Coherence and Decoherence of Large and Hot Molecules

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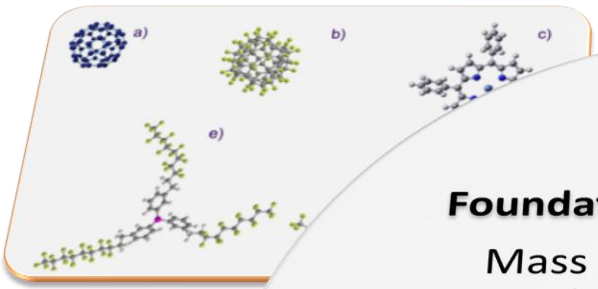
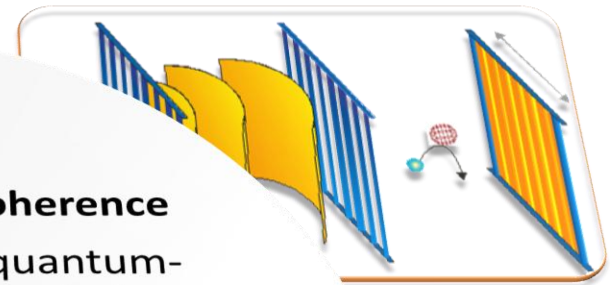
ECT Workshop, Trento 2010

