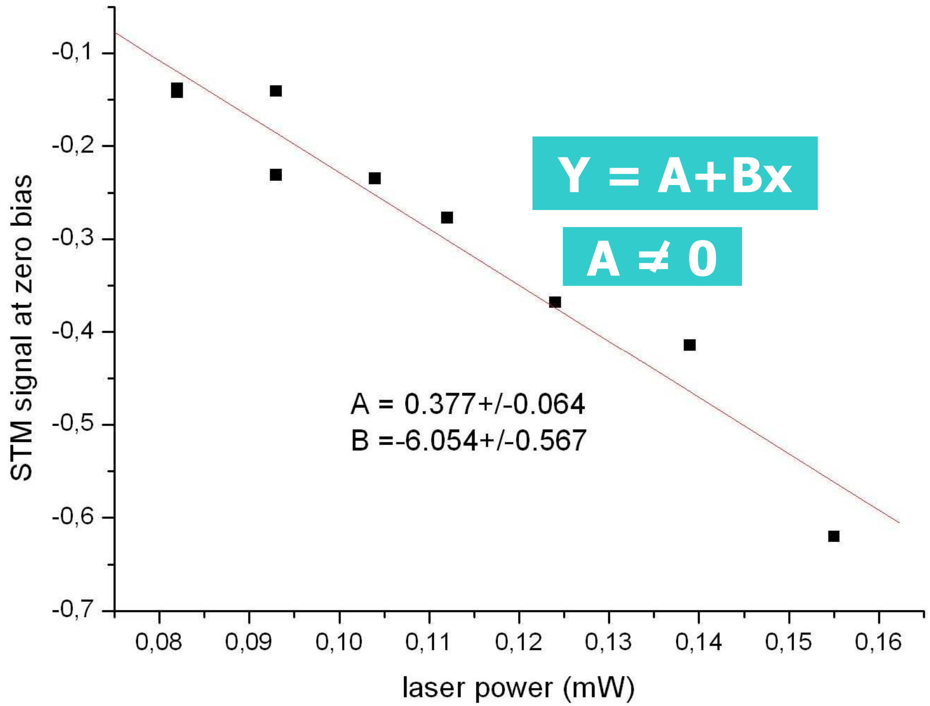

y = A + Bx
Thermal signal
SPO signal
SPO signal (picoamper)
0,4
0,2
0,0
-0,2
-0,4
-0,6
-100
-50
0
50
100
STM bias (mV)





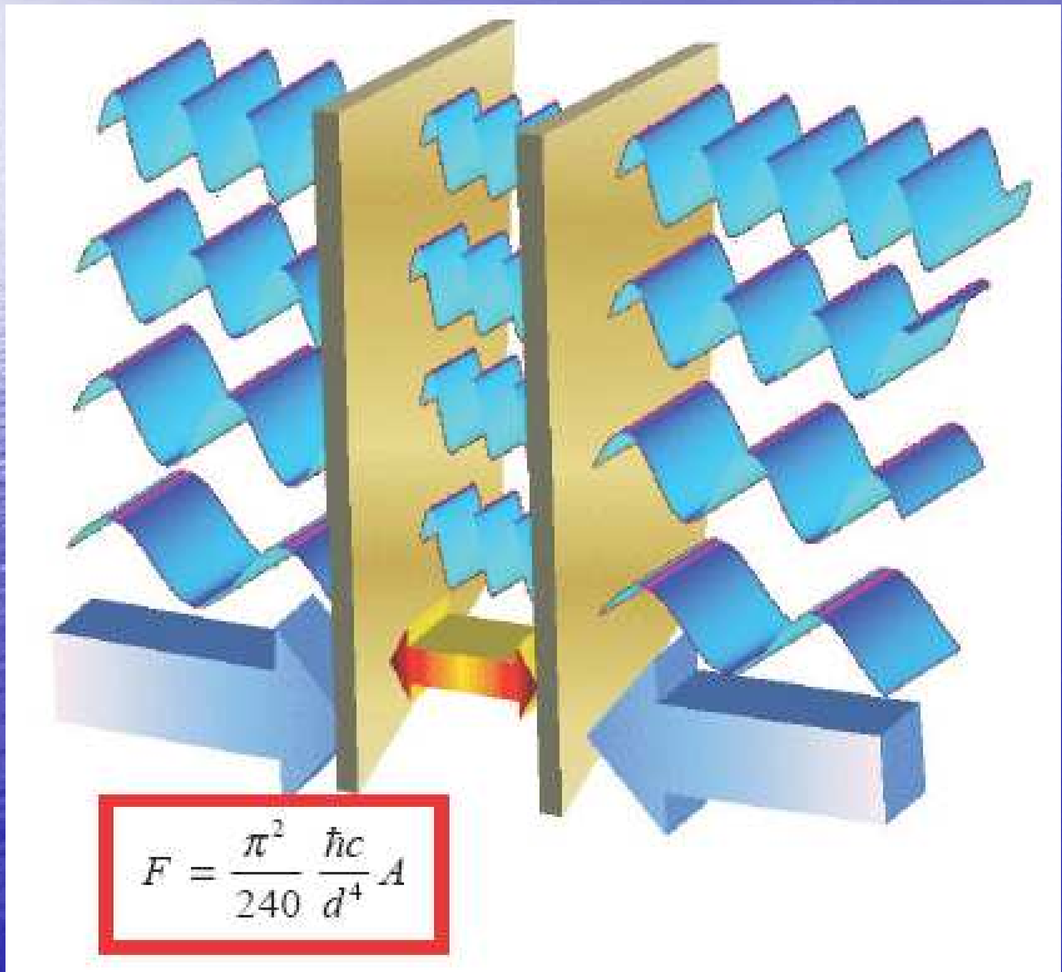
Different timescales!





Casimir force

- When the distance of the plates 10 nm -1 atm attractive pressure

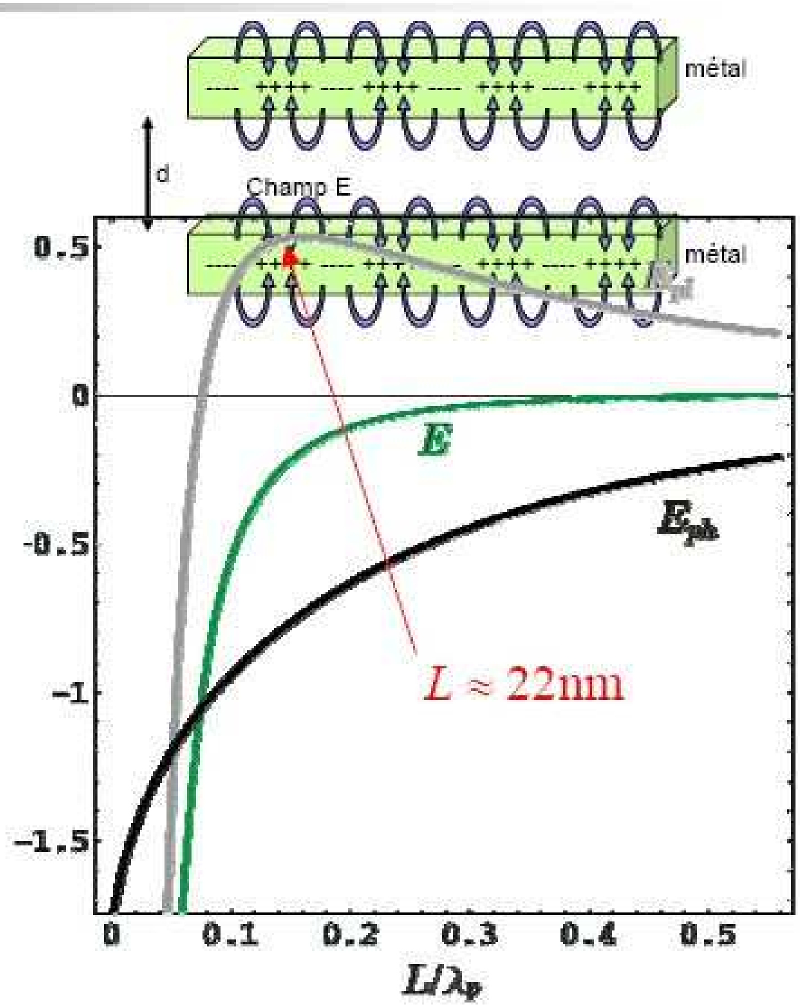


WHY MAY THIS BE IMPORTANT IN NANOTECHNOLOGY?

Surface plasmons

- For L<<10nm :
  Casimir effect = interaction between surface plasmons living on each mirror
- "Plasmonic modes" :
  evanescent waves
- "Photonic" or "cavity modes"
  propagating waves

- For L>20nm :
  Surface plasmon contribution to Casimir force is repulsive



F. Intravaia, & A. Lambrecht, Phys. Rev. Lett. , 94, 110404 (2005)

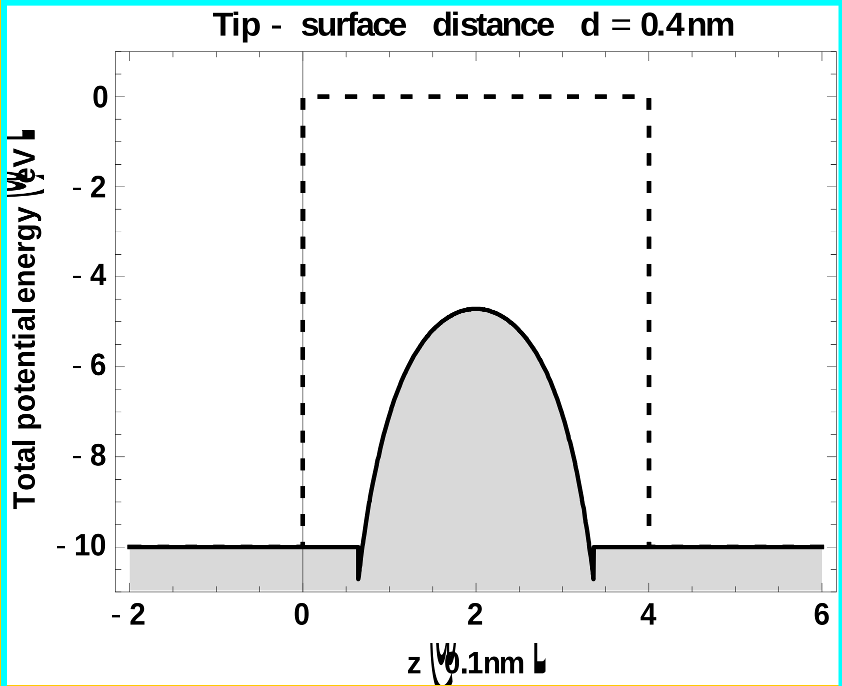
THE EFFECT OF MULTIPLE IMAGE CHARGE ON TUNNELING[a]

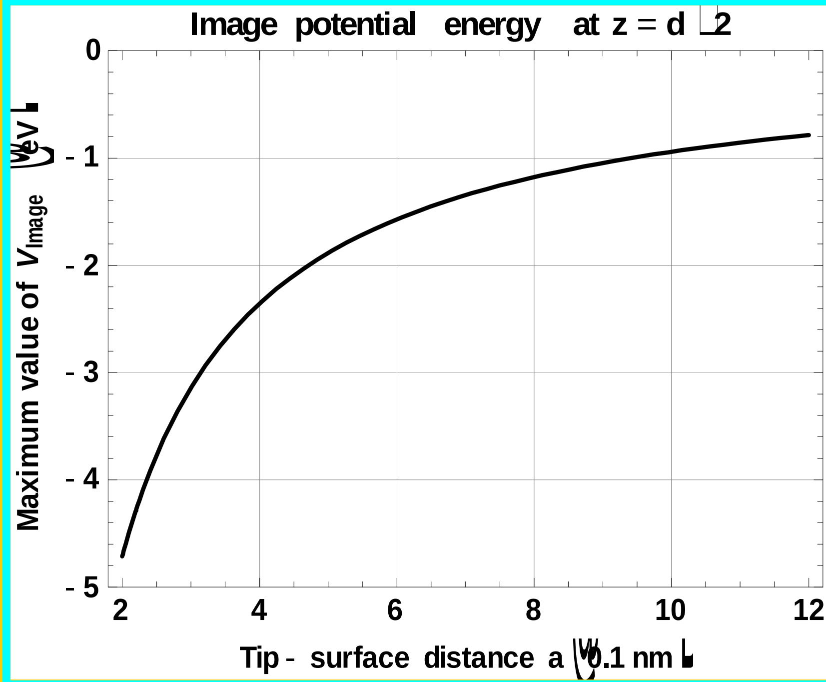
$$V_{\text{Im}}(z) = -\frac{e^2}{2d}\left[\psi(1) - \frac{1}{2}\psi(z/d) - \frac{1}{2}\psi(1-z/d)\right]$$

