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Laser techniques for bottoms-up nanofabrication at the atom scale

(Very successful) laser techniques developed in the last decade(s) to manipulation of neutral atoms in the vapor phase

How can those techniques be used in nanofabrication approaches?

- ***Some of our experimental results with atom lithography***
- ***A few words on how we plan to go beyond atom lithography***
- ***Related problems (at the present stage)***

The group

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NANOCOLD
2001-2004

Outline

1. The (conventional) atom lithography implementation

2. A few words on (basic) laser manipulation tools

3. Our “laser-cooled” cesium beam for atom-lithography

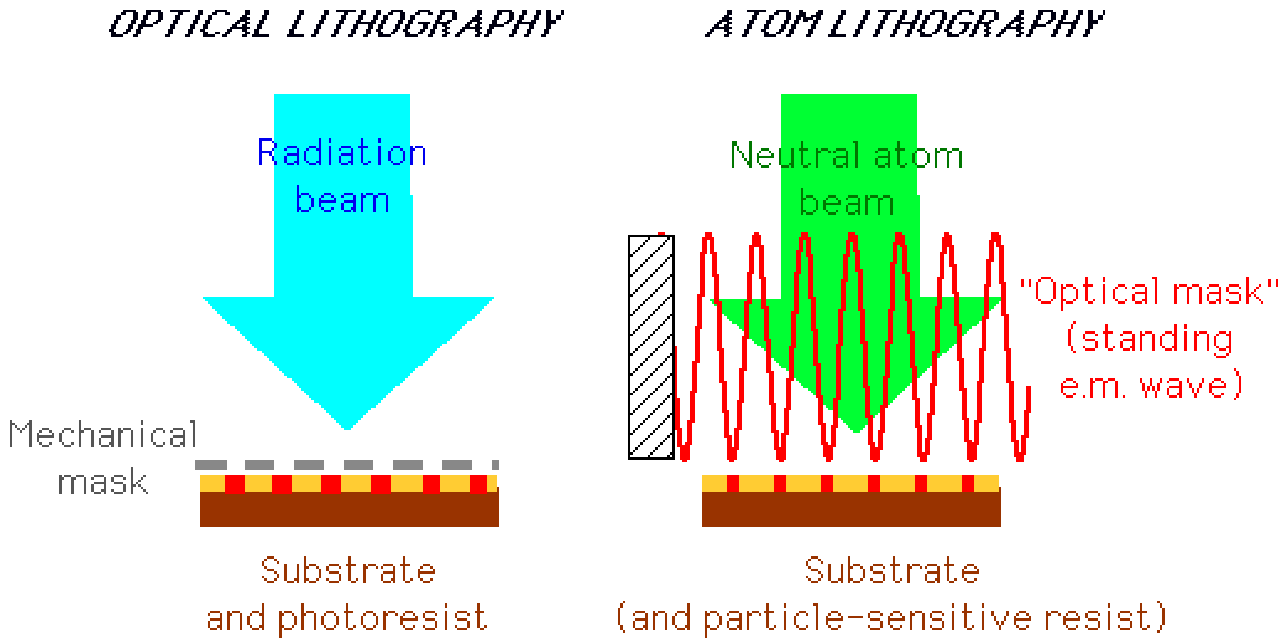
4. Some of our results:
    - Resist-assisted nanolithography;
    - Nanolines on HOPG substrates

5. Beyond atom lithography and our present work:
    - Better space control with few (may be, single) atoms;
    - Atom/surface interaction and related issues

6. Conclusions

1. The atom lithography as a nanofabrication tool

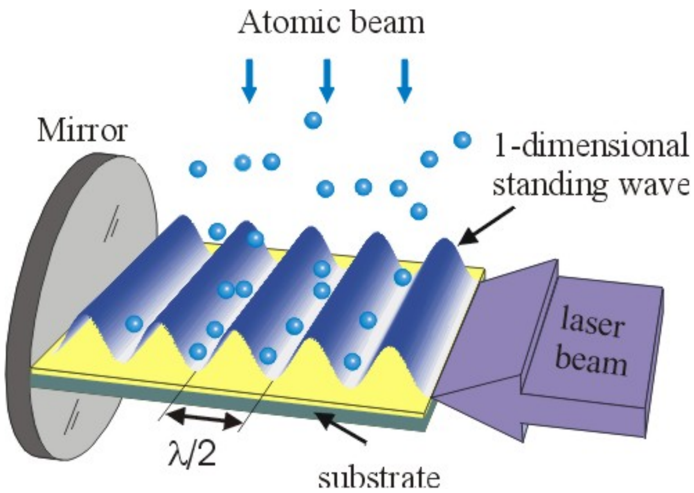
The atom lithography implementation



Ingredients:		Ingredients:
- **light;**	⇨	- **atoms;**
- **mechanical mask;**	⇨	- **optical mask (standing wave);**
- **photoresist;**		- **particle sensitive resist**
- **chemical processing, ...**		- **chemical processing, ...**

First introduced in early 90's (see, e.g., Timp et al., PRL 1992, McClelland et al., Science 1993)

Basic mechanism: dipolar forces



Optical mask (standing e.m. wave)
→ dipolar forces (conservative)
along a direction transverse to atom beam

D. Meschede, H. Metcalf, J. Phys. D 36 R17 (2003)

The optical dipole force acting on an atom with resonance frequency ω_A in a laser field of detuning $\delta = \omega_L - \omega_A$ is derived from the spatial variation of the light shift $\omega_{ls}(\mathbf{r})$ [1]. For a single laser beam travelling in the x-direction with Rabi frequency Ω, the light shift is given by

$$\omega_{ls} = \left[\sqrt{\Omega^2 + \delta^2} - \delta\right]/2\,. \qquad (1)$$

For sufficiently large detuning $\delta \gg \Omega$, approximation of Eq. 1 leads to $\omega_{ls} \approx \Omega^2/4\delta = \gamma^2 s/8\delta$, where $s \equiv I/I_{sat}$, I is the laser beam intensity, $I_{sat} \equiv \pi hc/3\lambda^3\tau$ is the saturation intensity, and $\tau \equiv 1/\gamma$ is the atomic excited state lifetime.

In a standing wave with $\delta \gg \Omega$, $\omega_{ls} = \omega_{ls}(x)$ varies sinusoidally from node to antinode and also spontaneous emission is inhibited so that $\hbar\omega_{ls}(x)$ may be treated as a potential $U(x)$. The resulting dipole force is

$$\mathbf{F}(x) = -\nabla U(x) = -\frac{\hbar\gamma^2}{8\delta I_{sat}}\nabla I(x) \equiv U_{max}\nabla f(x), \qquad (2)$$

where $I(x) = I_{max} f(x)$ is the total intensity distribution of the standing wave light field of period $\lambda/2$, I_{max} is the maximum intensity, and $f(x)$ describes the normalized modulation of the light field. For such a standing wave, the optical electric field (and the Rabi frequency) at the antinodes is double that of each travelling wave that composes it, and so the total intensity I_{max} at the antinodes is four times that of the single travelling wave.

Focused atom trajectories

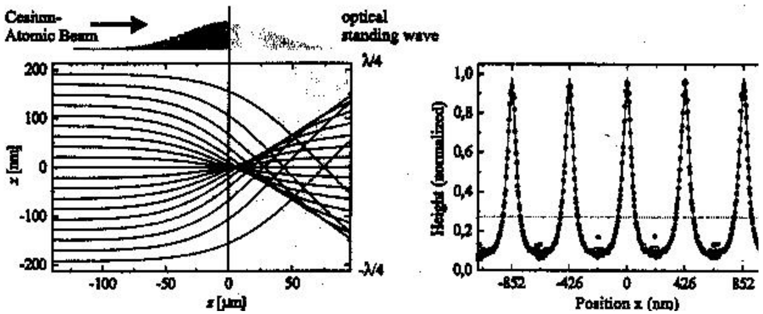


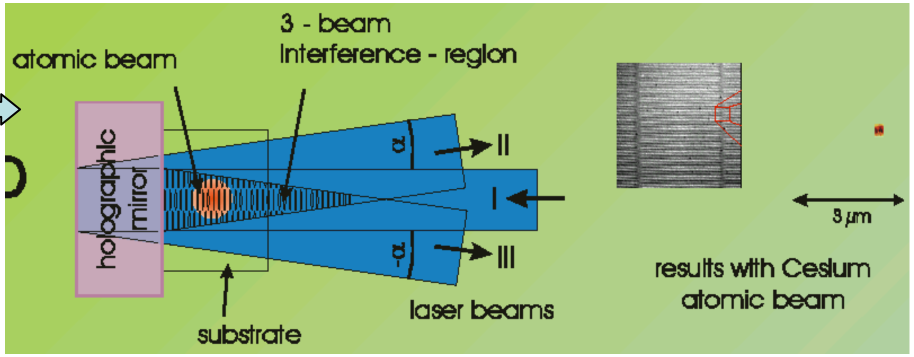
Figure 3. *Left: Numerically calculated trajectories of a laser cooled beam of atoms focussed to the center of a Gaussian envelope standing wave light field (thick lens limit). The focussed laser beam forming the optical standing wave is clipped by the substrate. Note the different scales in x- and z-directions.*
Right: Analysis [25] of flux concentration for a realisitic beam of thermal cesium atoms with 0.1 m/s transversal rms velocity at the focal plane z=0. The dotted line shows the flux distribution without the standing optical wave.