Overview





Foundations

Mass and complexity limits

Decoherence

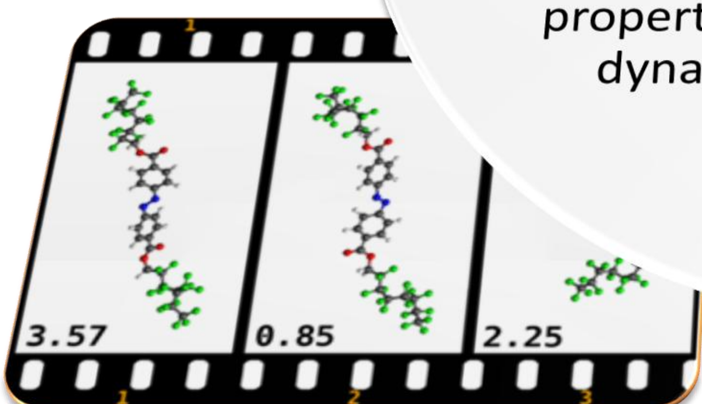
The quantum-classical transition

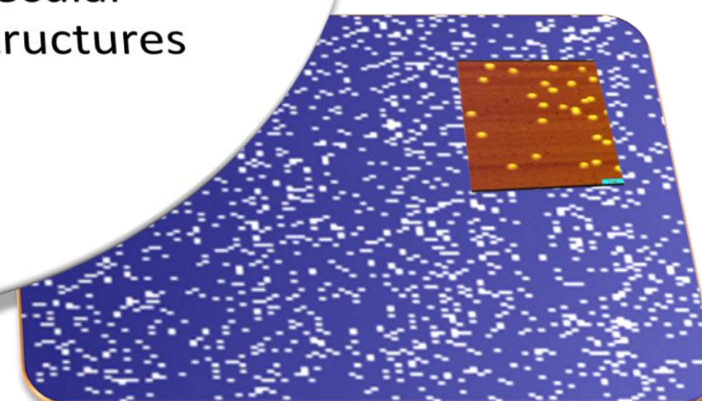
Metrology

Molecular properties and dynamics

Lithography

Molecular nanostructures

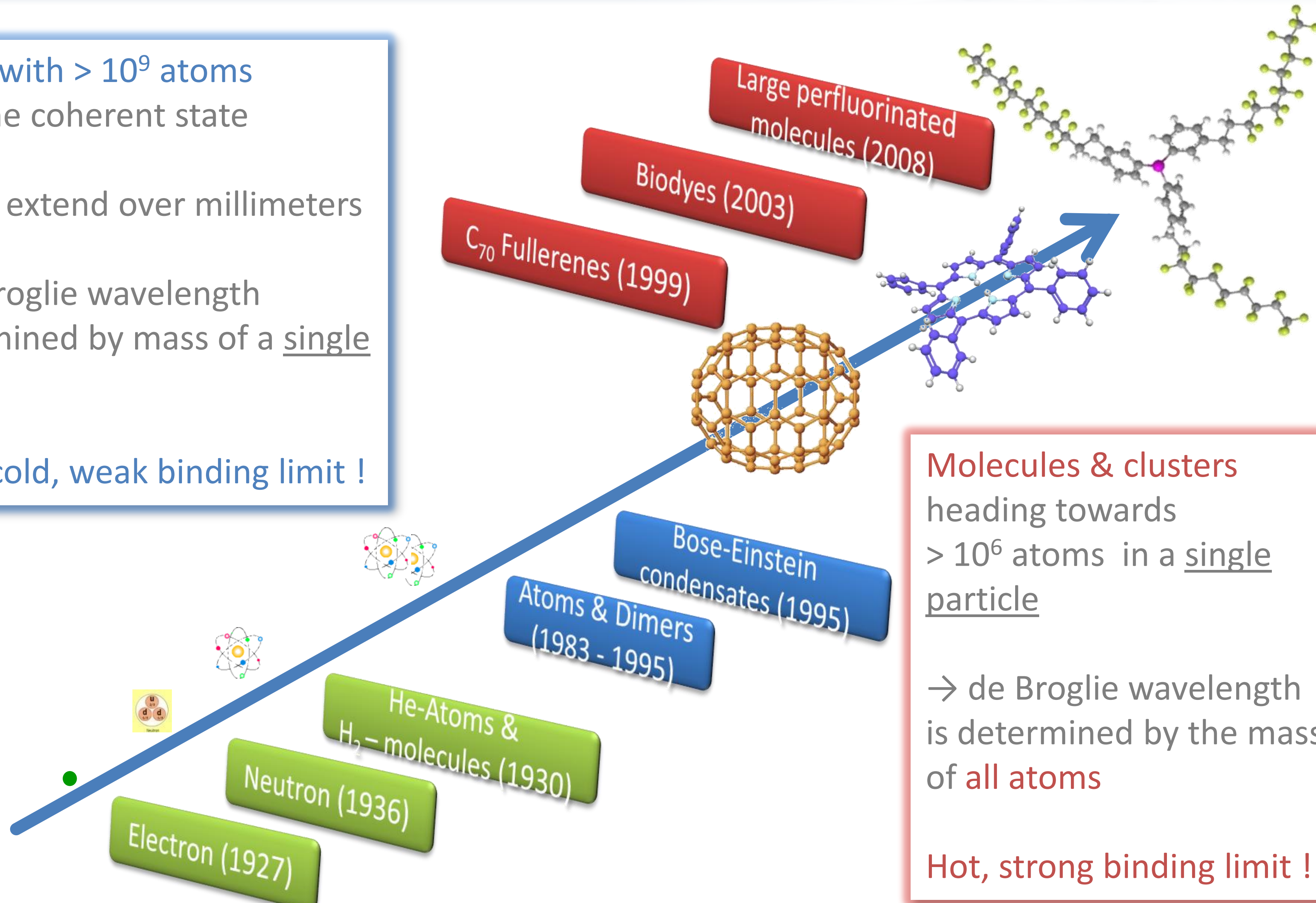




Timeline of matter wave interference

- BEC with > 10^9 atoms in one coherent state
- May extend over millimeters
- De Broglie wavelength determined by mass of a single atom.

Ultra-cold, weak binding limit !



Molecules & clusters heading towards > 10^6 atoms in a single particle

→ de Broglie wavelength is determined by the mass of all atoms

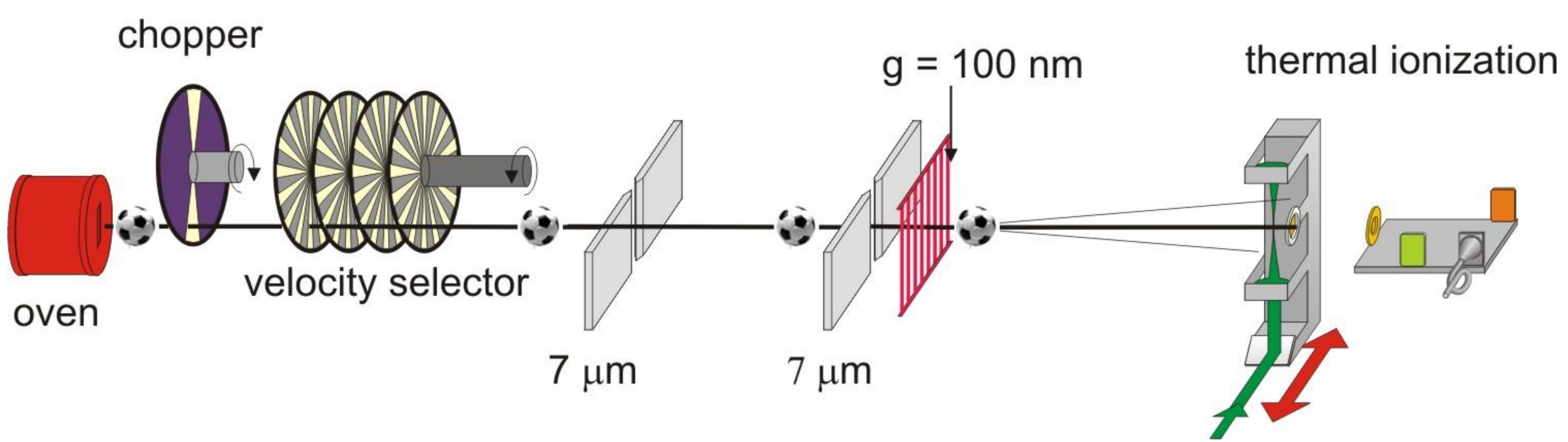
Hot, strong binding limit !

Challenges

- **Small de Broglie wavelengths (< picometer)**
- **No coherent beam sources available**
- **Low detection efficiencies**
- **Dispersive interaction between molecules and gratings**

➢ **Far-field experiments not feasible for masses >> 1000 amu !**

➢ Very small 1st order diffraction angle
➢ Long interferometer & very slow particles required
➢ Long passage time leads to strong environmental decoherence
➢ Huge signal loss due to required spatial coherence (tight velocity selection & collimation)