$$\psi(x) = \frac{1}{\Gamma(x)}\frac{d\Gamma(x)}{dx}$$

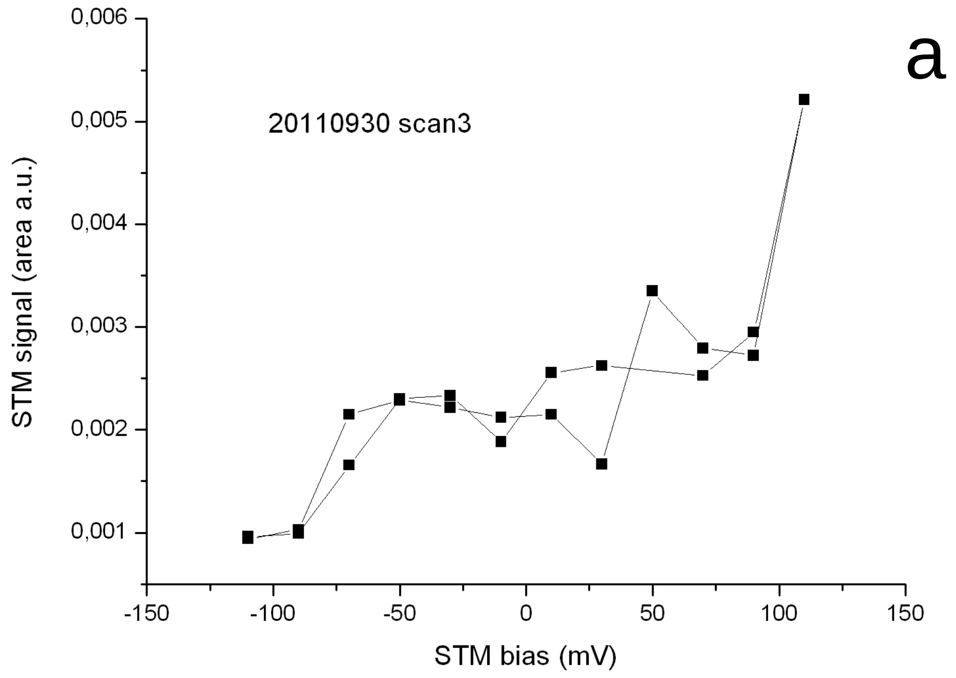
Even at zero bias, the multiple image charge may result in a considerable reduction of the barrier between the anod and cathode. This is described by the digamma function ψ (*x*).

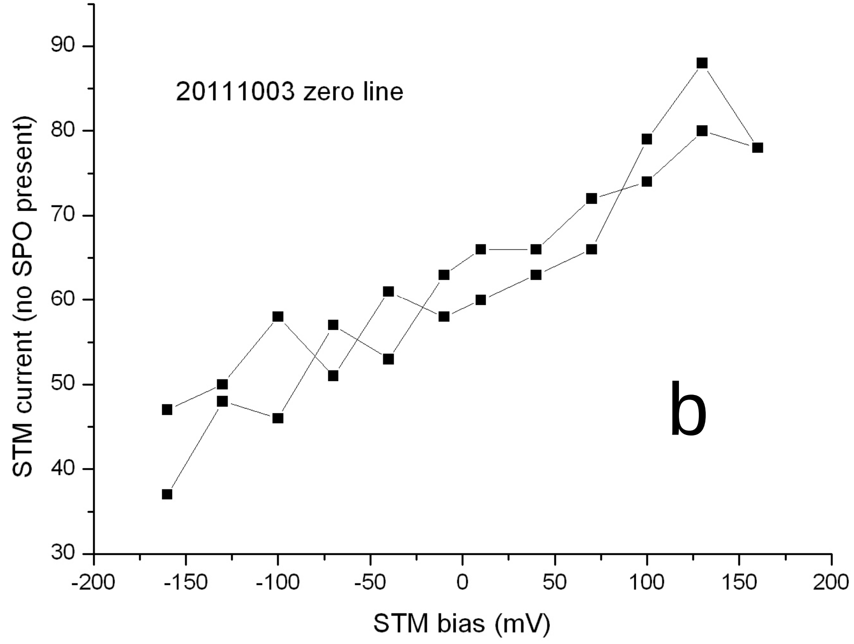
$$-V_{\text{Im}}^{\max} = -V_{\text{Im}}(d/2) = \frac{e^2}{d}\ln 2 = \frac{9.427}{[d/0.1nm]}eV$$





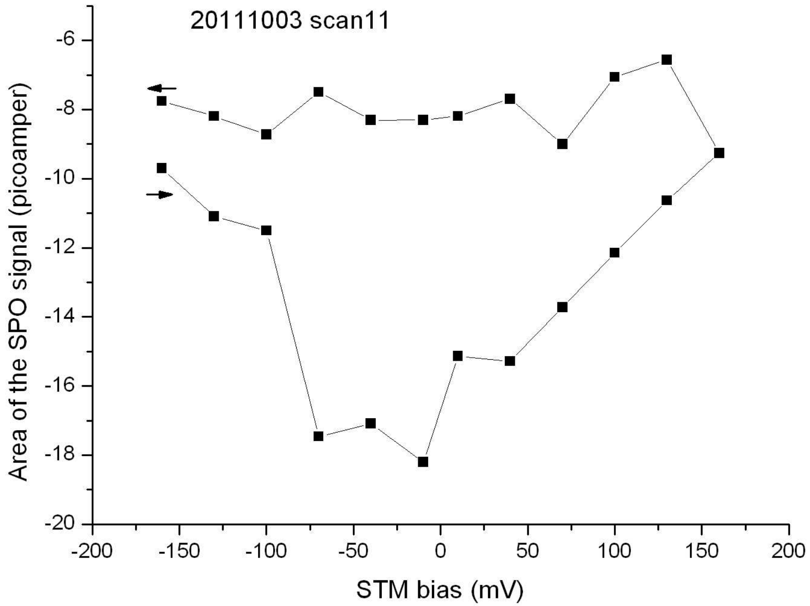
To decide which model might be valid?

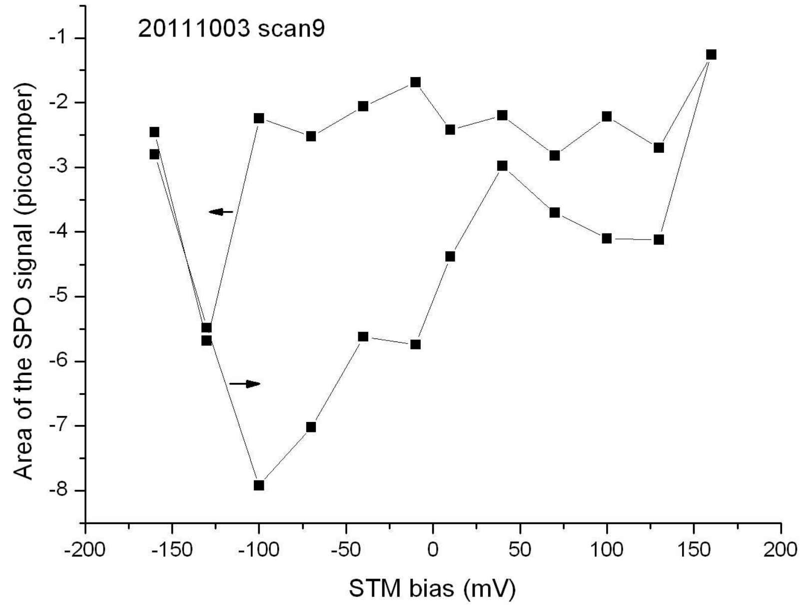




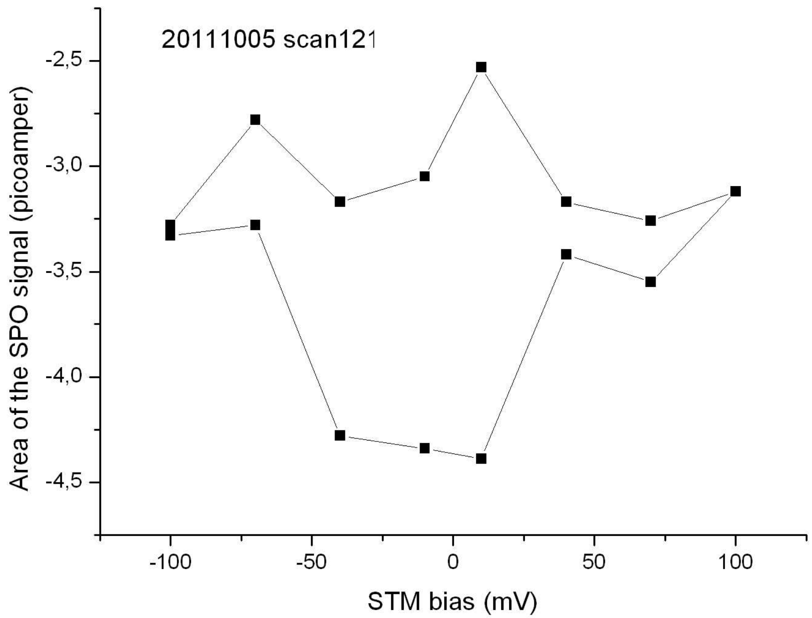
HYSTERESIS NOT OBSERVED IN THE THERMAL SIGNAL (a) AND IN THE TUNNEL CURRENT WITHOUT SPO EXCITATION (b)

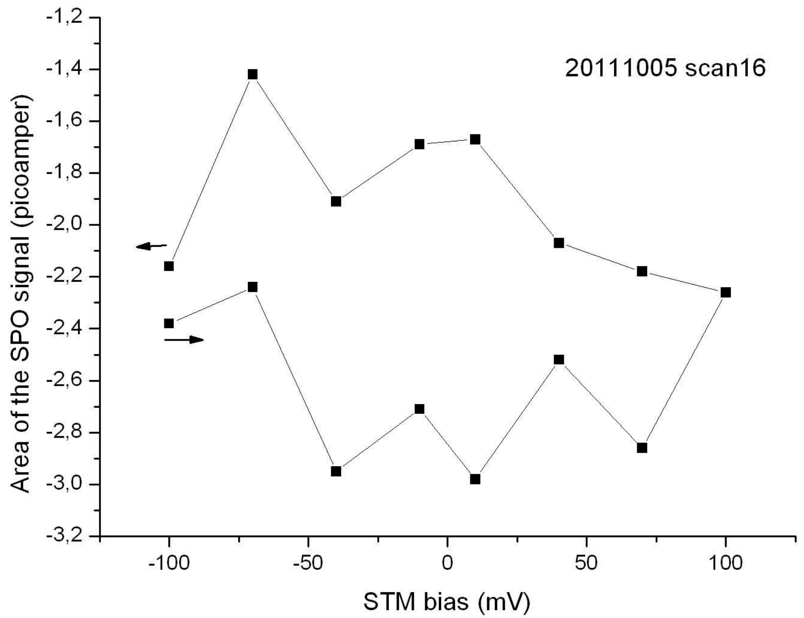
HYSTERESIS!





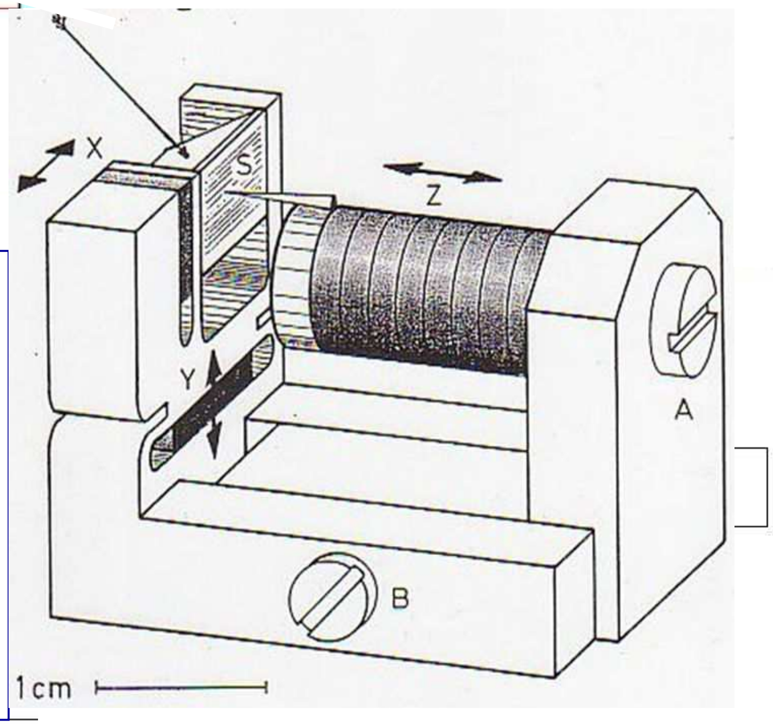
CASIMIR-EFFECT EXCLUDED!