A few pros and cons of atom lithography

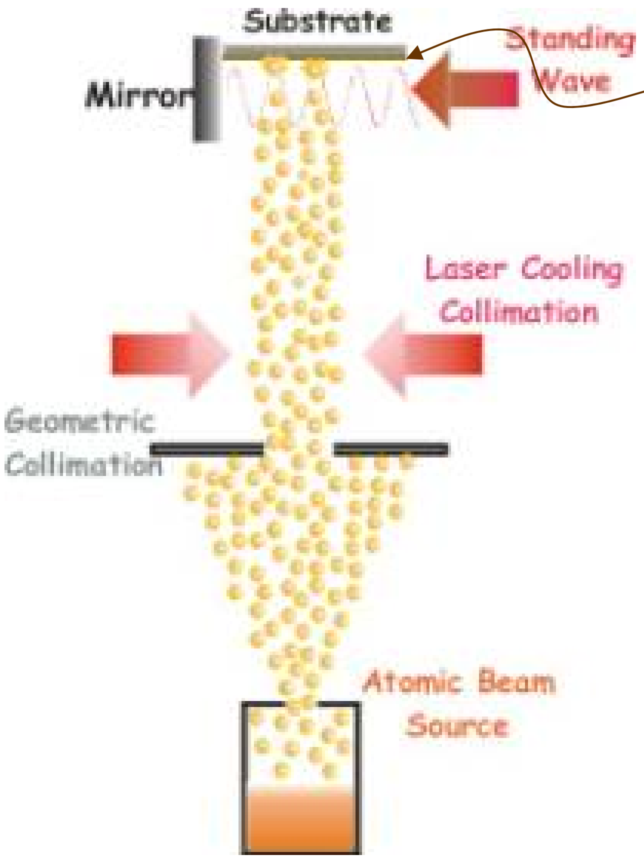
Potential advantages of atom lithography:
- **negligible diffraction (sub-nm de Broglie wavelength)**
- **parallel operation;**
- **non obtrusive, virtually perfect, species-selective mask**

(Evident) limitations:
- **overall efficiency in large-scale applications;**
- **limitations in defining arbitrary patterns (*but attempts have been made to overcome that limitation*)**

Examples:
- ***holograpically generated standing waves (Muetzel et al., PRL 2002)***
- ***“frequency encoding” methods to tune the standing wave phase (Thywissen et al., NJP 2005)***



Requirements on the atom beam



In terms of atom optics, the e.m. standing wave can be seen as an array of lenses for atoms or as an array of potential wells

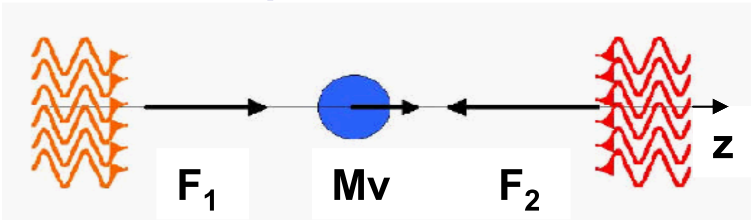
Highly collimated atom beams are required (typ. divergence in the mrad range)

Laser manipulation can provide the required collimation

2. Laser manipulation techniques

Laser-cooling and trapping of atoms

1-D optical molasse

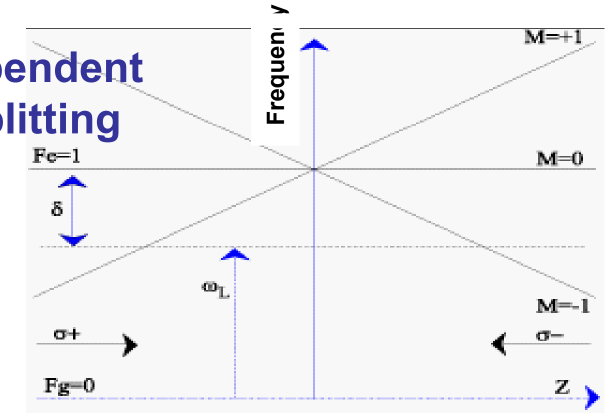


$$\boxed{F = F_1 - F_2 \propto -\gamma v}$$

(dissipative force –
Doppler cooling T~100μK)

Atom/photon momentum exchange

Position dependent
Zeeman splitting



z = 0

$$\boxed{F \propto -k\,z}$$

(restoring force)

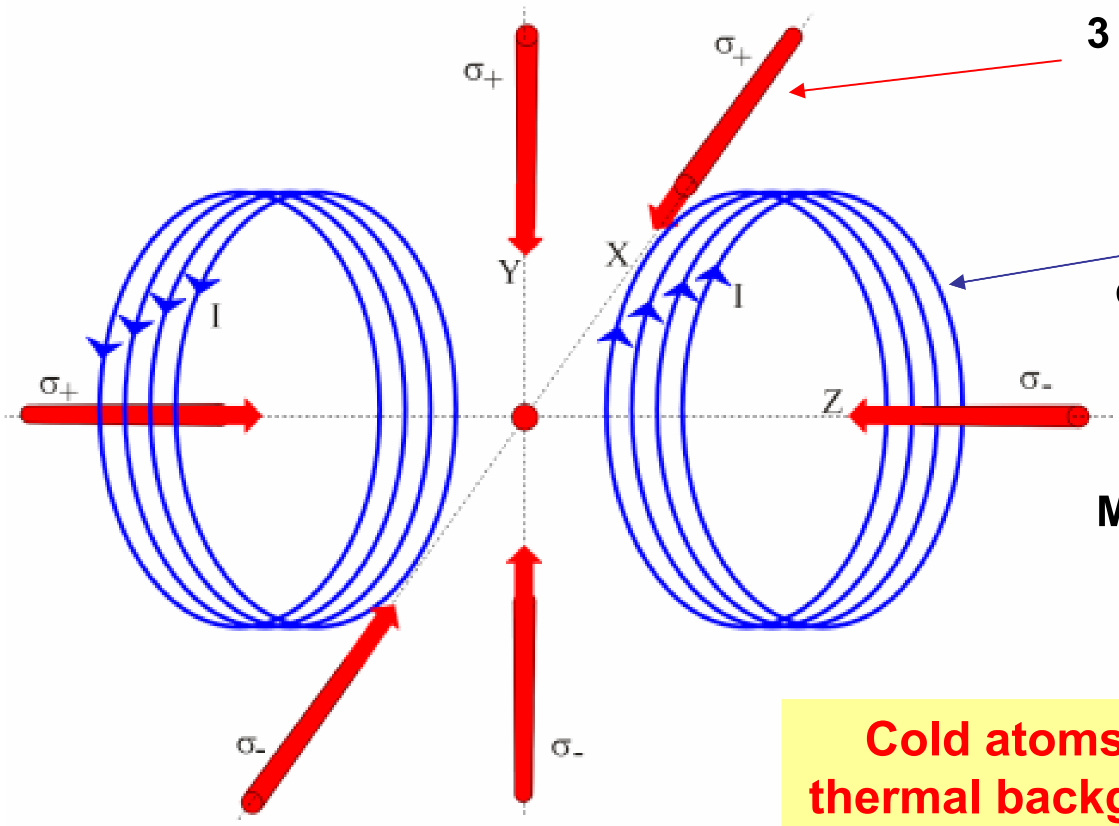
Addition of a linearly varying static magnetic field and trapping close to the zero-field region

Cooling and trapping of cold atoms can be accomplished through an interplay between internal and external degrees of freedom

A very large of additional schemes (e.g., polarization gradient, Sysiphus, RF evaporation, pure optical and magnetic traps, ...) established and demonstrated able to achieve cooling below the 100 nK range

The magneto-optical trap (MOT)

The standard **six-beam MOT**



3 pairs of counterpropagating laser beams (circularly polarized)
+
anti-Helmoltz coils for quadrupolar magnetic field

MOT is typically loaded from the background vapor

Cold atoms are captured from the thermal background, cooled down and confined close to the MOT center

The atom-on-demand scheme

APPLIED PHYSICS LETTERS VOLUME 82, NUMBER 18 5 MAY 2003

Atoms on demand: Fast, deterministic production of single Cr atoms

S. B. Hill and J. J. McClelland[a)]
Electron Physics Group, National Institute of Standards and Technology, Gaithersburg, Maryland 20899-8412