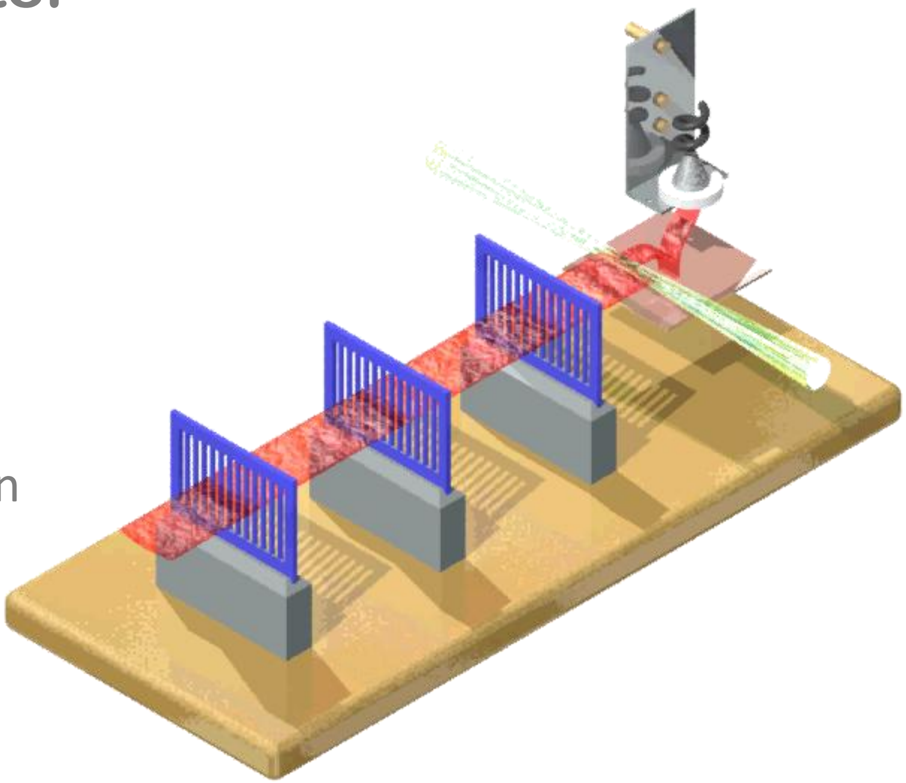


The Talbot-Lau interferometer (TLI)

- **Talbot-Lau-Interferometer**
  - 3 Gratings of equal periods, spaced equidistantly

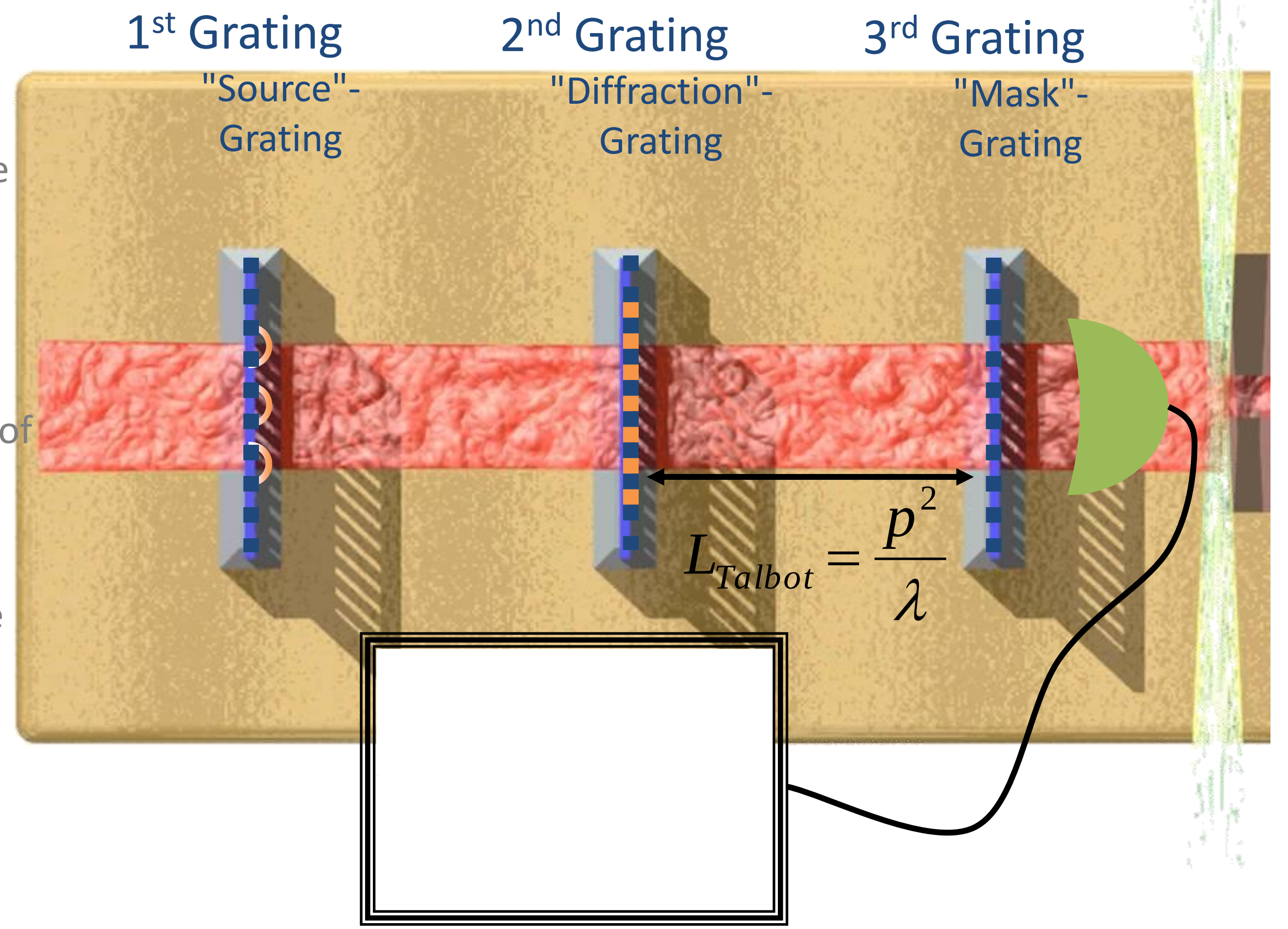
- **Advantages:**
  - Period scales with $\lambda^{1/2}$
  - Spatially incoherent illumination
  - Short (Intensity ~$1/r^2$)

  - => Allows investigation of the characteristics of complex molecules



Principle of operation

- 1^{st} grating is illuminated incoherently
- Each slit acts as coherent source
- 2^{nd} grating acts as diffraction-grating
- The transmission function is reproduced at integer multiples of the Talbot length
- If periods match, patterns originating from different source slits add (incoherently)
- 3^{rd} grating is used to scan the molecule pattern