ARRANGEMENT FOR FEMTOSECOND LASER EXCITED SPO-STM MEASUREMENTS

790nm Ti:Sa laser;
3.6MHz⇒1.2kHz,
110-120fs pulsees



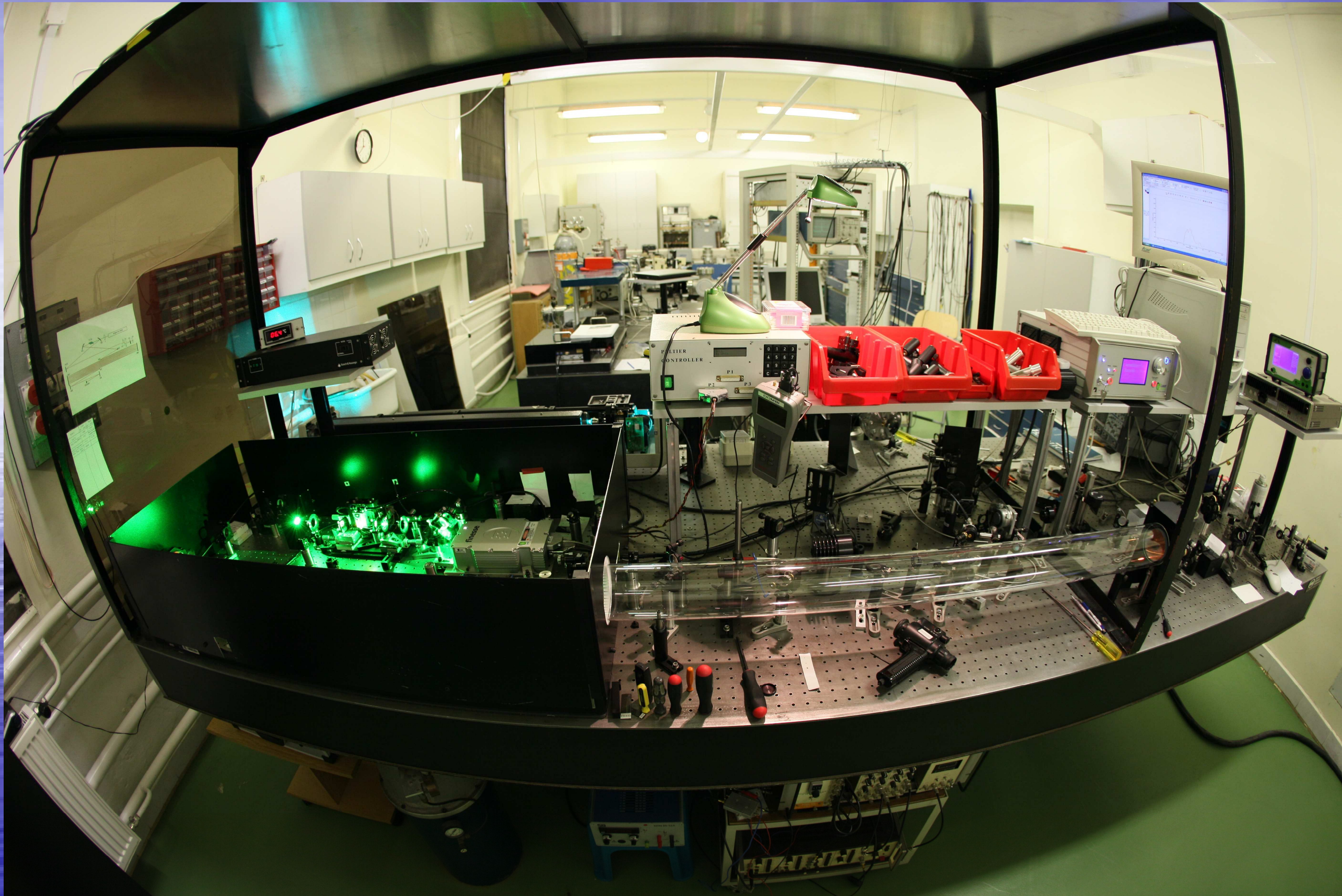
The long resonator (~80m)