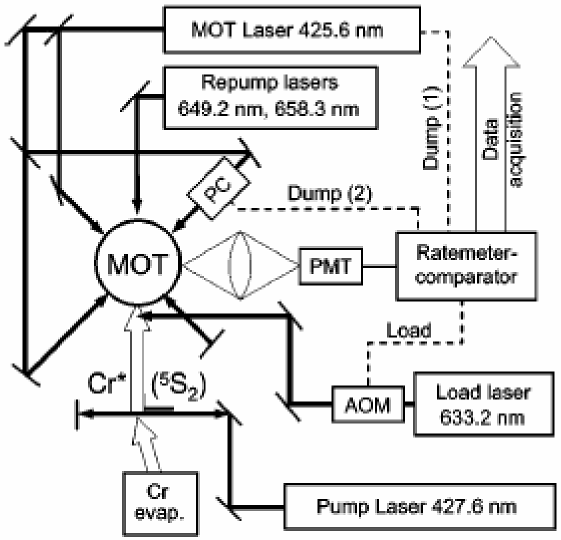
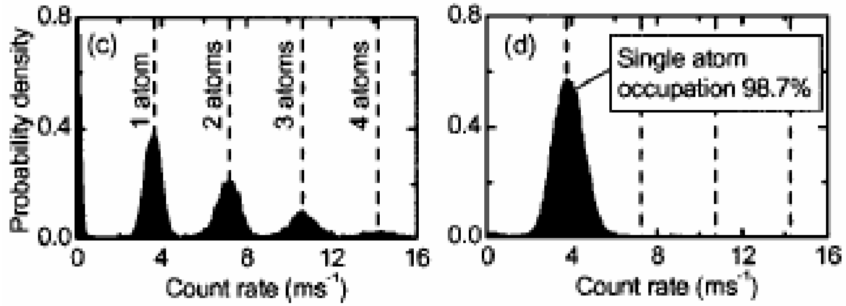


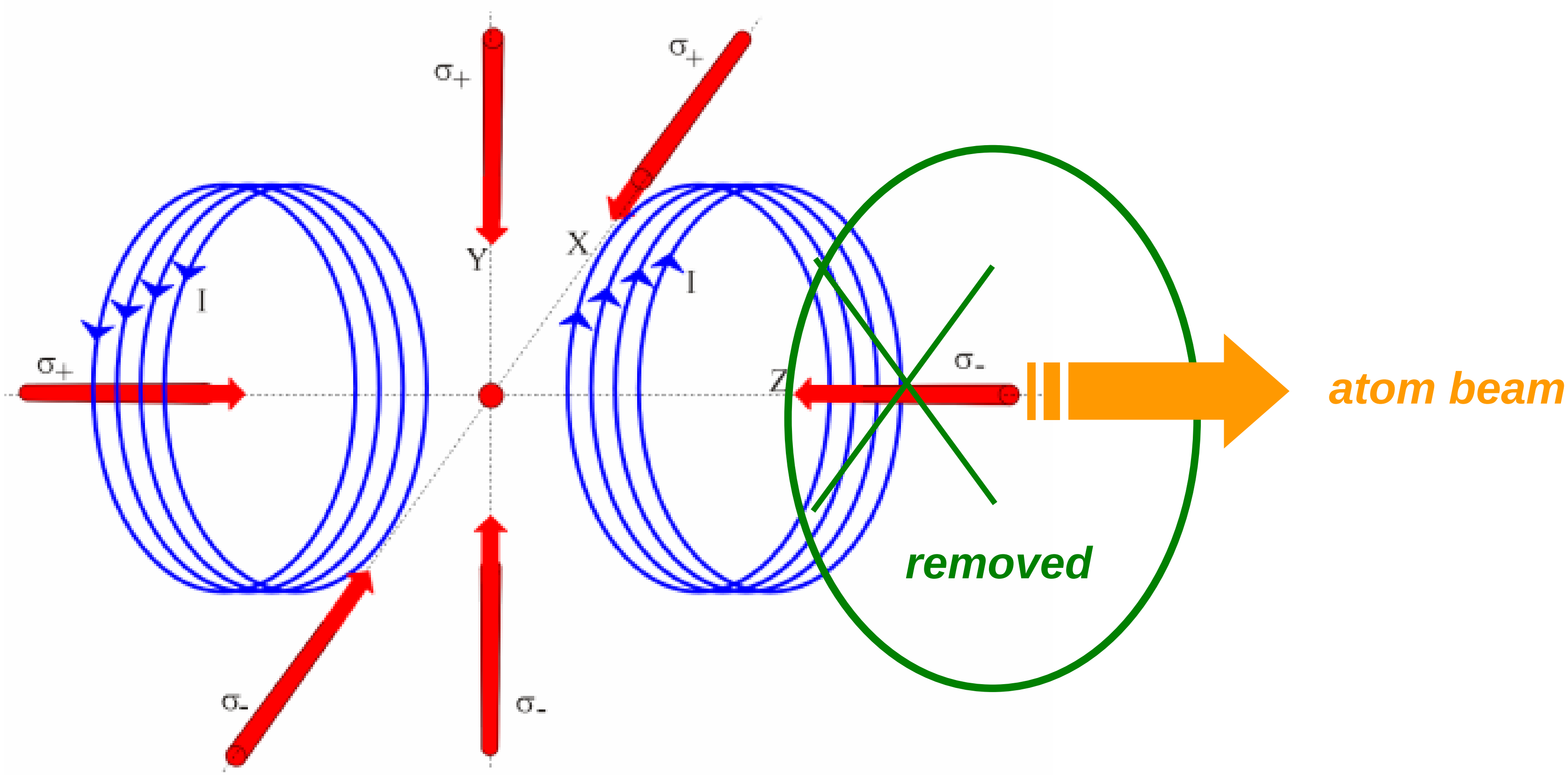
FIG. 1. (a) Schematic of the experiment. A MOT is formed by six beams (two axial beams not shown) from the MOT laser. Two repump lasers prevent trap loss to the metastable $^5D_3$ and $^5D_4$ levels. MOT fluorescence is detected by a photomultiplier (PMT), and then integrated and analyzed by a ratemeter-comparator that generates load and dump gates based on preset thresholds. Line-of-sight loading from a Cr evaporator is blocked. Instead, Cr atoms are deflected, collimated, and optically pumped into the metastable $^5S_2$ state via a pump laser tuned to the $^7S_3 \rightarrow {}^7P_3^o$ transition. When loading is desired, the $^5S_2$ atoms are optically pumped back to the ground state by a load laser gated with an acousto-optic modulator (AOM). The MOT is dumped by either of two methods: (1) momentarily (~1 ms) shifting the MOT laser frequency slightly above resonance; or (2) briefly (~5 ms) pulsing the MOT magnetic field off while simultaneously blocking one MOT laser beam with a Pockels cell (PC). (b) Energy levels of Cr (not to scale),



Schemes can be *readily* implented to have single or few atom control

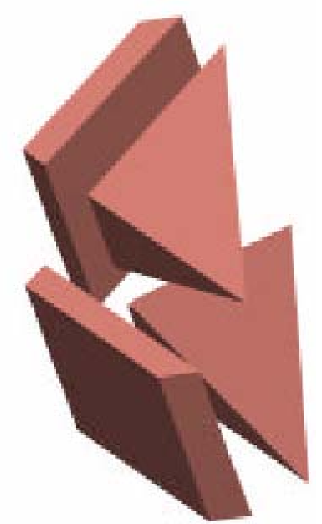
3. Our atom beam

Atom beam from a MOT

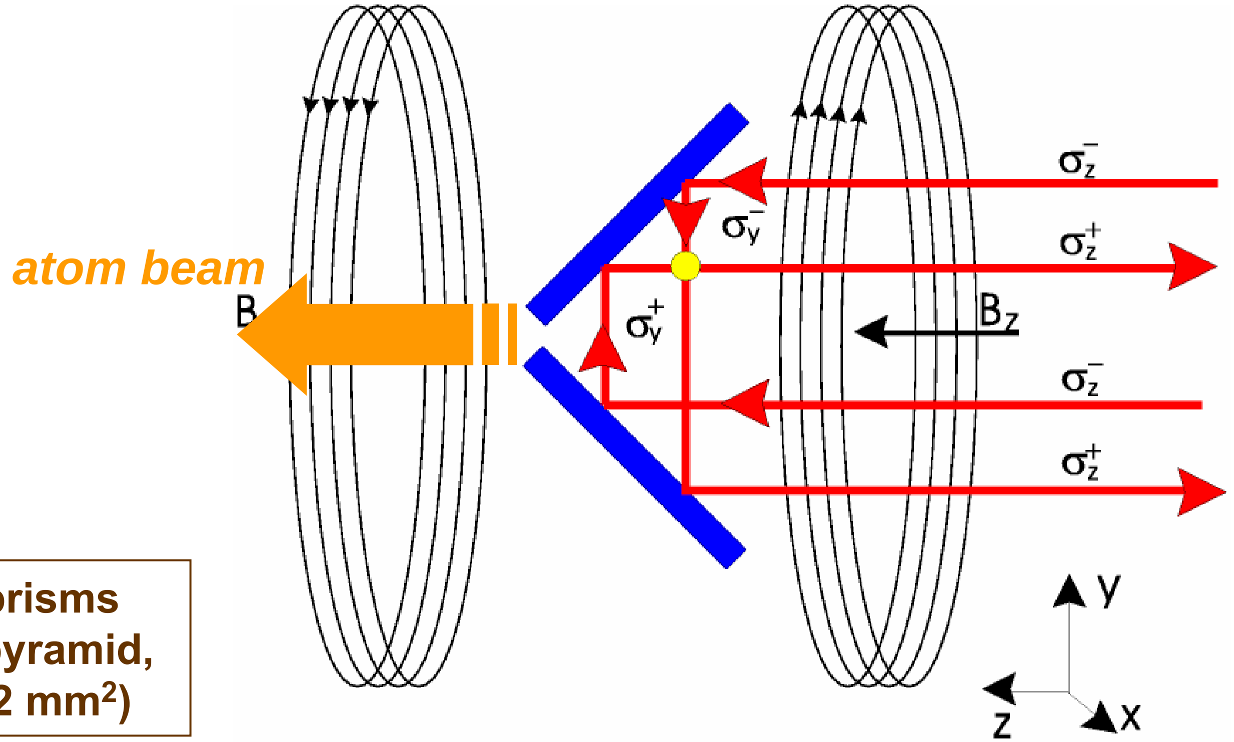


One trapping laser beam removed in a six-beam MOT
$\Rightarrow$ force on atoms unbalanced along one axis
$\Rightarrow$ continuous atom beam achieved