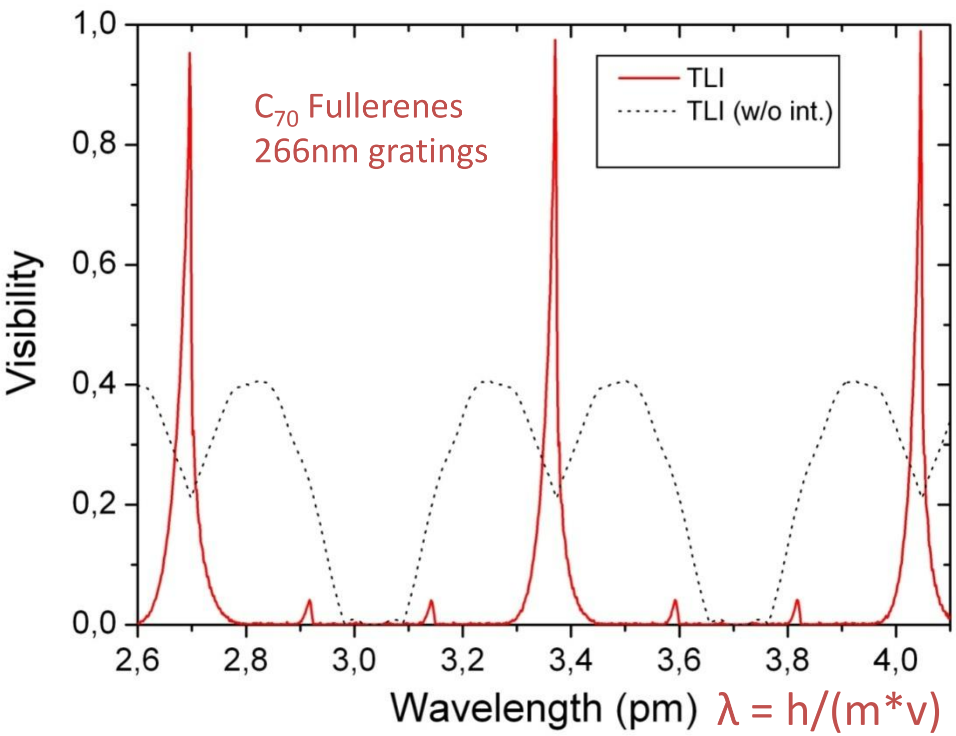


Impact of the molecule-grating interaction

Molecule-Grating interaction:

- More significant for smaller grating periods
- More significant for larger molecules (higher polarizabilities)

- **Prohibitively narrow velocity distribution required!**
- **Not feasible for molecules >>1000 amu!**



$\lambda = h/(m*v)$

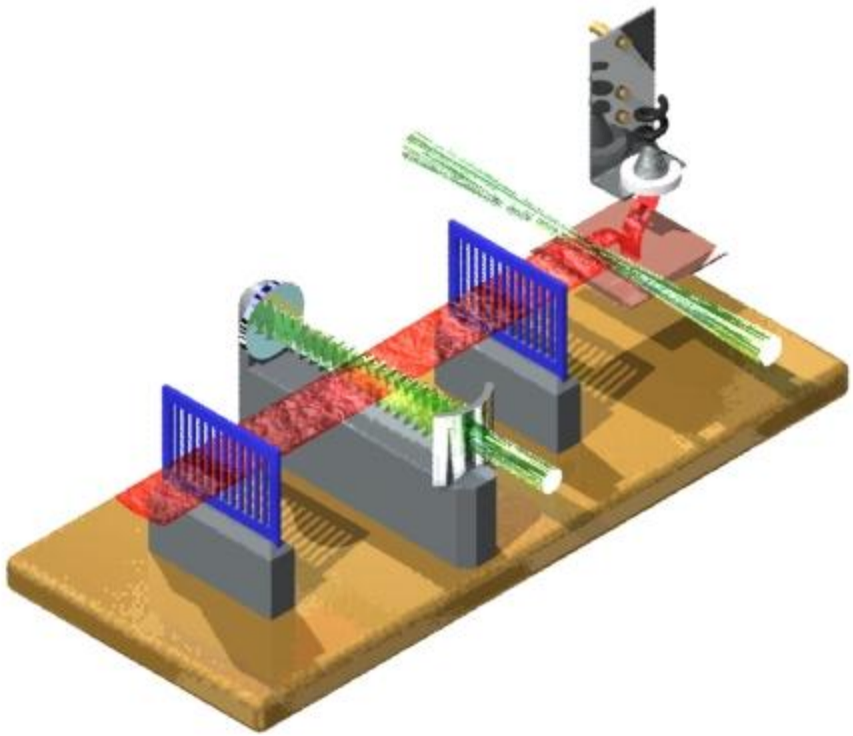
$\Delta v/v$ = 0.3% => Visibility reduced by 50%!

Kapitza-Dirac TLI

Solution:

Light Grating employed as central grating

- No van-der-Waals interaction
- Acts as pure phase grating
- Variable potential
- 100% transmission
- Cannot be blocked or destroyed