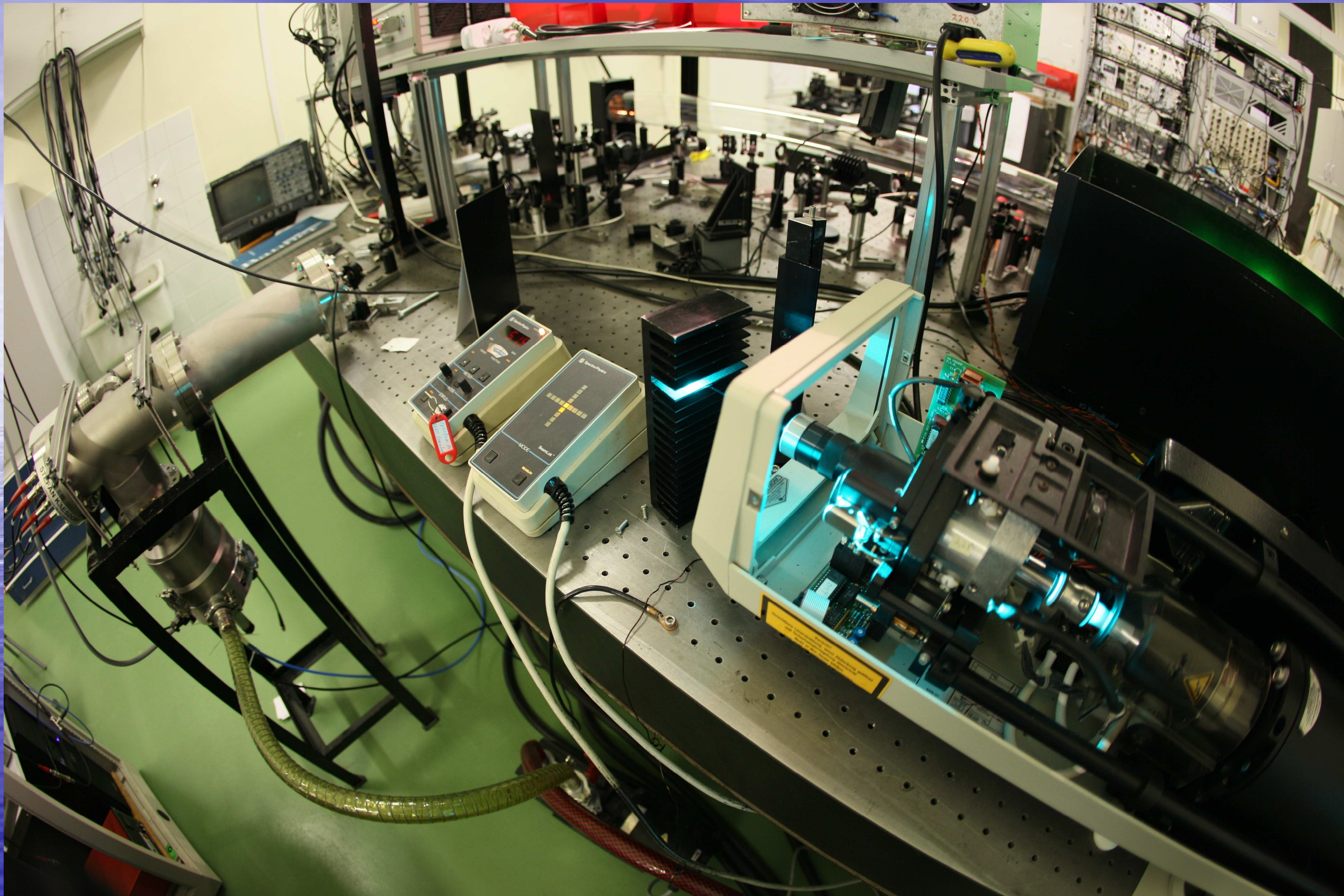
Nd:YAG laser pumped Ti:Sa laser

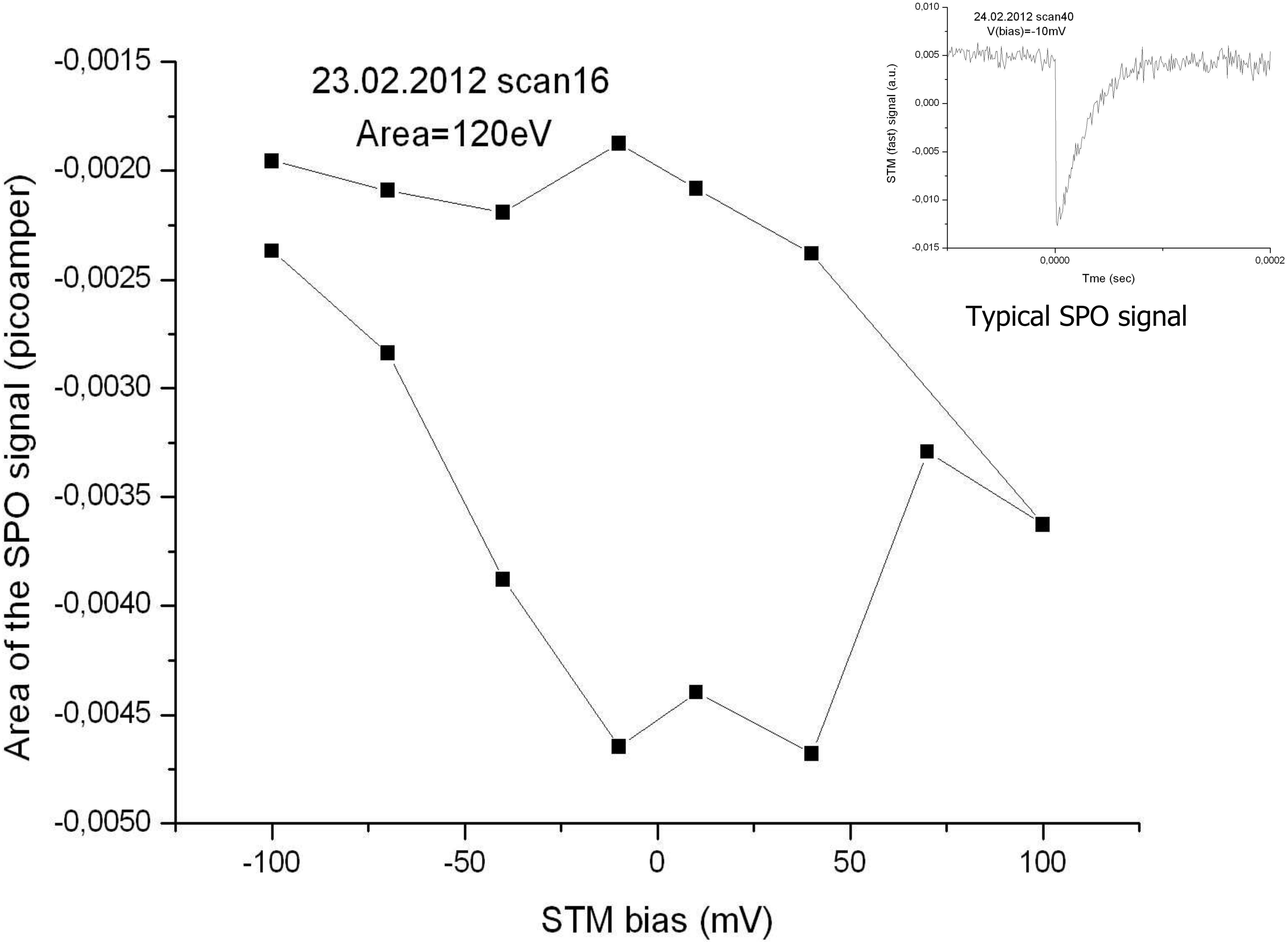
produced a 3.6MHz pulse train,

reduced to 1.6kHz, coupled into the

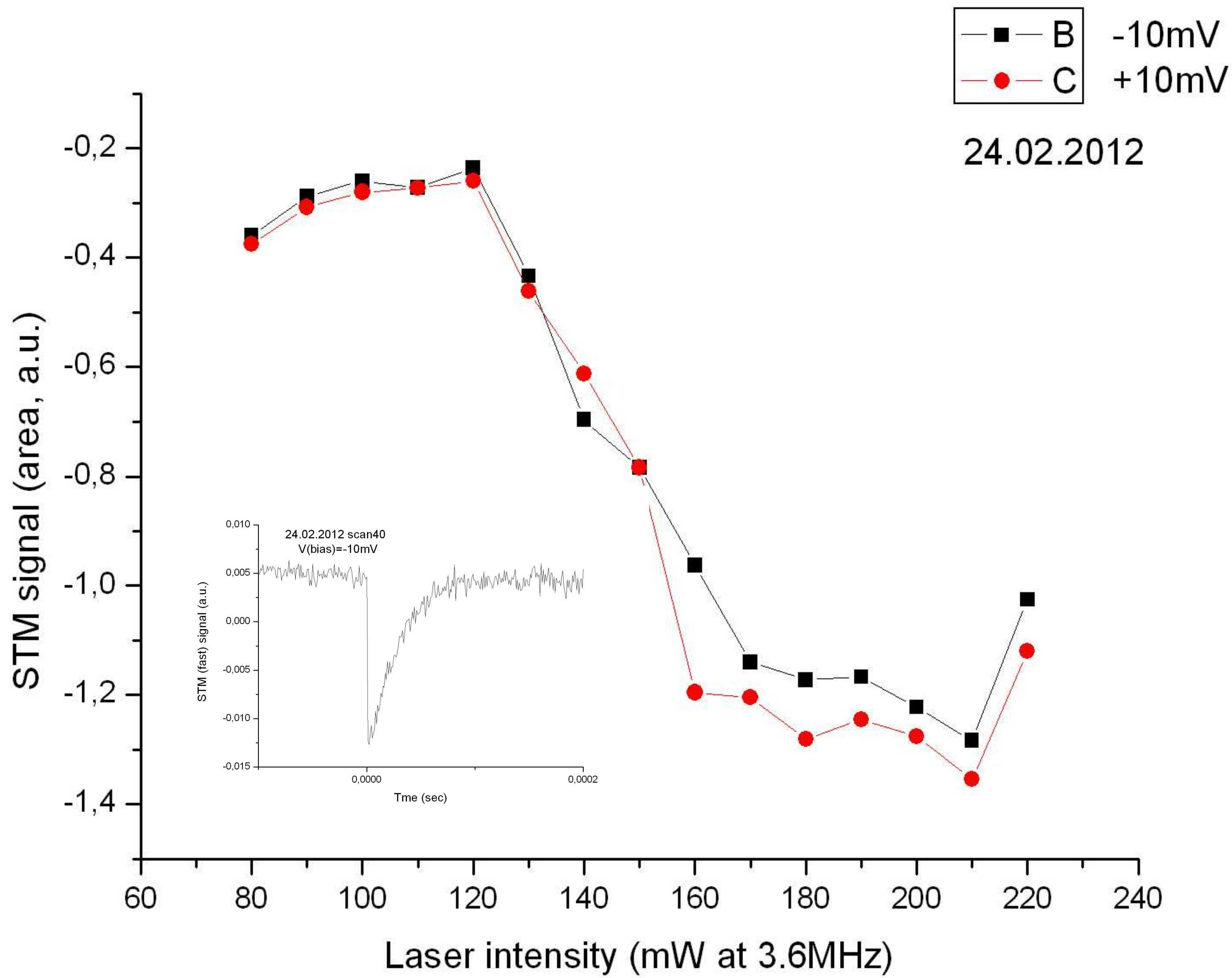
STM in the Kretschmann geometry



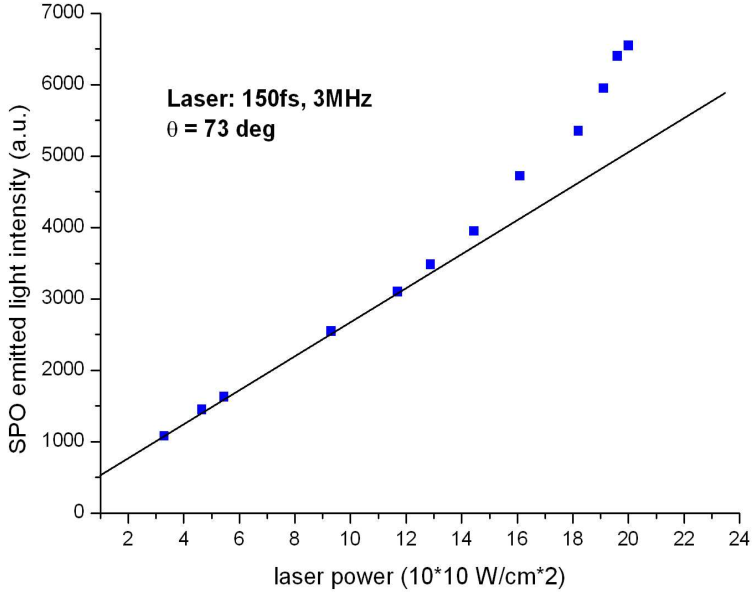


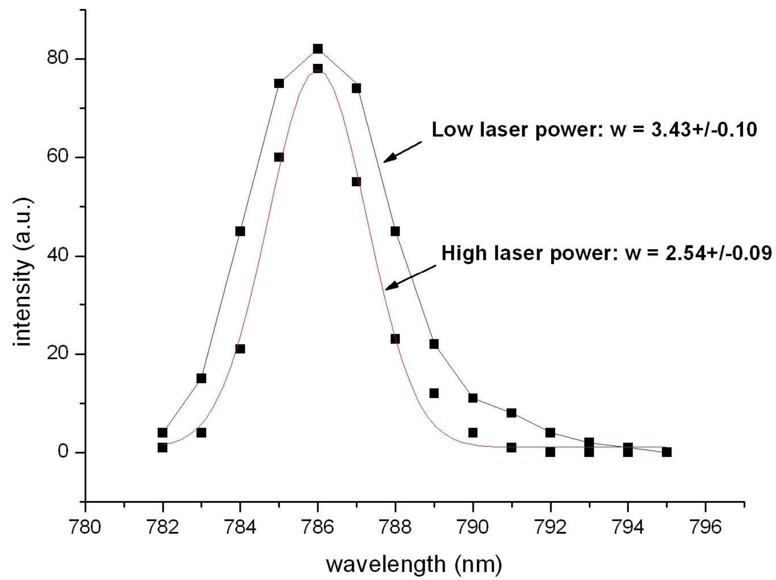


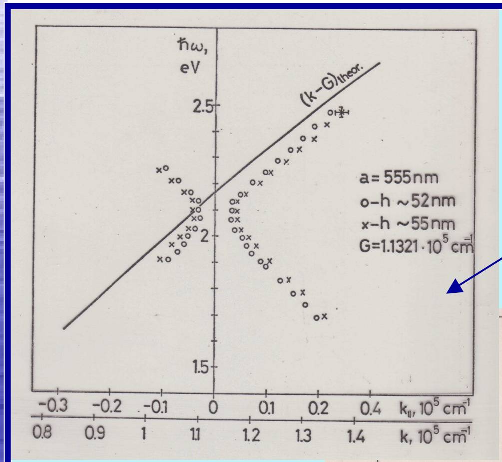
23.02.2012 scan16
Area=120eV
Area of the SPO signal (picoamper)
-0,0015
-0,0020
-0,0025
-0,0030
-0,0035
-0,0040
-0,0045
-0,0050
-100
-50
0
50
100
STM bias (mV)
24.02.2012 scan40
V(bias)=-10mV
STM (fast) signal (a.u.)
0,010
0,005
0,000
-0,005
-0,010
-0,015
0,0000
0,0002
Tme (sec)
Typical SPO signal



B -10mV
C +10mV
24.02.2012
STM signal (area, a.u.)
-0,2
-0,4
-0,6
-0,8
-1,0
-1,2
-1,4
60
80
100
120
140
160
180
200
220
240
Laser intensity (mW at 3.6MHz)
24.02.2012 scan40
V(bias)=-10mV
STM (fast) signal (a.u.)
0,010
0,005
0,000
-0,005
-0,010
-0,015
0,0000
0,0002
Tme (sec)



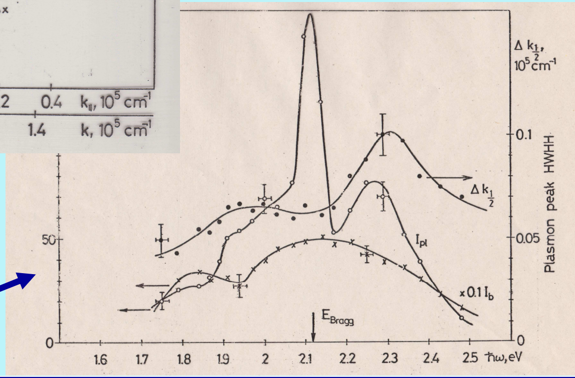




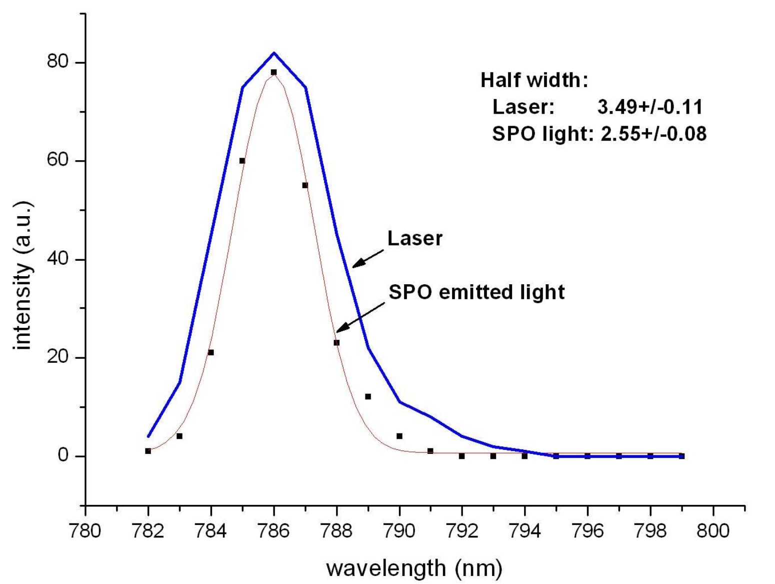
$Ag\text{-}Al_2O_3\text{-}Al$ diode

Electron excitation

Momentum gap
Phys Lett
86A,445(1981)