Pyramidal MOT



Two mirrors and two prisms arranged as an hollow pyramid, with an apical hole (1x2 mm^2)



By shining a *single laser beam* into the pyramid, a six-beam MOT is recovered

The MOT *x y* position depends on compensation magnetic fields

When *B* = 0 on the pyramid axis, atoms are pushed out of the hole (atom funnel)

[A. Camposeo, A. Piombini, F. Cervelli, F. Tantussi, F. Fuso, and E. Arimondo, *Opt. Commun.* 200 231 (2001)]

Collimation stage

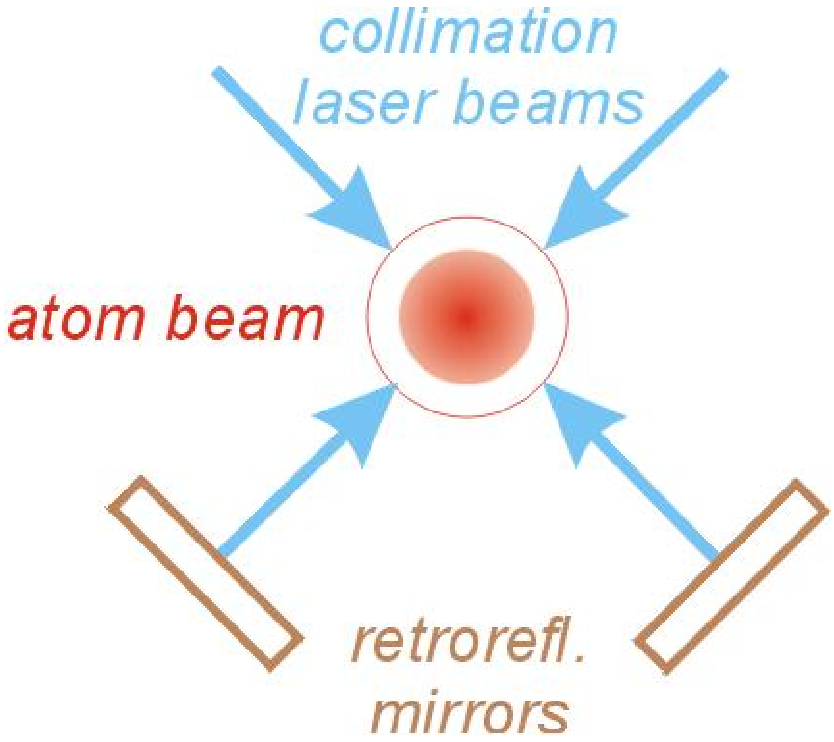
Atoms leaving the "funnel" (the pyramidal MOT) belong to a beam with a ~ 40 mrad divergence, too large for the optical mask to be efficient

Laser manipulation tools (optical molasses) are used to reduce the beam divergence

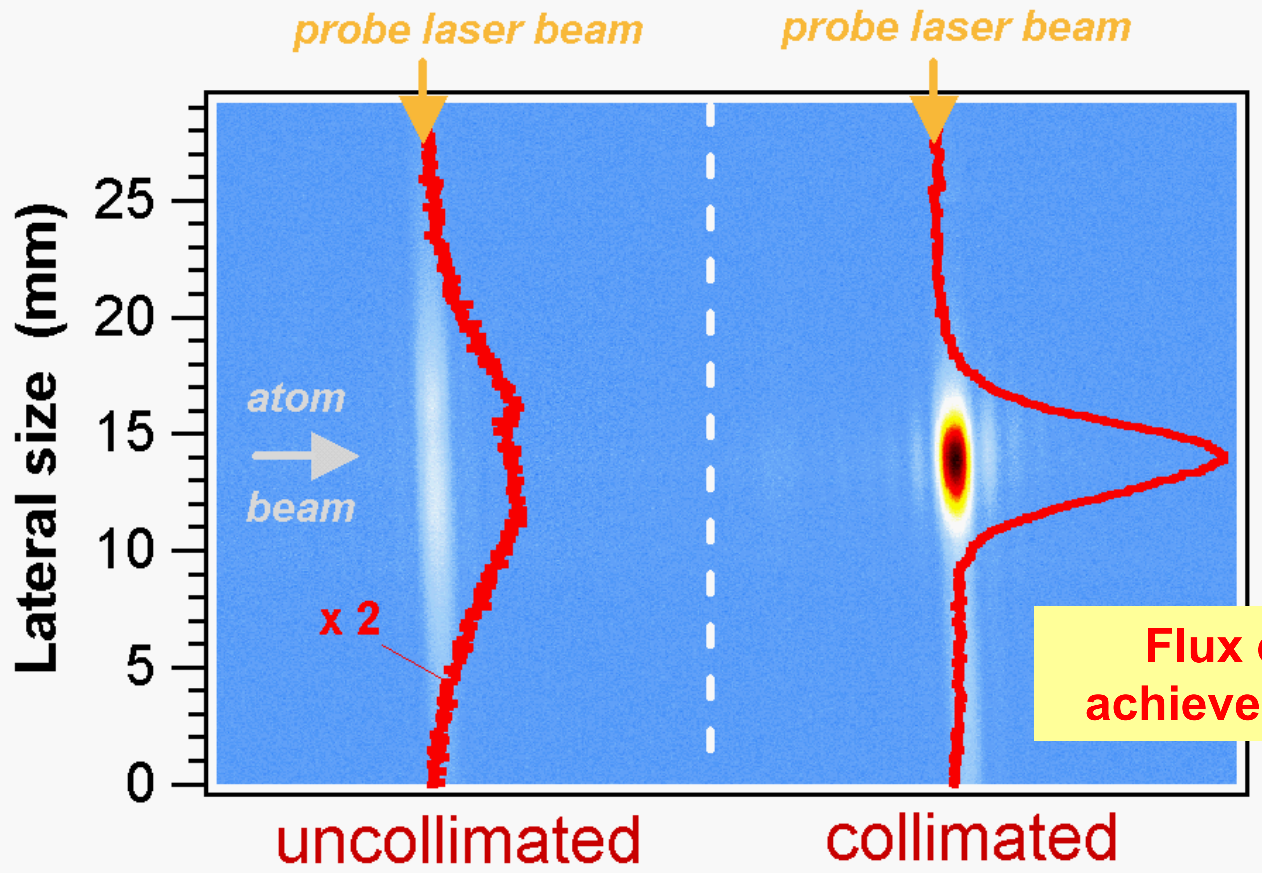
The atom beam must be further collimated

⇩

2D transverse laser-cooling right after the pyramid hole



Atom beam collimation



$\delta_{L,MOT}$=-3.0Γ
dB_{MOT}/dz=12G/cm
$I_{L,MOT}$= =10mW/cm^2
$\delta_{L,coll}$=7.5Γ
$I_{L,coll}$= =4.4mW/cm^2
σ^+/σ^-

Flux enahncement achieved by collimation

Fluorescence emission imaging at the deposition region (around 350 mm downward the pyramid hole)

[A. Camposeo, F. Cervelli, A. Piombini, F. Tantussi, F. Fuso, M. Allegrini, and E. Arimondo, *Mat. Sci. Eng. C* 23 217 (2003); A. Camposeo, F: Cervelli, F. Tantussi, M. Lindholdt, F. Fuso, M. Allegrini, and E. Arimondo, *Mat. Sci. Eng. C* 23 1087 (2003)]

Features of the collimated atom beam

Collimated beam properties at the deposition region (i.e., 600 mm downward the pyramid)	
Long. velocity	7-15 m/s (λ_{dB} ~ 0.3 nm)
Width of long. vel.	$\leq$ 1.5 m/s ($T_{//}$ <10mK)
Beam divergence	< 2 mrad
Atom flux	~ 10^{10} atoms/s
Beam transv. size	~ 4 mm (fwhm)
Atom flux density	up to 10^{11} at. $cm^{-2}s^{-1}$

Relatively intense, collimated, laser-cooled (i.e., longitudinally slow) atom beam produced