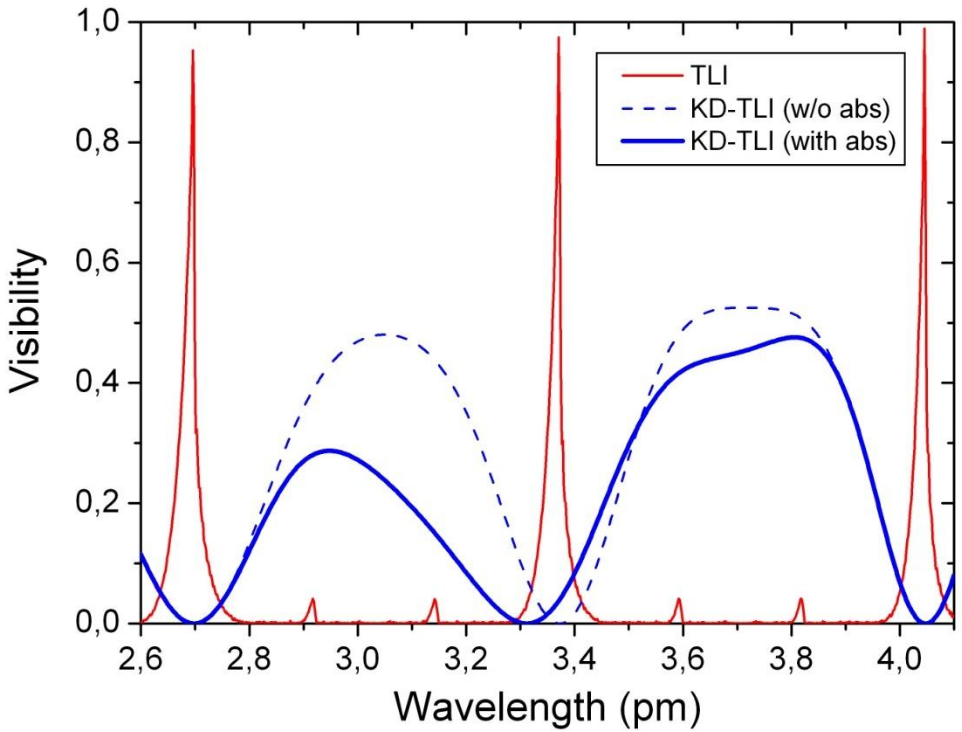


Kapitza-Dirac TLI

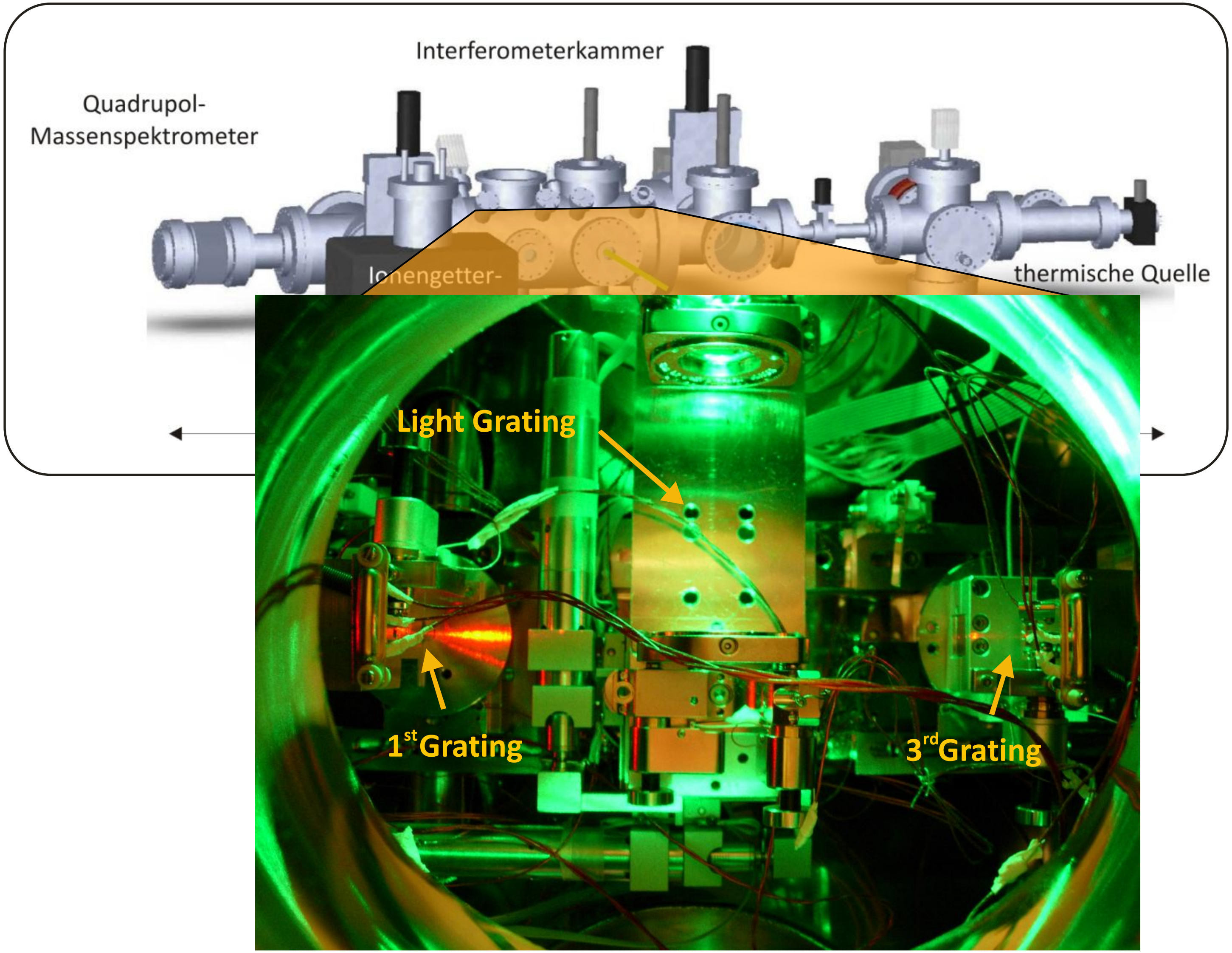
Solution:

Light Grating employed as central grating

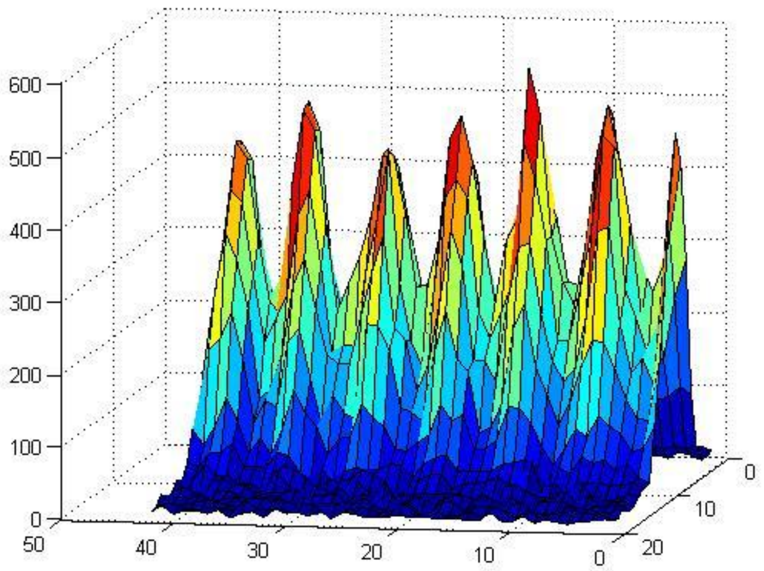
- No interaction btw. molecles and grating surfaces
- Acts as pure phase grating
- Variable potential
- 100% transmission
- Cannot be blocked or destroyed



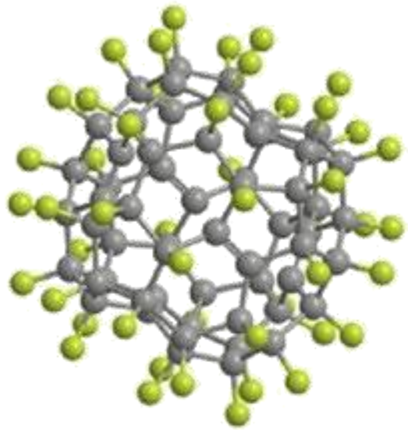
The experimental setup



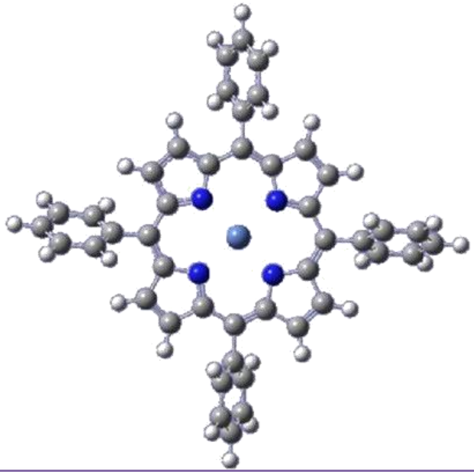
High-contrast quantum interference has been observed with…