Emitted light intensity

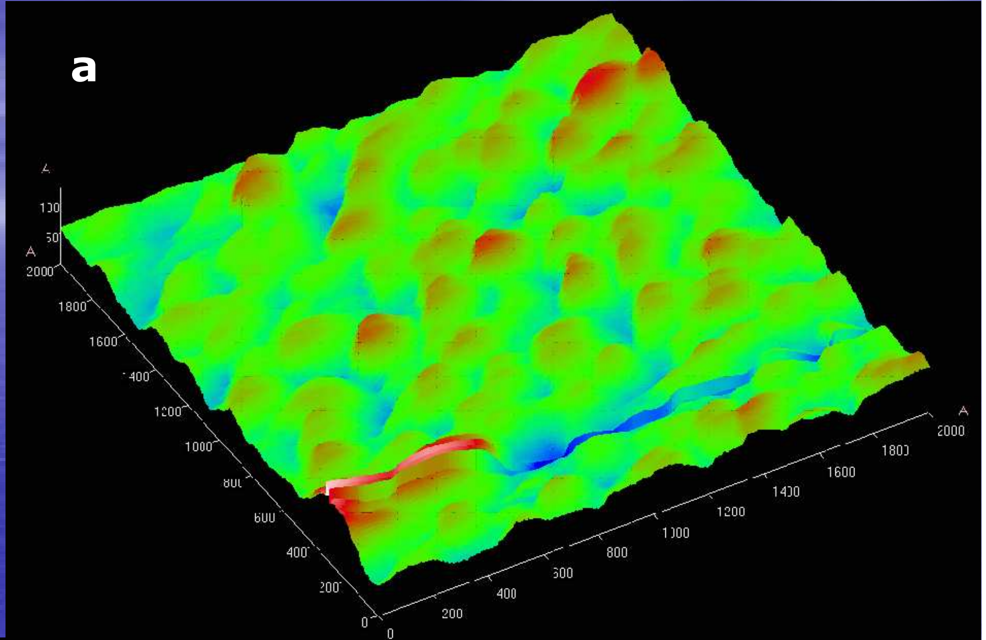


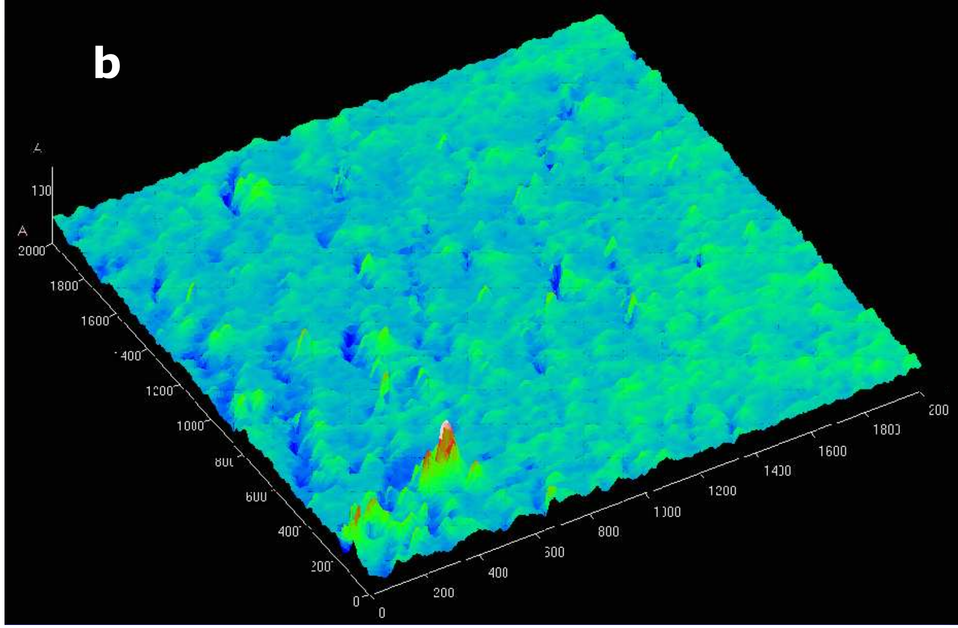
Au(Ag) surface, Au tip, 200nm

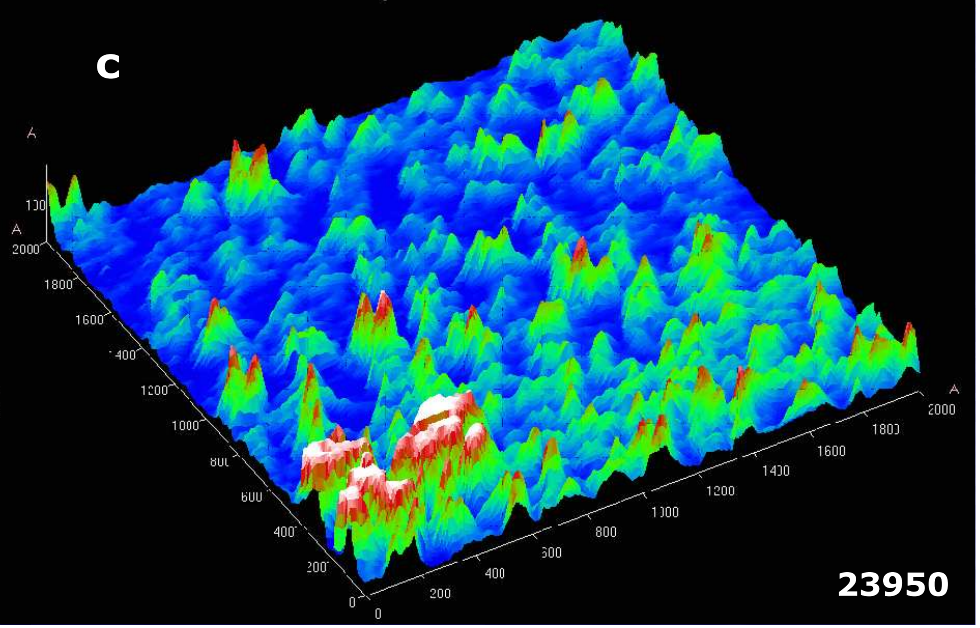
a: topography

b: SPO image

c: thermal image

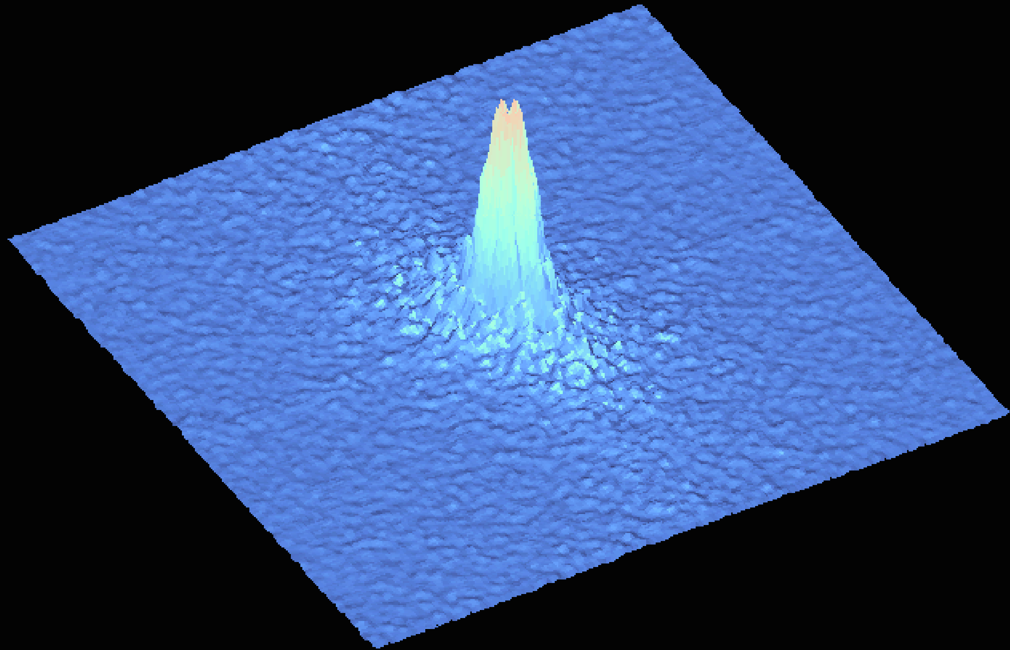


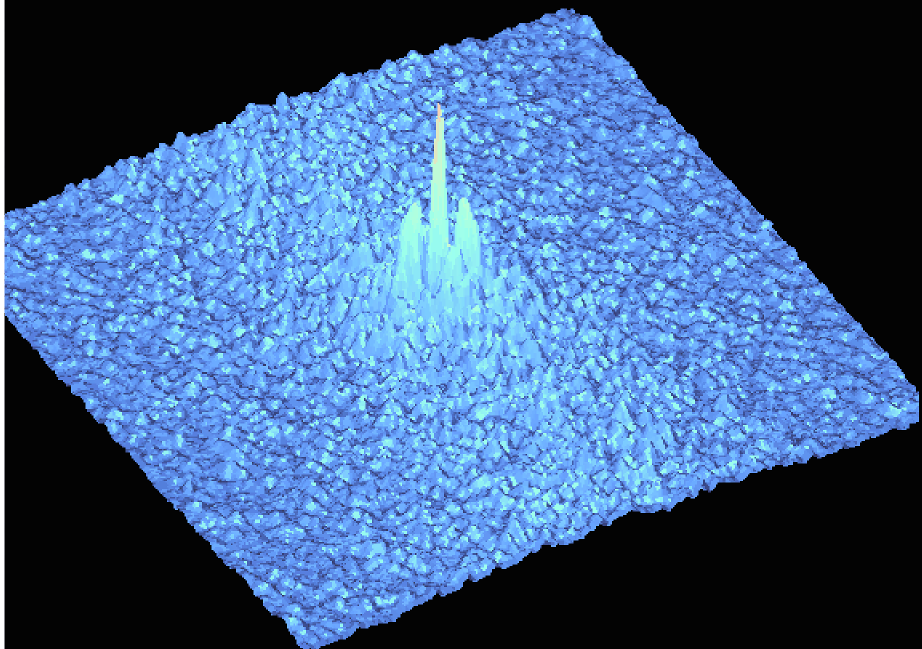


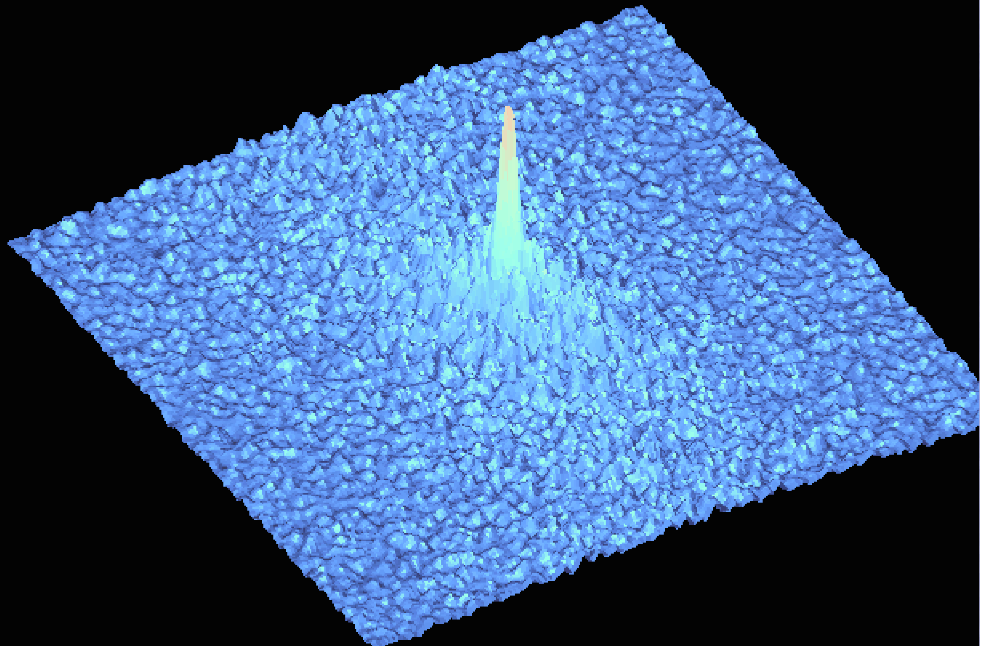


23950

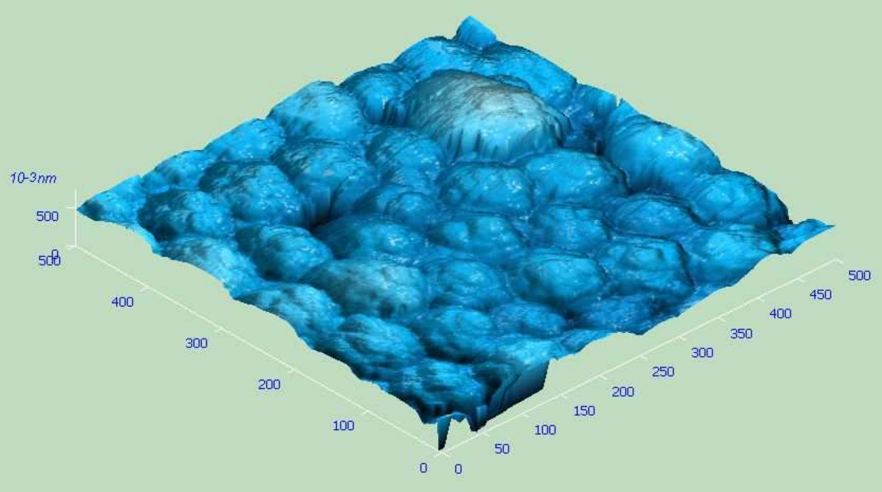
FFT OF THE TOPOGRAPHIC SURFACE PLASMON AND THERMAL IMAGES