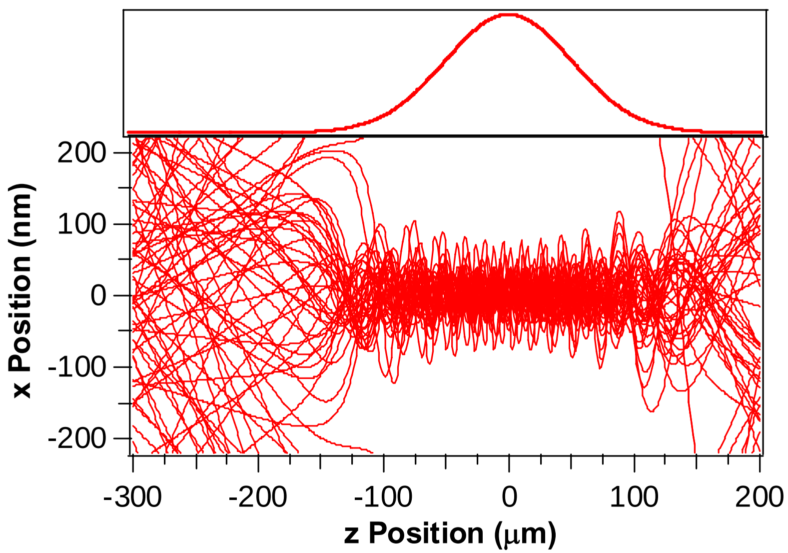
4. A few results

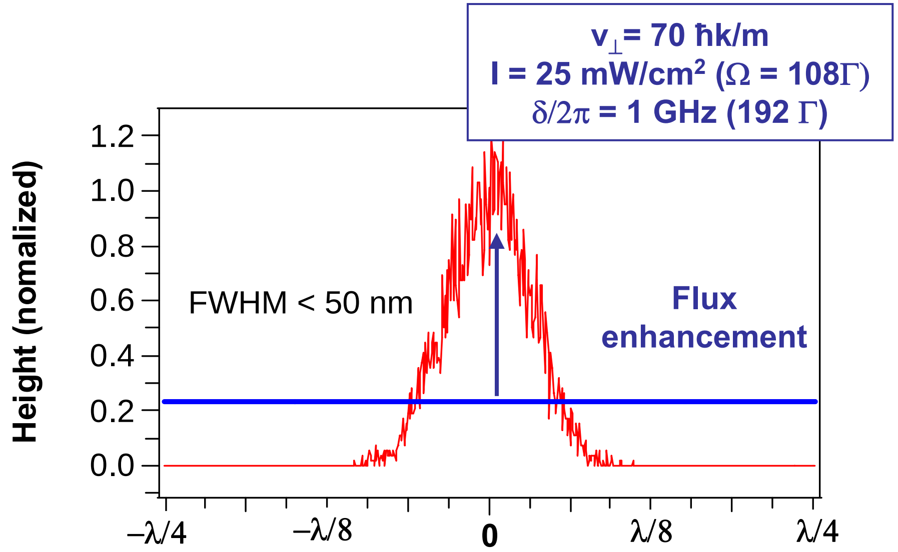
Trajectories of the atoms impinging onto the substrate

Simple model:
dipolar force between atoms and quasi-resonant, space inhomogenous e.m. field

$$F_{Dipole} = -\frac{\hbar\,\Gamma^2}{8\,\Delta}\,\frac{\nabla I(x)}{I_{Sat}}$$

Standing wave space profile

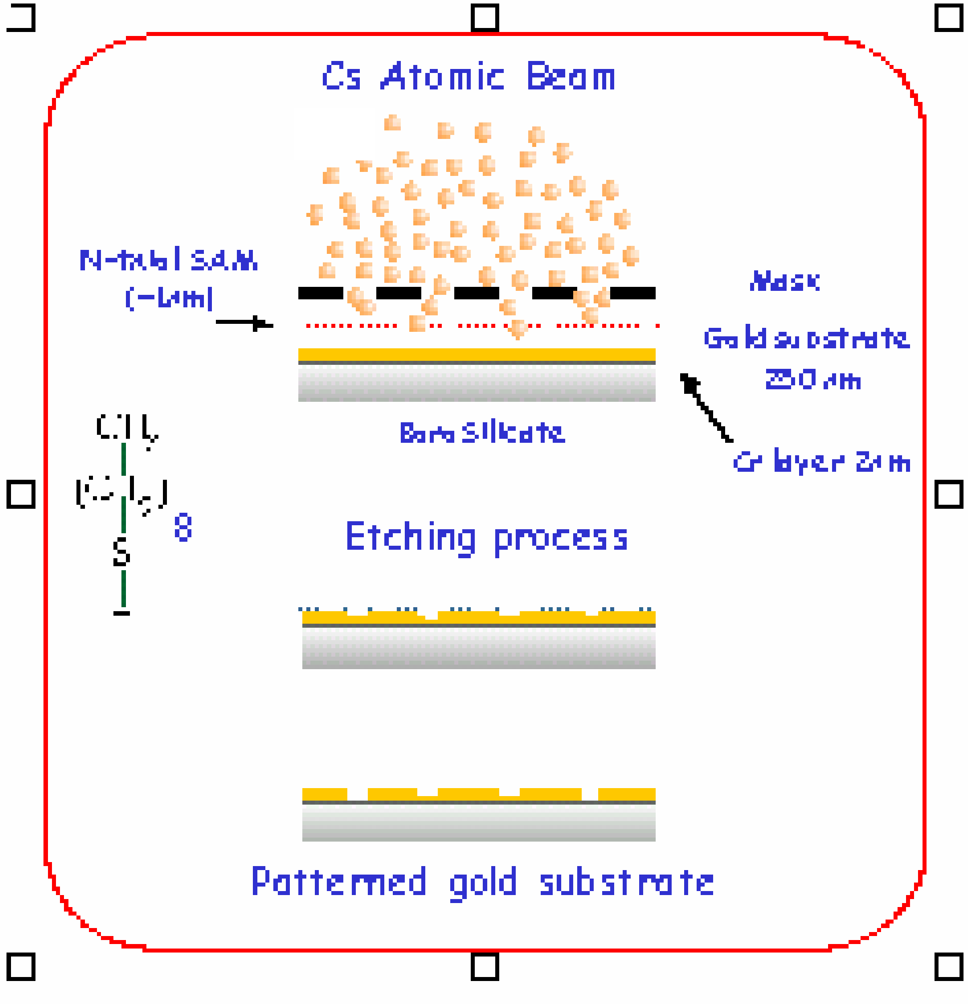




[F. Tantussi et al., MSEC (2006)]

Due to the small velocity (and the long interaction time) atoms are channeled by the standing wave rather than focused as in conventional schemes → *negligible background*

Resist-assisted atom lithography

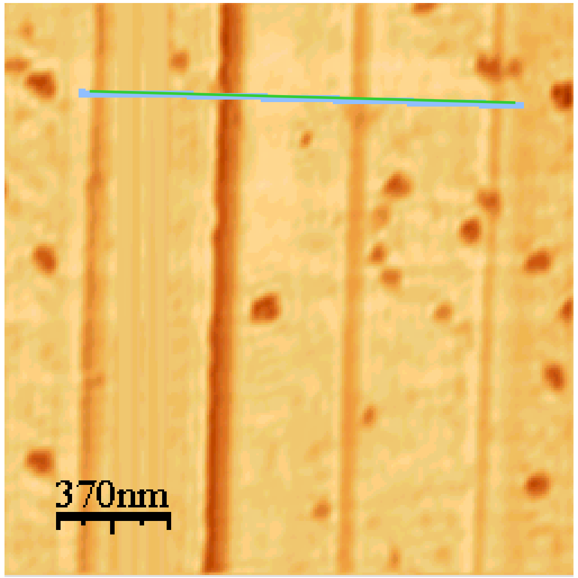


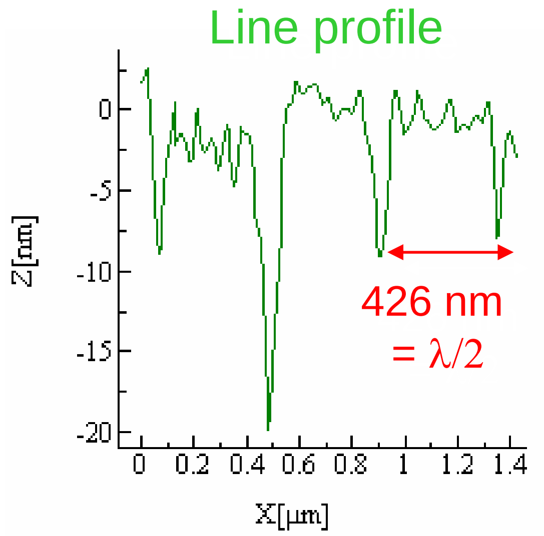
SAM impression mechanism possibly interpreted
by Cs: C. Di Valentin,
A. Scagnelli, G. Pacchioni, J. Phys. Chem. 109 1815 (2005)

Nanopatterning of the resist and subsequent etching accomplished as a test for the technique

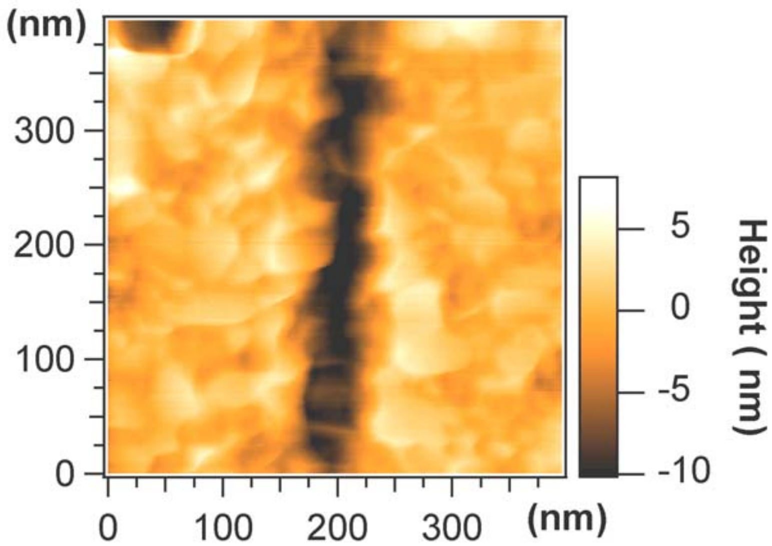
- Particle-sensitive resist:
  - Self Assembled Monolayer of nonanethiol [$CH_3(CH_2)_8SH$] onto gold
  - substrate cleaning with Piranha
  - 24 hrs dipping into 1-5 mM SAM solution in ethanol
- Substrates: gold layers deposited onto Si (or glass), with a Cr adhesion layer
- Wet-etching process:
  - cyanate-based solution
  - etching speed ~ 4 nm/min