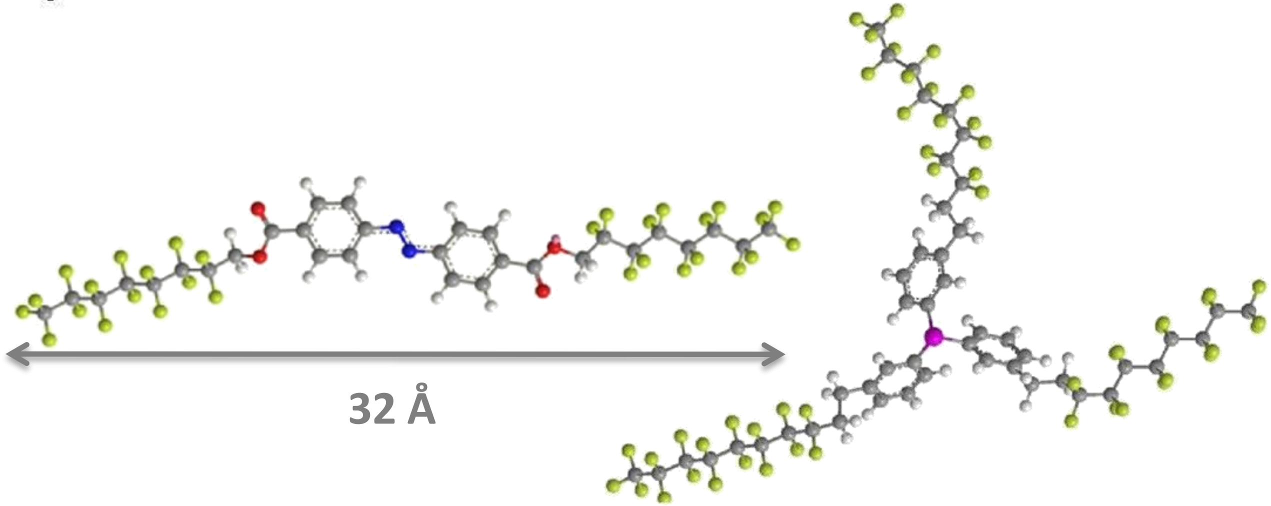
Fullerene C_{60} & C_{70}



Fluoro-Fullerene $C_{60}F_{36}$ & $C_{60}F_{48}$



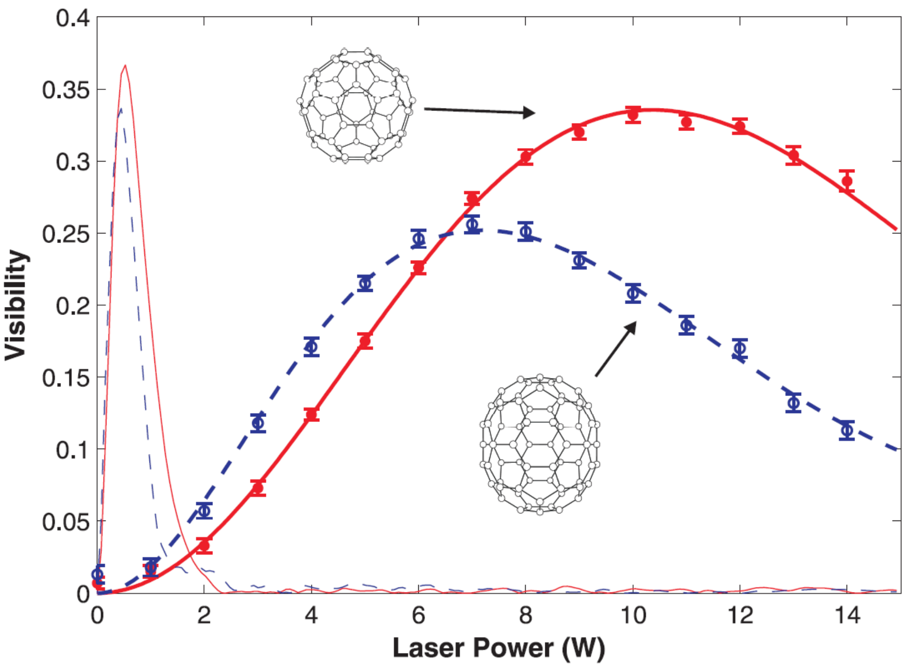
Porphyrins & derivatives



Perfluoroalkyl-functionalized molecules

Measurement of optical polarisabilities

- Perfect agreement with theory
- Different functional shapes of the visibility-power dependence
- This can be used to:
  - Measure the optical polarisabilities.
  - Indentify different molecules (including isomeres)

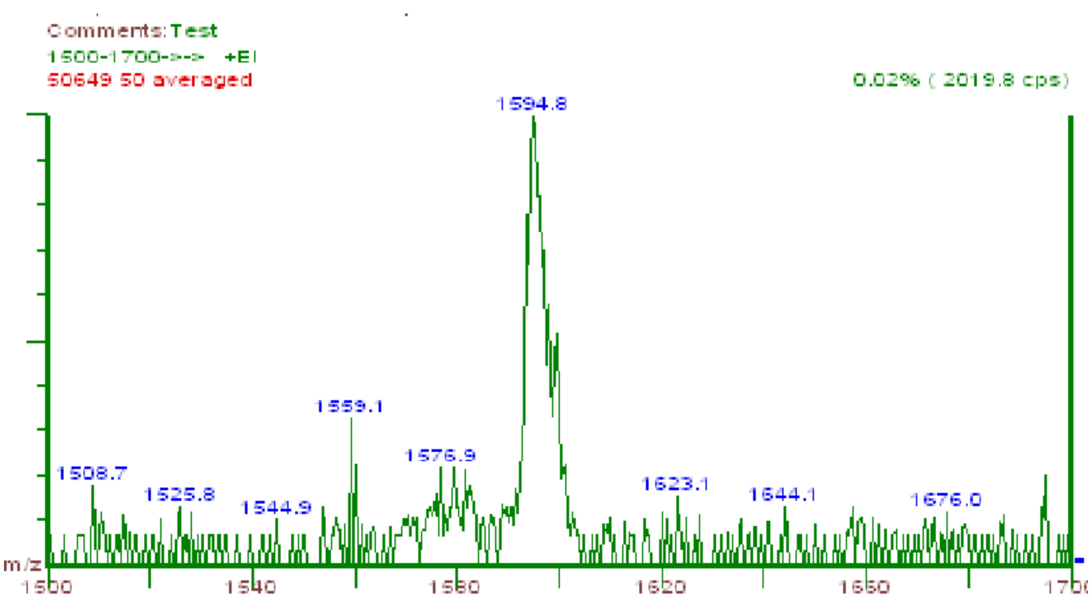


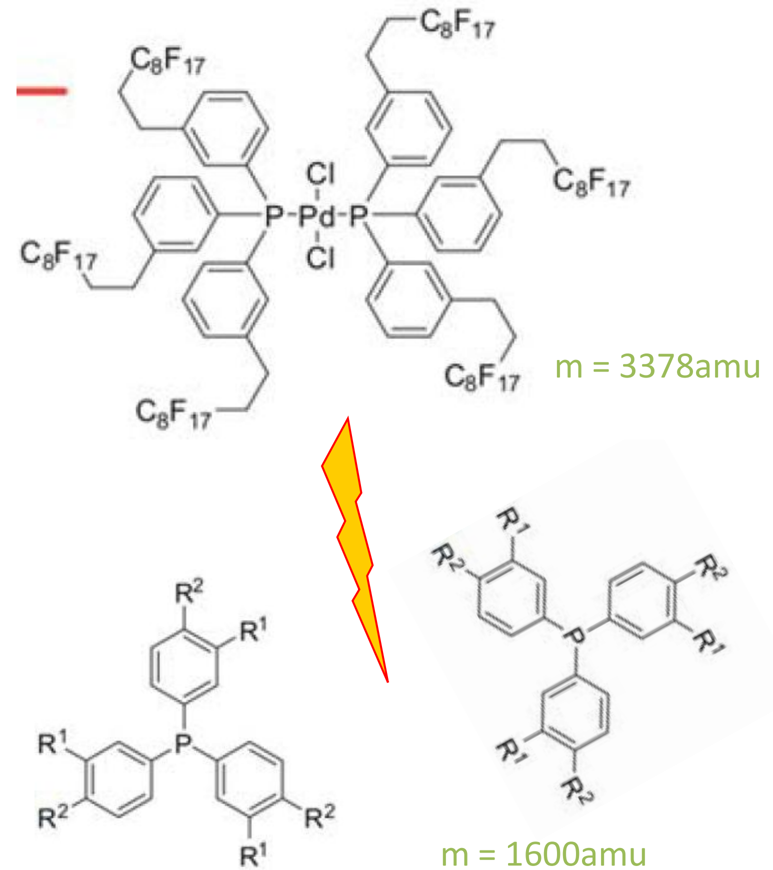
Interferometry complementing mass spectrometry