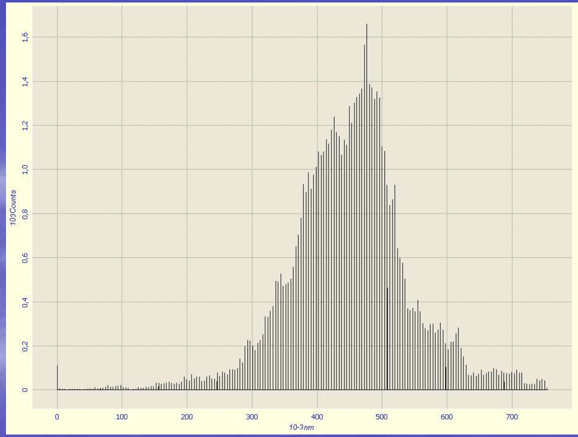


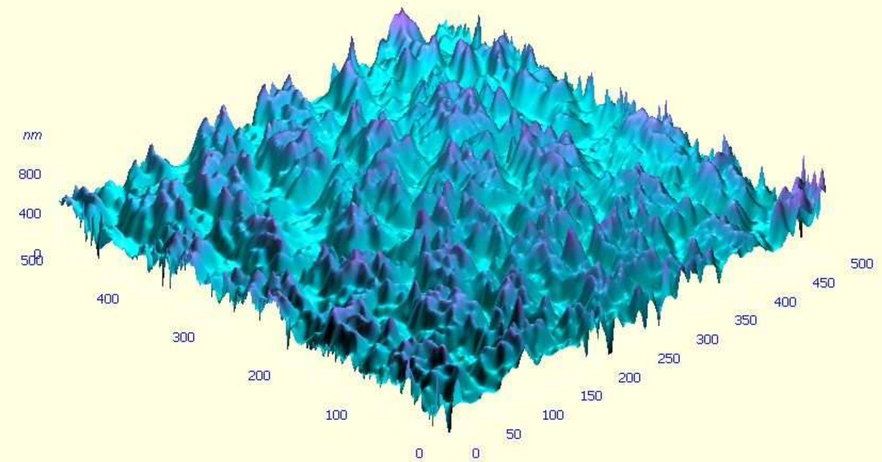


23950ss

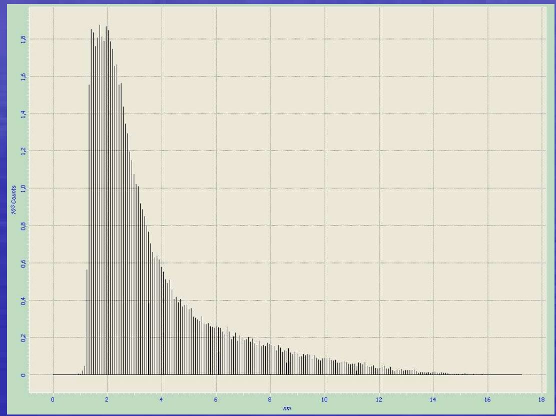


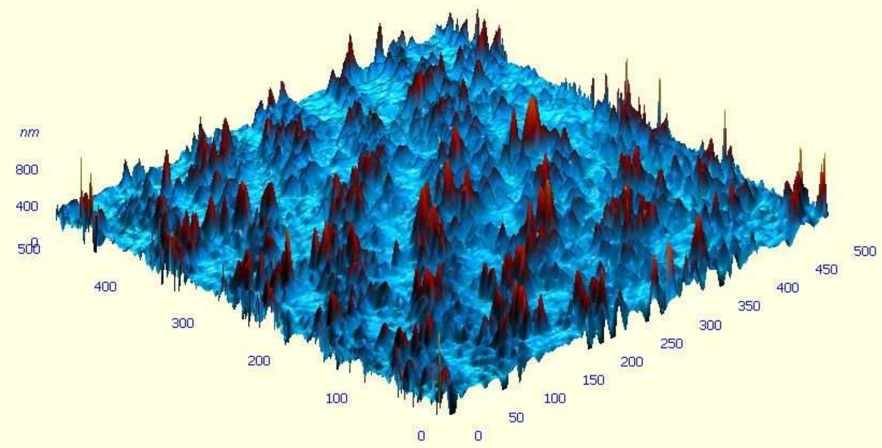
topography



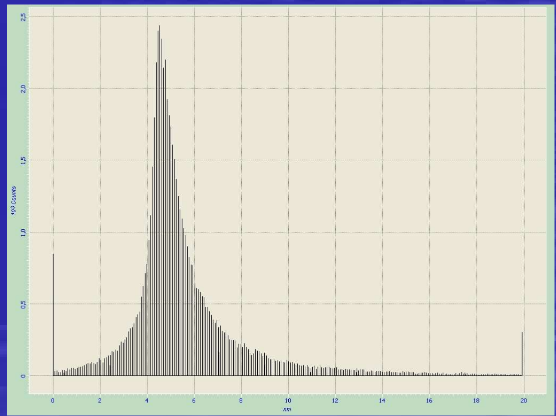


thermal

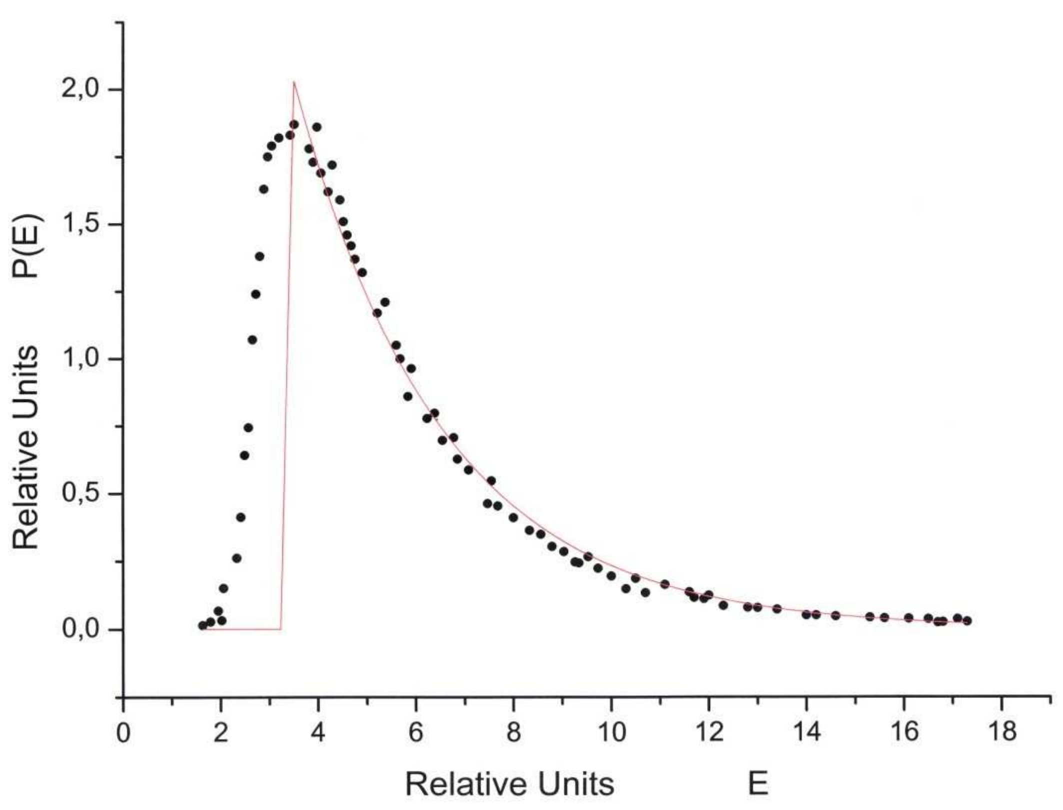


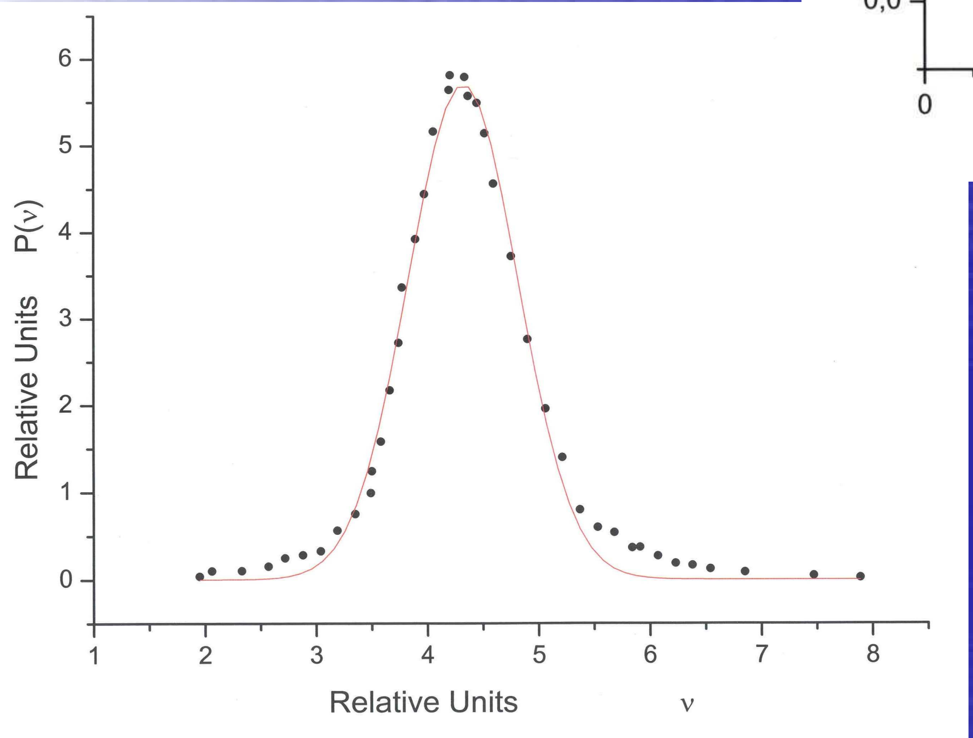


SPO



SURFACE ANALYSIS



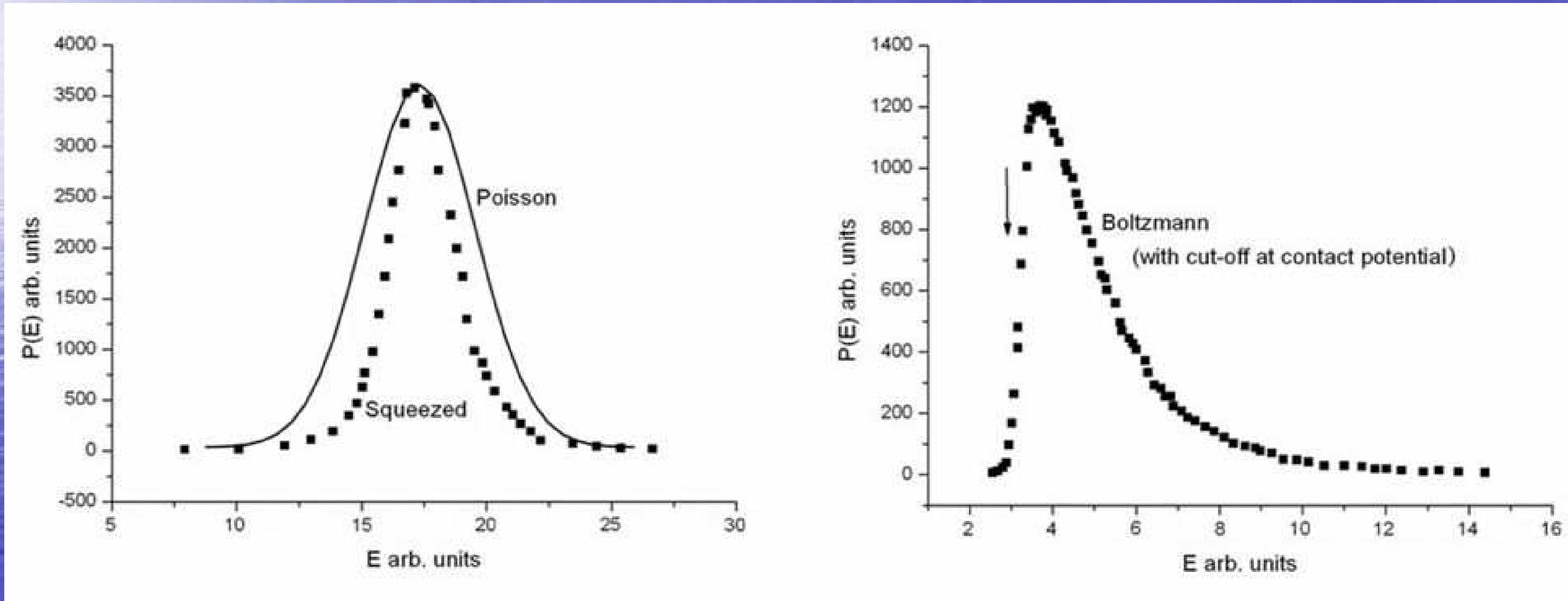


Surface statistics of the thermal image. Boltzmann distribution with a cut-off

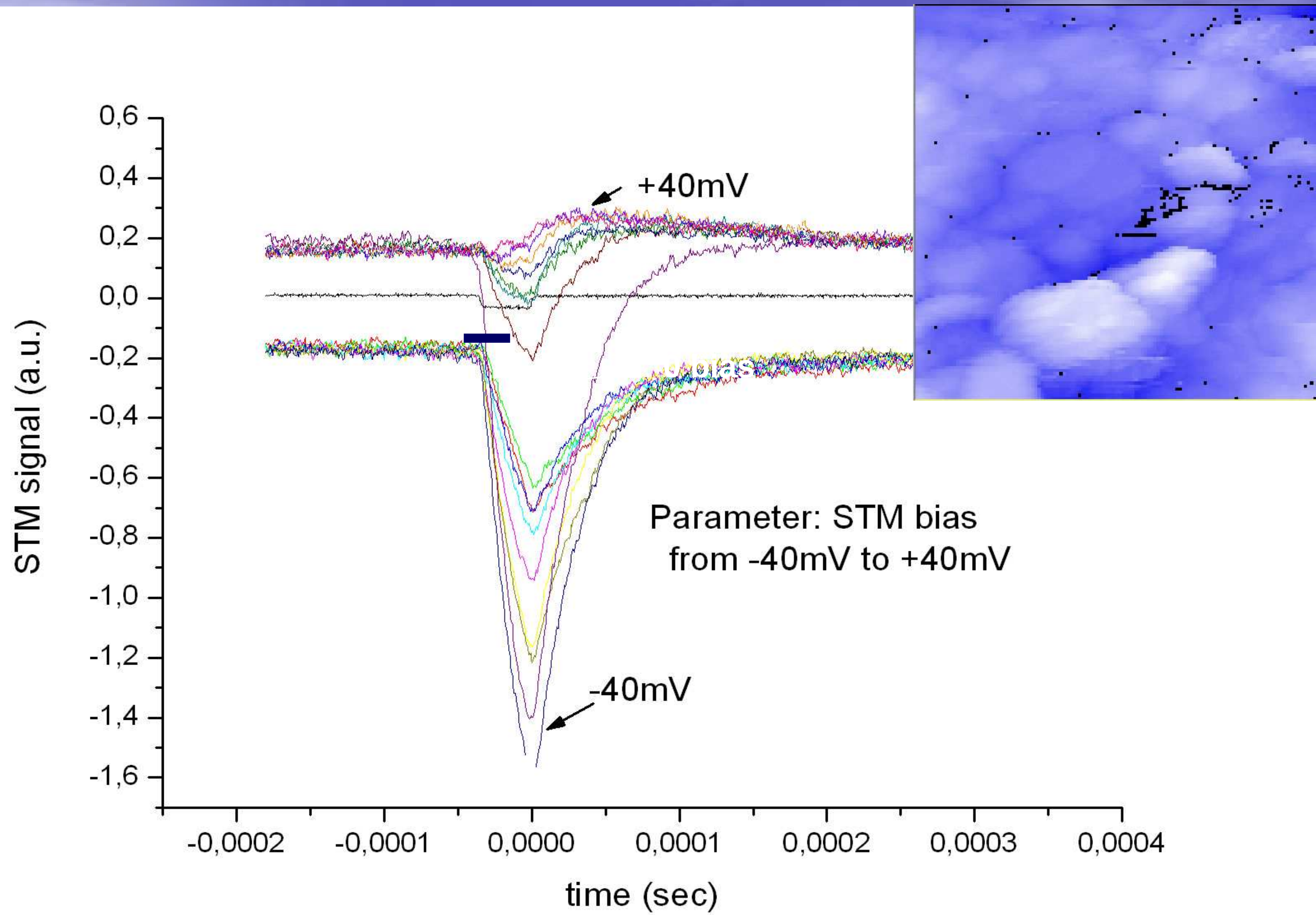
Surface statistics of the SPO image. Gaussian (sub-Poissonian) distribution