A “representative” example





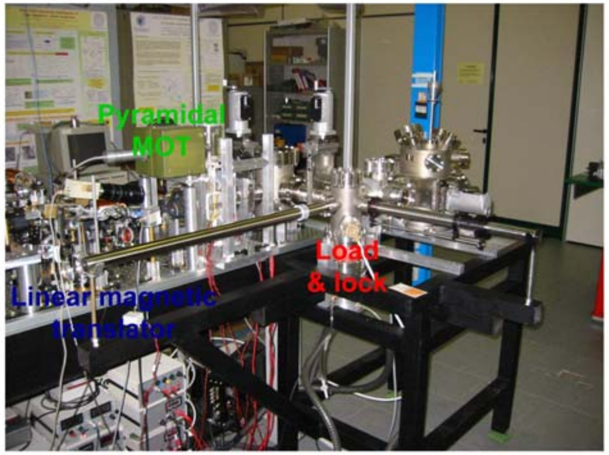
AFM image – plan view
Sputtered gold 30 nm thick
2h deposition and 13 min etching
Estimated dose: 2 atoms/SAM molecule

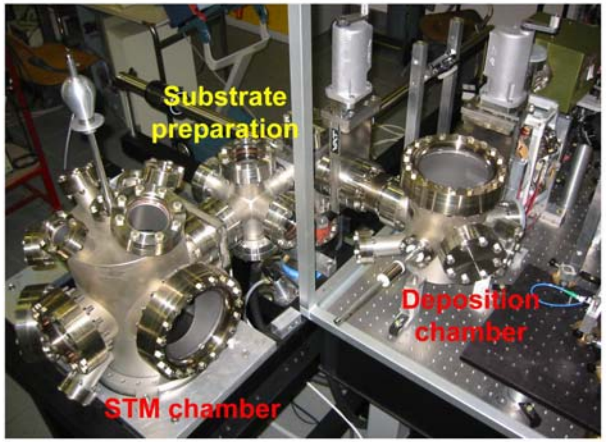


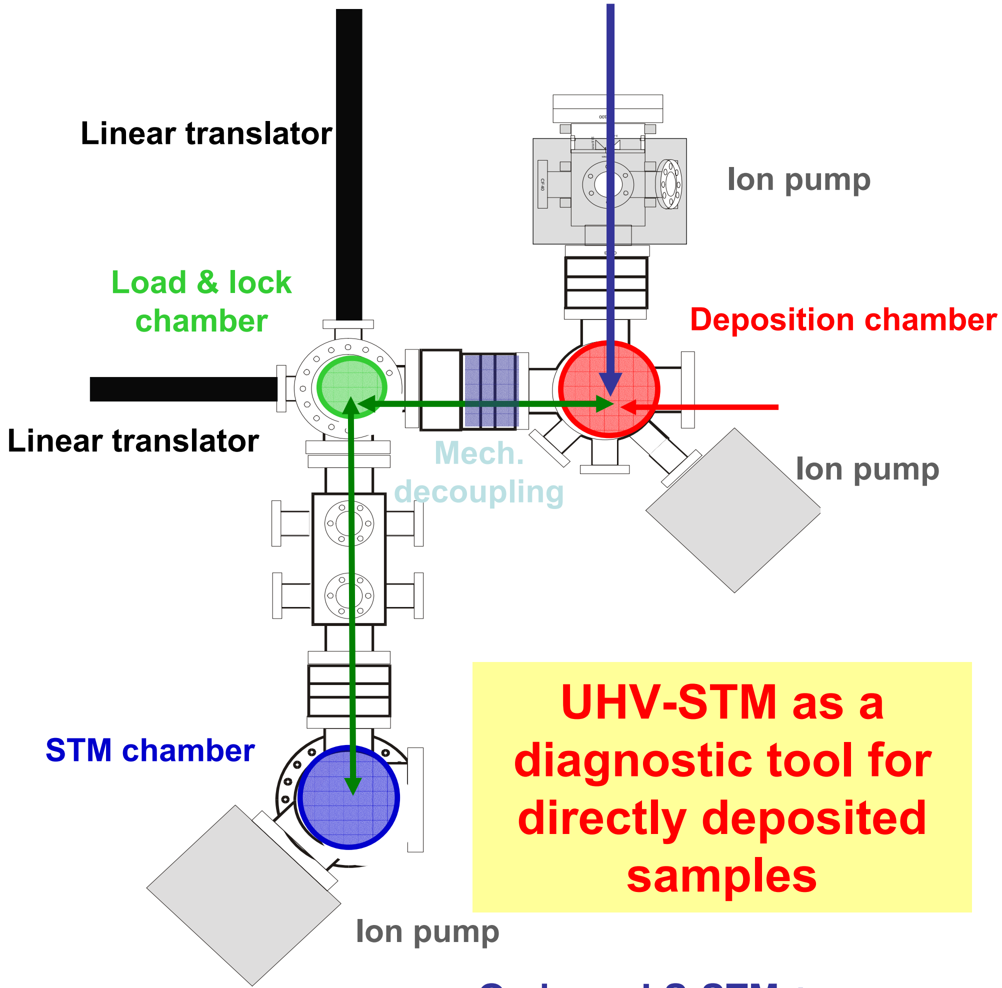
The granular structure of substrate/SAM affects the results

[C.O’Dwyer, G.Gay et al., Nanotechnology 16 1536 (2005)]

Direct deposition and diagnostics

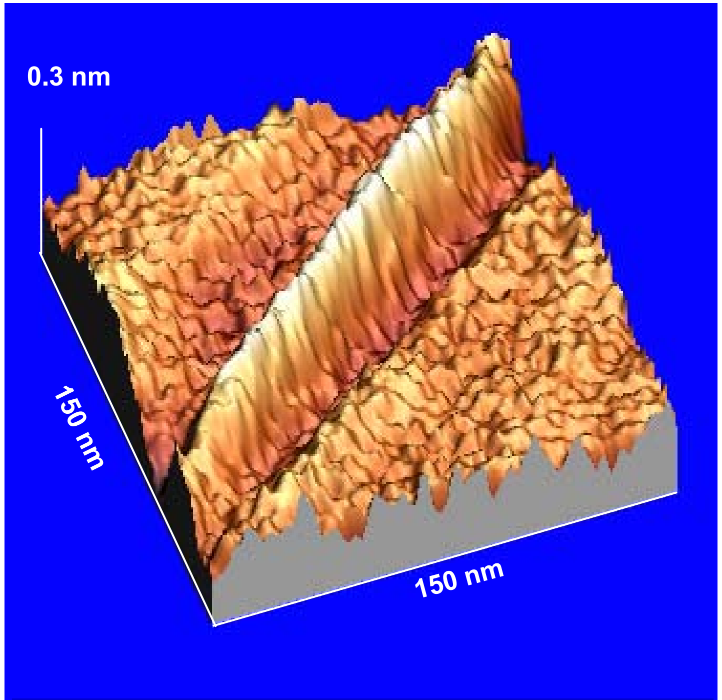






Omicron LS-STM + Nanotec Dulcinea controller

Nanolines on HOPG

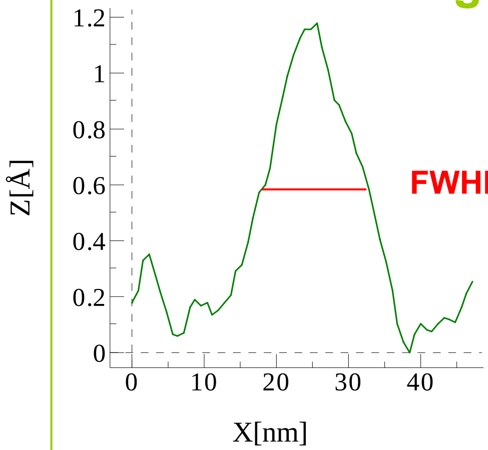


UHV-STM image – plan view of a single line
HOPG substrate (peeled off)
3h deposition – detuning 1 GHz
Standing wave intensity 22 mW/cm^2

Nanolines are observed after exposure of a HOPG substrate (previously "flat") in the presence of standing wave (optical mask)

Nanoline direction is in agreement with the standing wave geometry

Profile of a single line



Improving reproducibility is in progress along with detailed interpretations

5. Future directions, “surface” issues, our present work

A few planned steps and related problems

A few thougths for going beyond atom lithography we are presently working on:

- To exploit the small atom kinetic energy with soft-materials
  → test of other resists and surface decoration
- To enhance pattern definition flexibility (parallel operation)
  → e.g., excitation to Rydberg states + inhomeg. electrostatic fields

- To reduce number of atoms while keeping spatial control ability (e.g., for *precise-doping* purposes)
  → atom-on-demand schemes
  → use of the beam to load a *localized* atom trap and subsequent *soft-landing*-like arrival onto a substrate

Basic question: can we really have the chance to place *few atoms* in predefined positions over a surface?