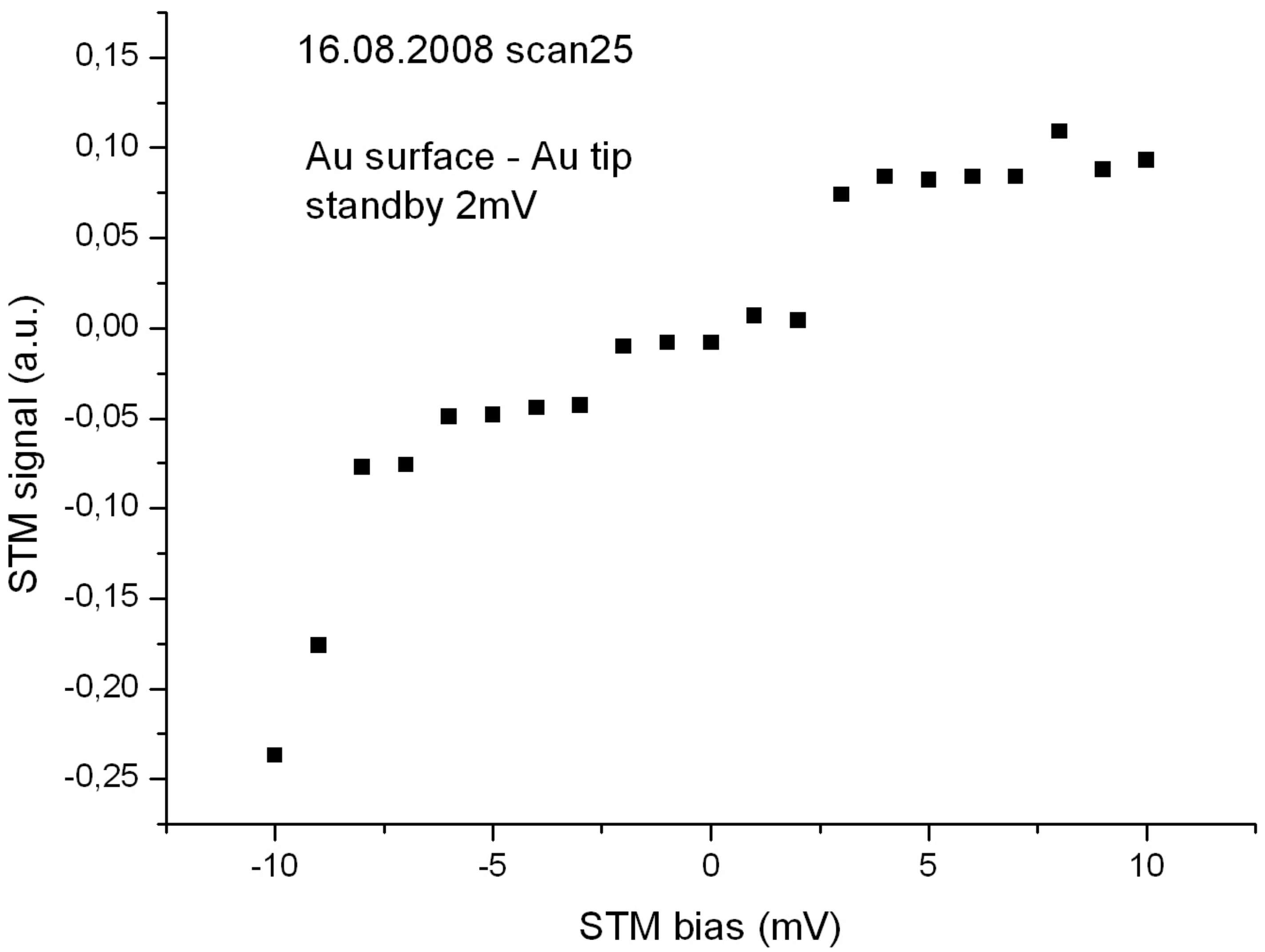
23952-3

SQUEEZING IN THE SPO IMAGE



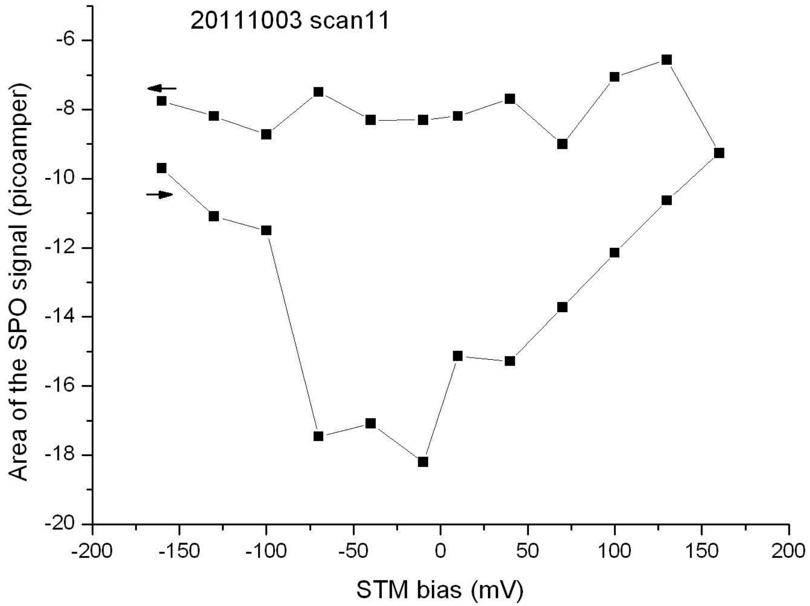
„NEGATIVE" STM SIGNALS IN SOME (HOT) SPOTS

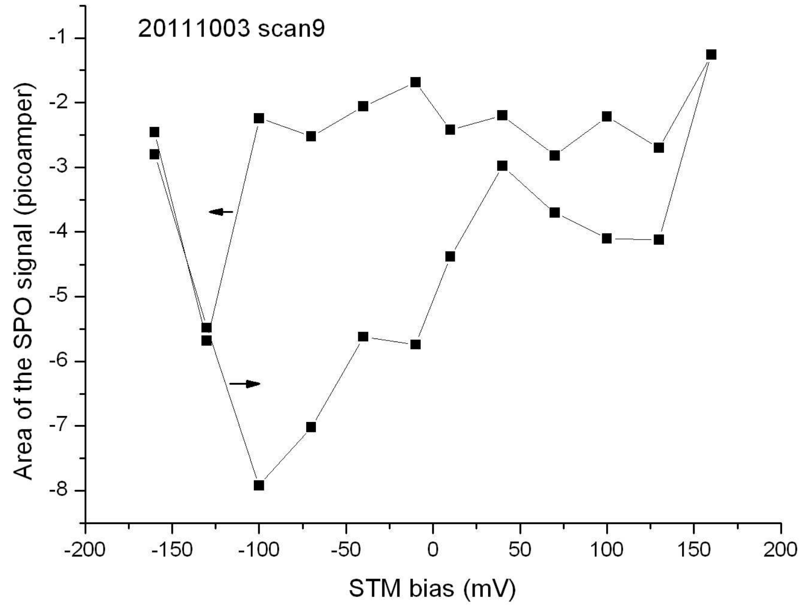




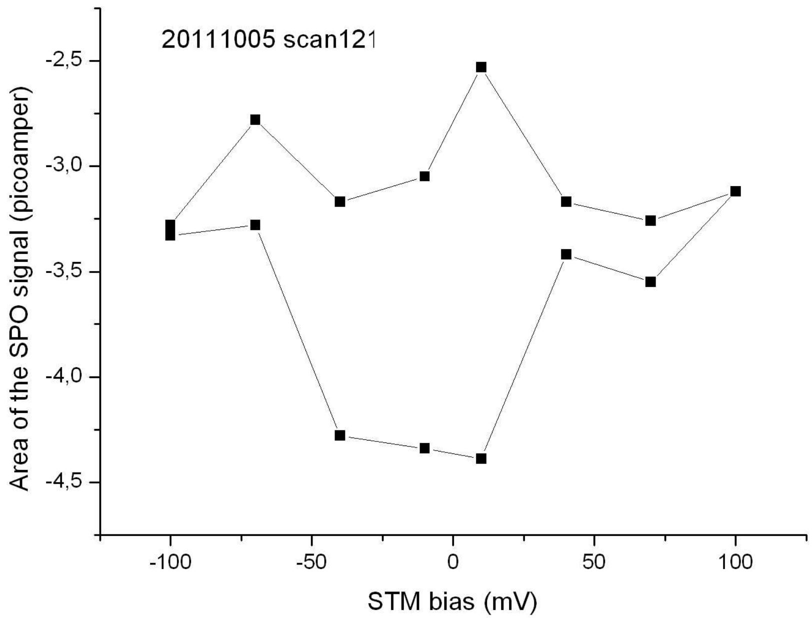
16.08.2008 scan25
Au surface - Au tip
standby 2mV
STM signal (a.u.)
0,15
0,10
0,05
0,00
-0,05
-0,10
-0,15
-0,20
-0,25
-10
-5
0
5
10
STM bias (mV)

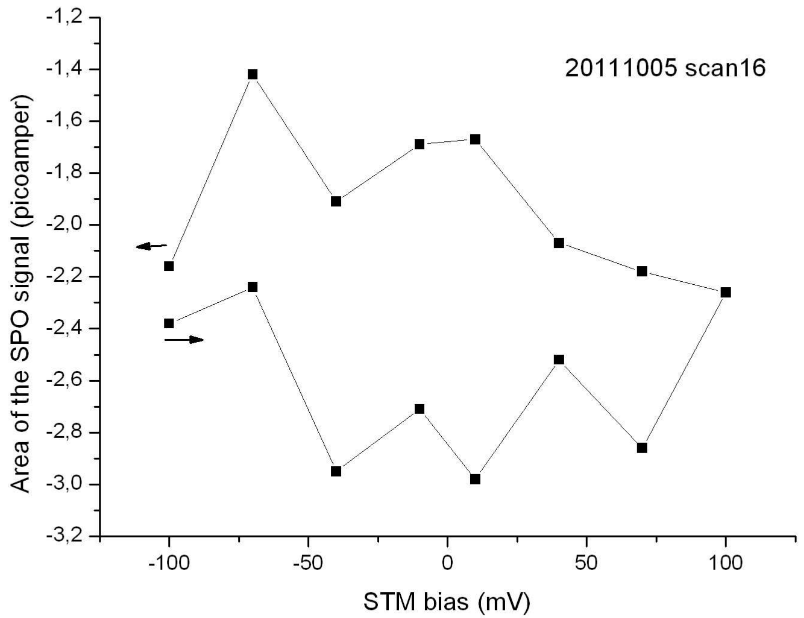
HYSTERESIS!

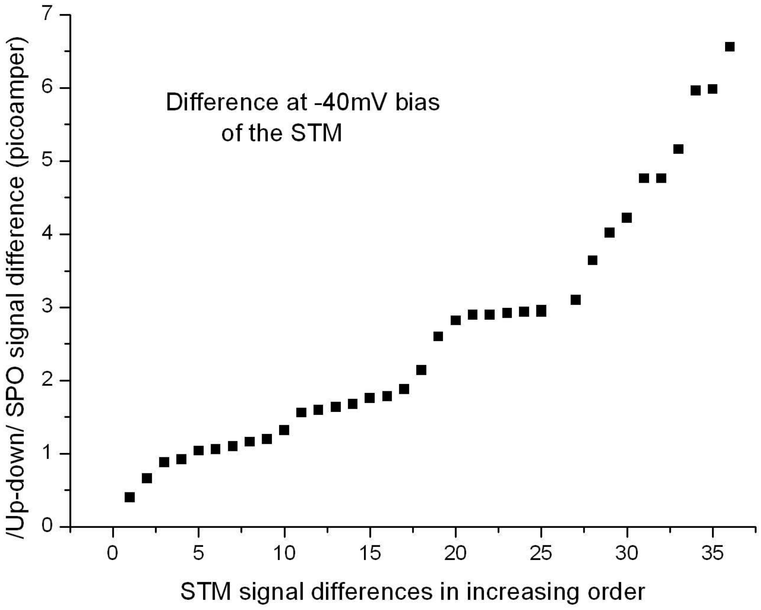


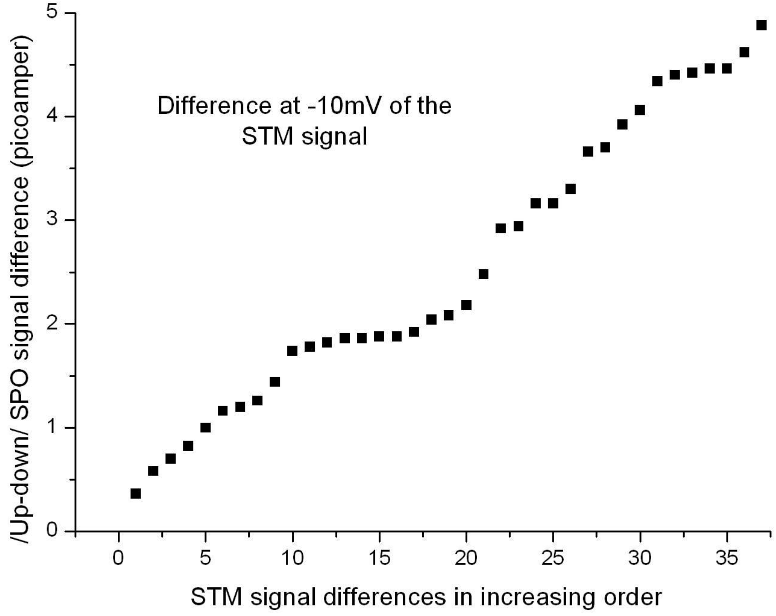


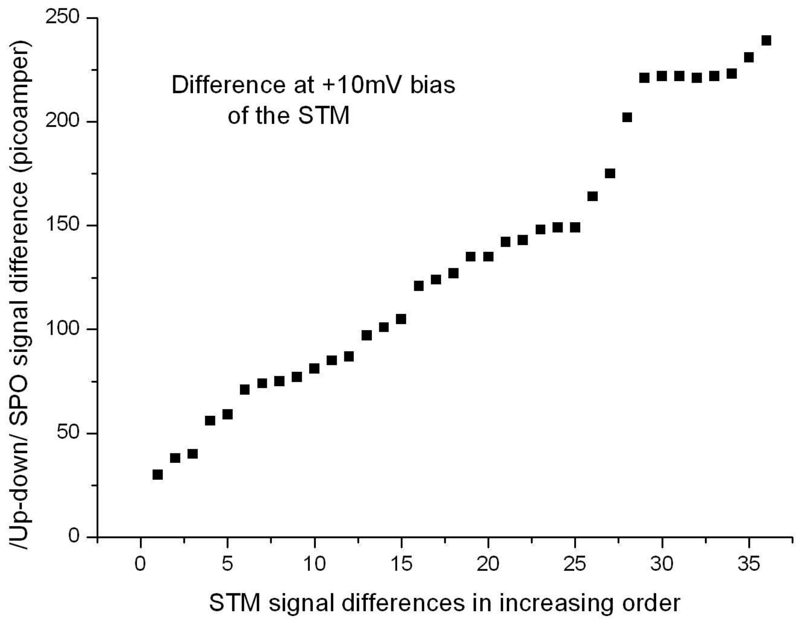
CASIMIR-EFFECT EXCLUDED!

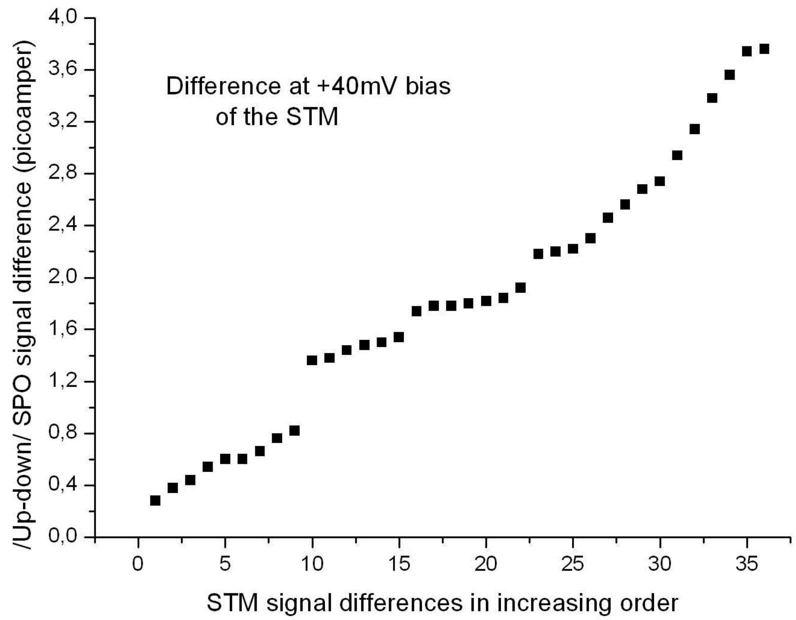


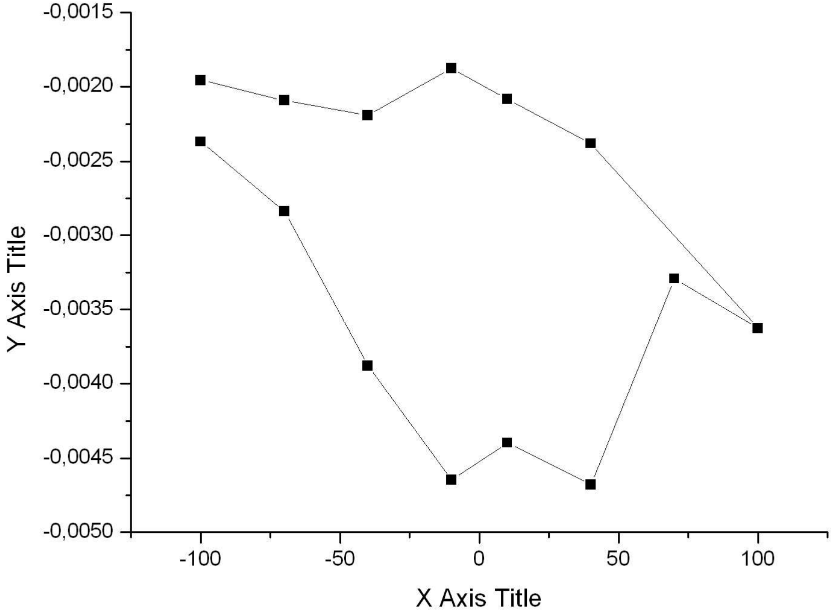


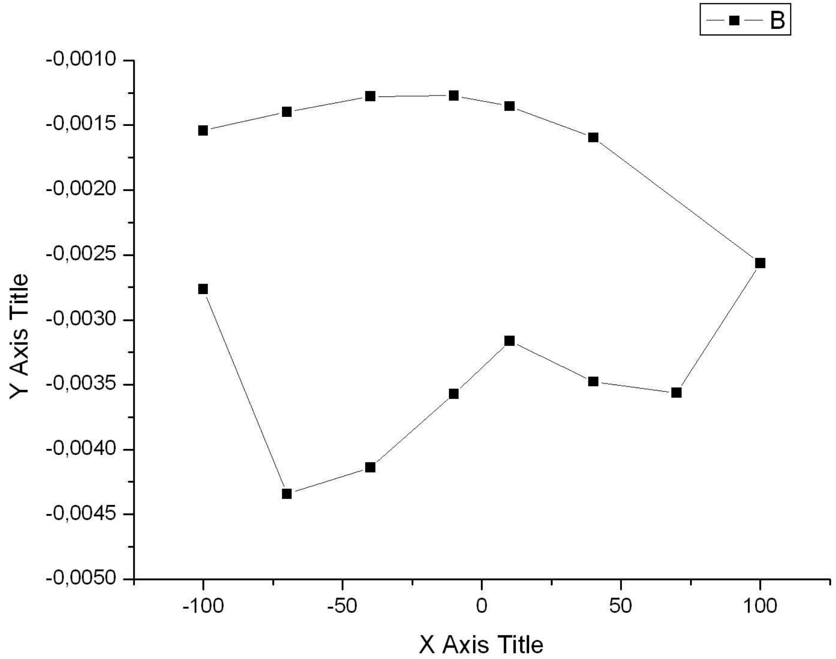

Difference at -40mV bias
of the STM
/Up-down/ SPO signal difference (picoamper)
STM signal differences in increasing order


Difference at -10mV of the
STM signal
/Up-down/ SPO signal difference (picoamper)
STM signal differences in increasing order

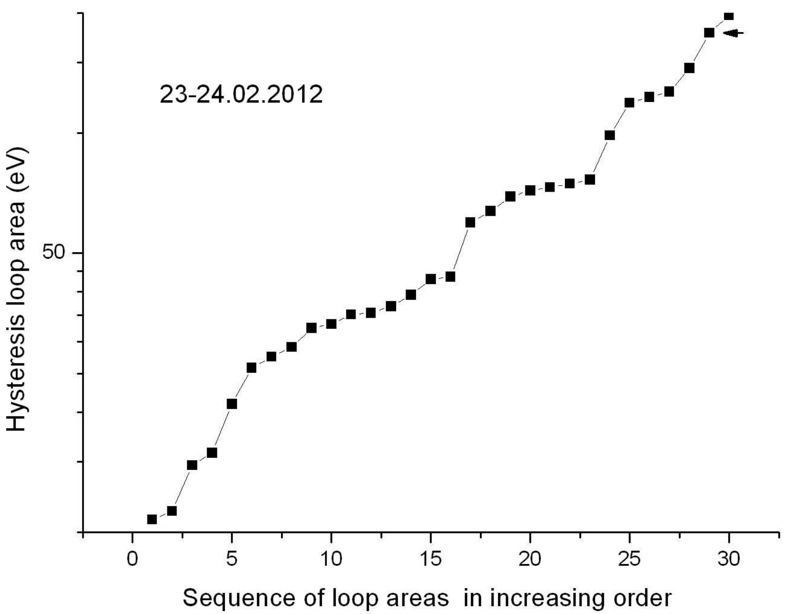

Difference at +10mV bias
of the STM
/Up-down/ SPO signal difference (picoamper)
STM signal differences in increasing order

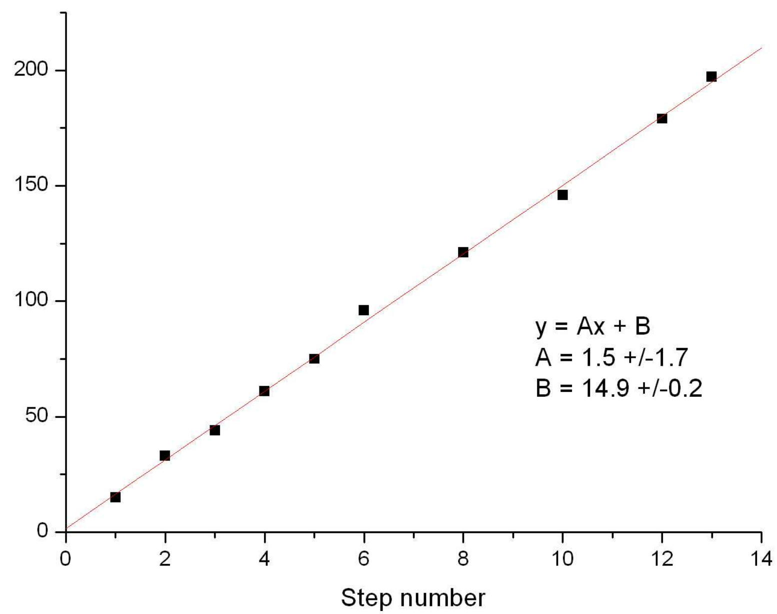

Difference at +40mV bias
of the STM
/Up-down/ SPO signal difference (picoamper)
STM signal differences in increasing order





$I_0 = 4.5.10^{11} W/cm^{2},$ $E_{laser} = 2.10^{7} V/cm$





KÖSZÖNÖM FIGYELMÜKET