⇓

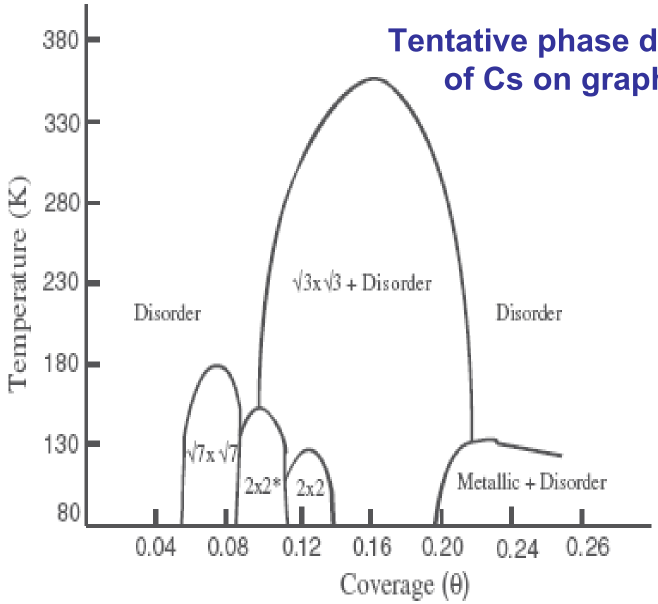
"Surface" issues!!

(e.g., diffusion, coalescence, role of substrate defects,..)

Deposition from the laser-cooled Cesium beam

Furthermore:
our beam is "laser-cooled" (subthermal transverse *and longitudinal* velocity):
is this playing any role? (we expect no, since substrate is at room temperature)

Models for nanostructure growth usually predict thermal-activated barriers
(see, e.g., E te Sligte et al., JAP (2004) and F. Nita, A. Pimpinelli, JAP (2005))



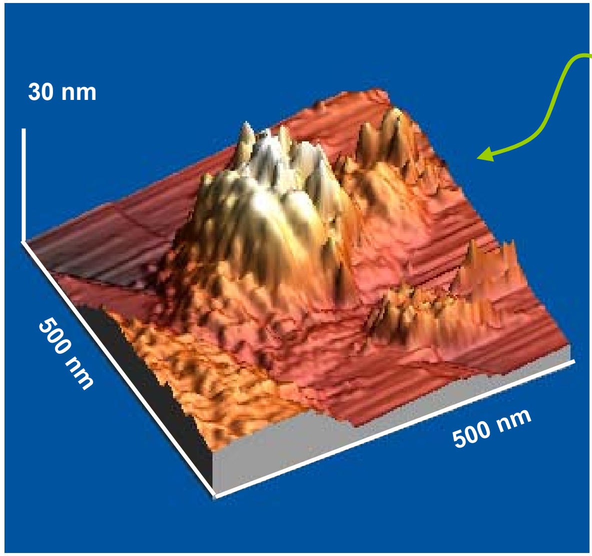
See, e.g., ZP Hu et al., PRB (1986);
M. Caragiu, S. Finberg, JPC (2005)

Alkali growth (including Cs) on graphite well studied in the past in conventional (thermal) deposition conditions

Preliminary assessment of laser-cooled Cs deposition onto HOPG worth to be carried out

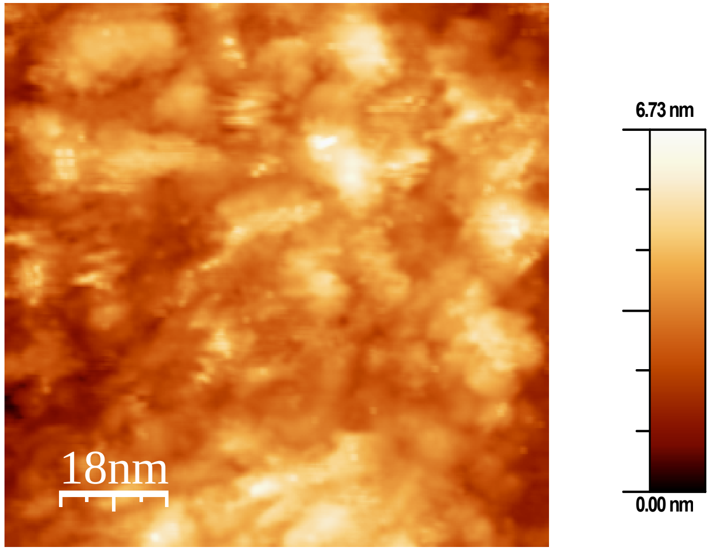
Depositions without the standing wave

"Unstructured" depositions at "large" exposure dose



UHV-STM image – 3D view
HOPG substrate (peeled off)
Standing wave off
Estimated coverage ~ 0.4

Non uniform substrate coverage
Density enhanced close to the HOPG fracture planes

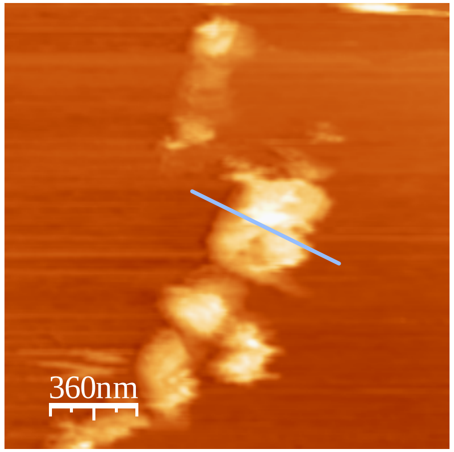


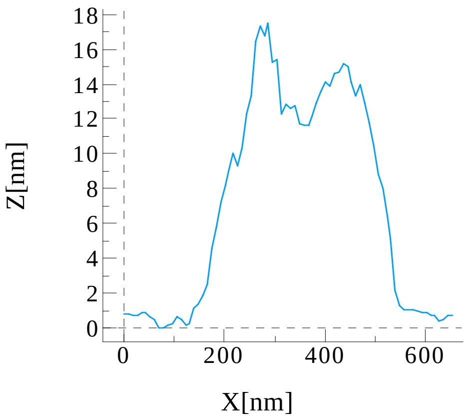
UHV-STM image – plan view
HOPG substrate (peeled off)
Standing wave off
Estimated coverage ~ 0.4

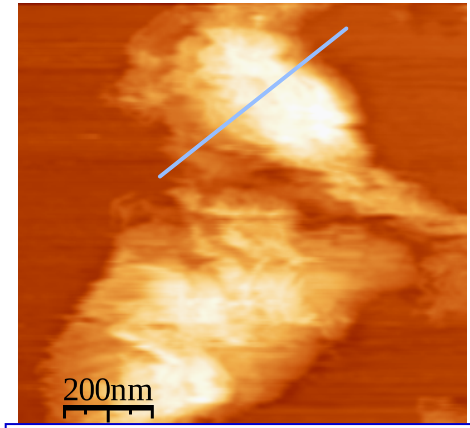
More homogeneously covered regions show grain-like features

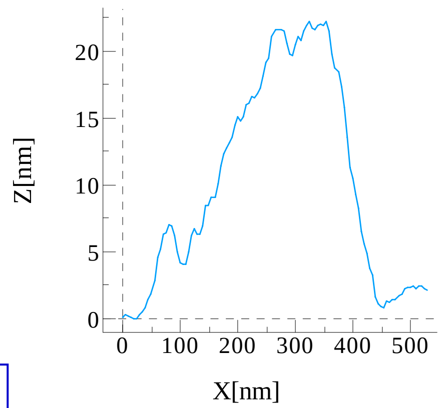
Depositions from a thermal source

For a comparison: depositions from an effusive (hot) Cs source







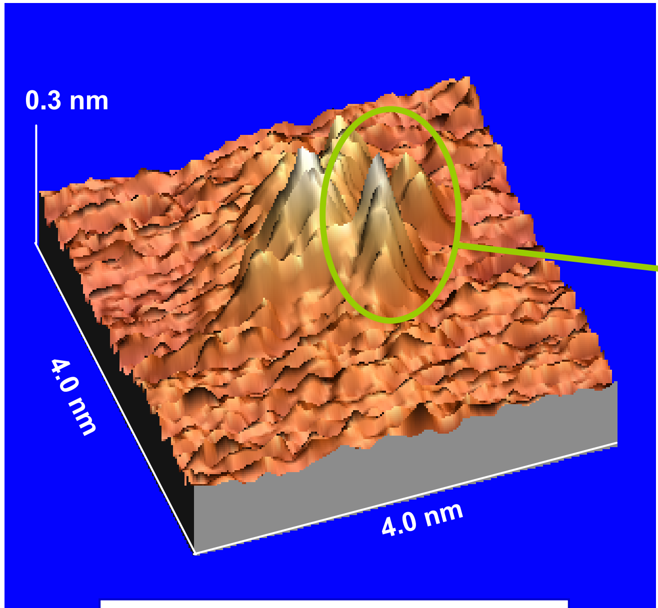


UHV-STM images – plan view
HOPG substrate (peeled off)
Standing wave off
Estimated coverage > 0.4
Hot cesium beam

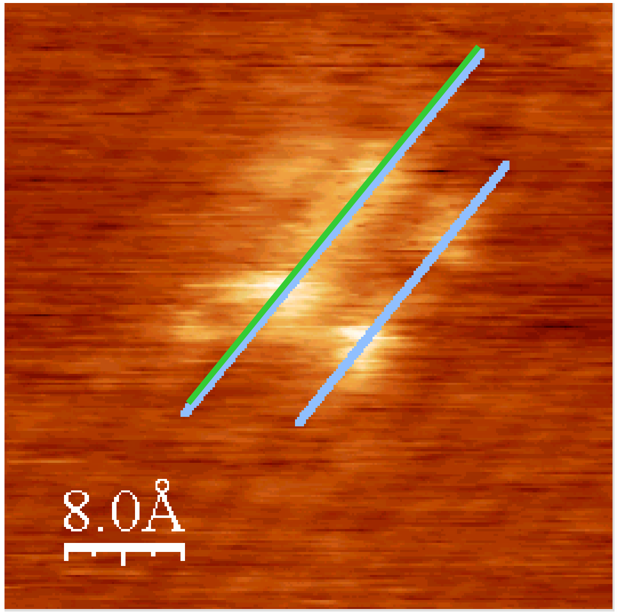
- Strongly non homogeneous space distribution
- "Droplet-like" morphologies possibly related with wetting properties of cesium

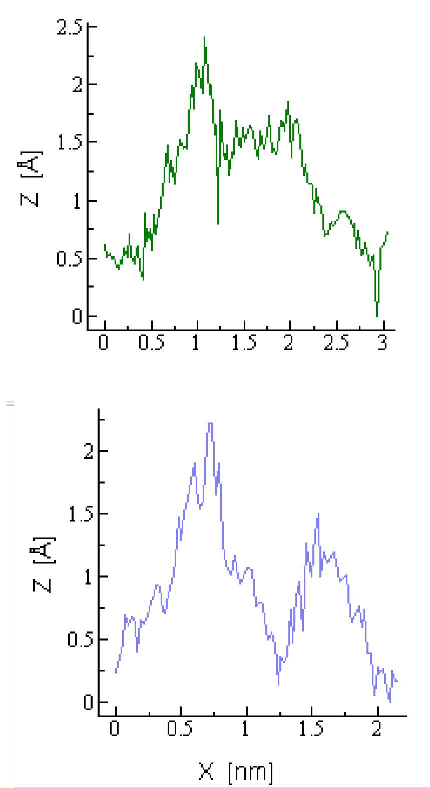
Comparison between thermal and laser-cooled deposition features is presently under investigation

Cesium nanoislands at the low coverage, cold regime



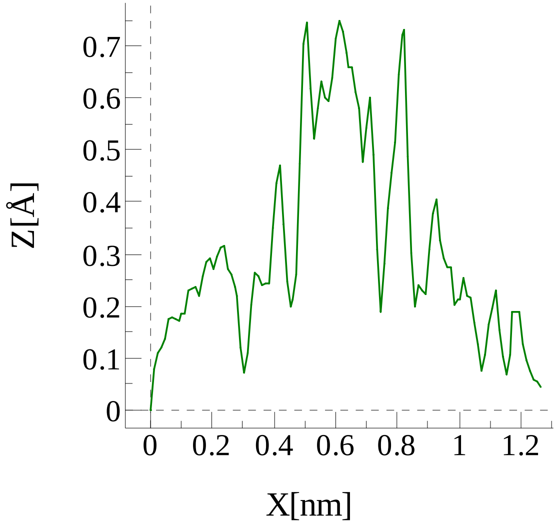
UHV-STM image
HOPG substrate (peeled off)
estimated coverage <0.1



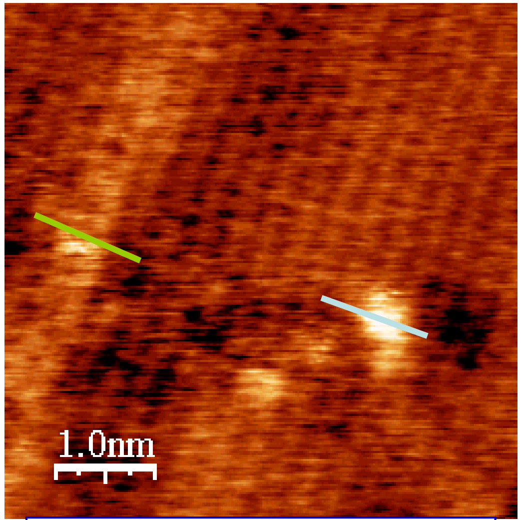


Cesium nanoislands observed after (unstructured) exposure at a small dose appear consisting of "few" atoms

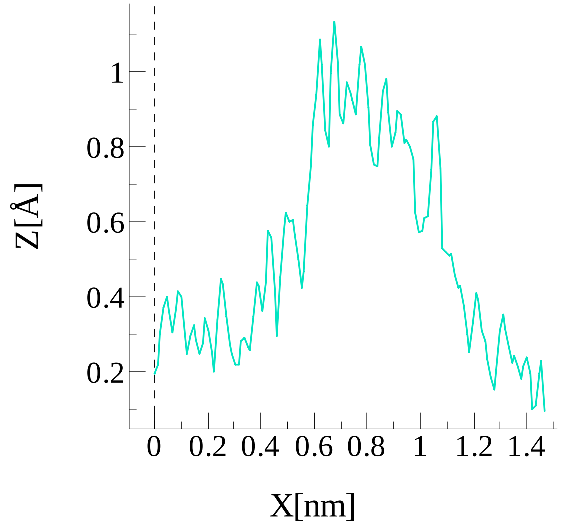
Cesium nanoislands II



A wide variety of nanostructure morphologies is observed at small exposure dose (without standing wave)



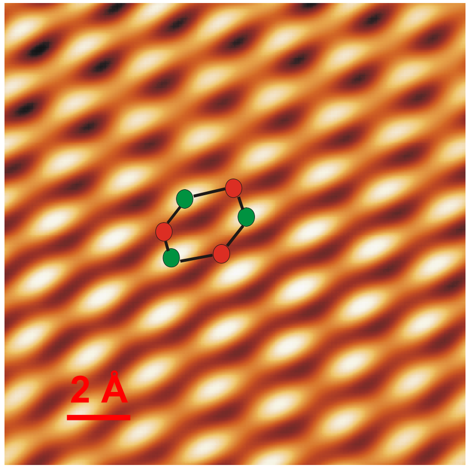
UHV-STM image
HOPG substrate (peeled off)
esimated coverage <0.1

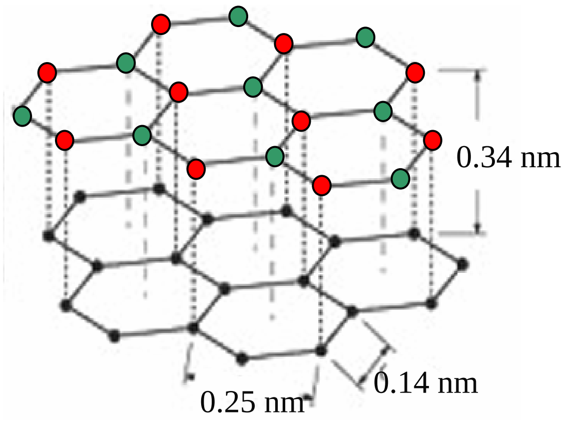


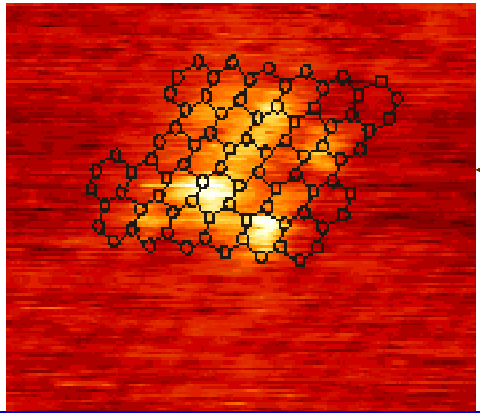
Diffusion below the uppermost graphite layer (intercalation) might play a role, possibly enhanced along a graphite plane fracture

Cesium atom sites on HOPG

Pure (unexposed) graphite surface







UHV-STM image – plan view
HOPG substrate (peeled off)
estimated surface coverage <0.1

After exposure to the laser-cooled cesium beam (at "small" exposure dose) without the standing wave

Cesium atoms appear to occupy hollow sites in graphite

Islands are formed which are stable
(at least on the few days timescale)

Conclusions

- Laser techniques have been developed to accurately control the dynamical properties of neutral toms in the vapor phase
- Laser techniques can be used in fabrication schemes, e.g., in atom lithography
- Laser manipulation schemes can be implemented able to address (positional and dynamical) control of few or single atoms: *we are working on that*
- Issues pertaining to the atom/surface interaction are expected to get larger and larger relevance whenever few atoms are concerned: *we are working on that*