Quantum interference of the fluorinated catalyst: $C_{96}H_{48}Cl_2F_{102}P_2Pd$ (3378 amu)

Detection using EI-QMS on the fragment m = 1595 amu

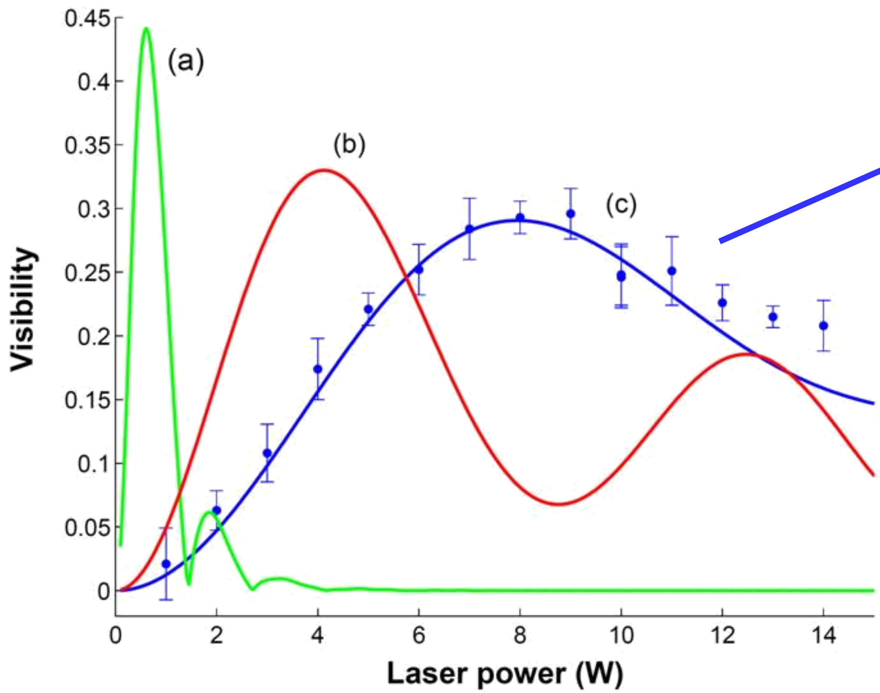
Do the molecules break in the detector or in the source **?**

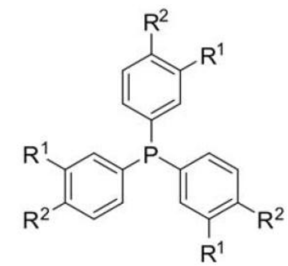




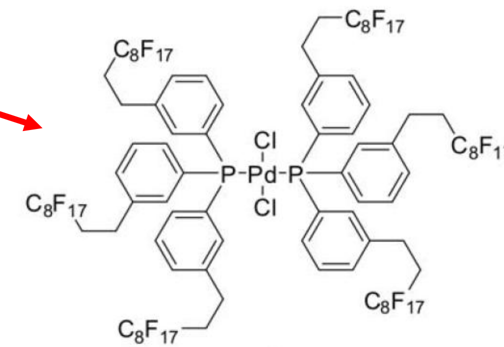
Interferometric mass spectrometry

The power dependence of the fringe visibility gives the answer!



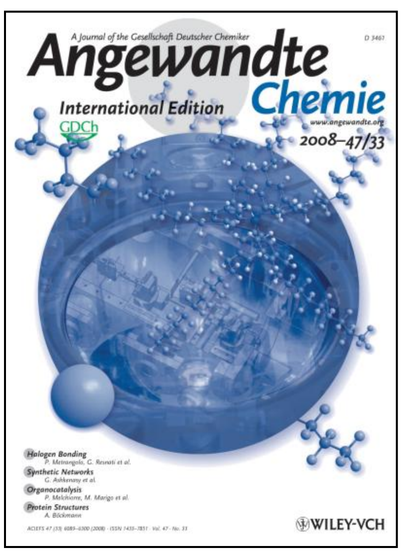


$R^1 = (CH_2)_2(CF_2)_7CF_3$; $R^2 = H$; **2**
$R^1 = H$; $R^2 = (CH_2)_2(CF_2)_7CF_3$; **3**
$C_{48}H_{24}F_{51}P$
Mwt.: 1600.60
m/z: 1600.08 (100.0%)



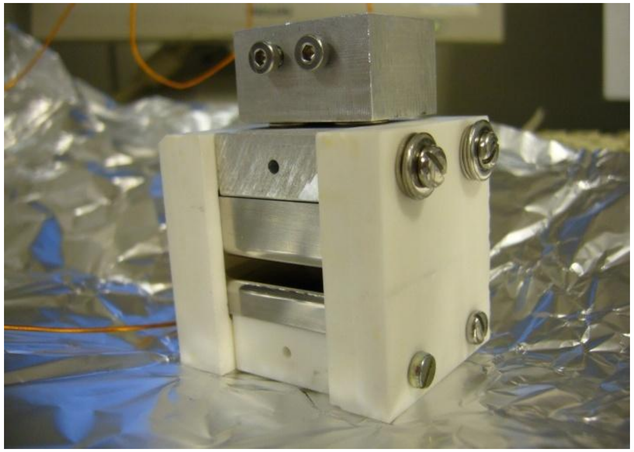
1
$C_{96}H_{48}Cl_2F_{102}P_2Pd$
Mwt.: 3378.52
m/z: 3378.00 (100.0%)

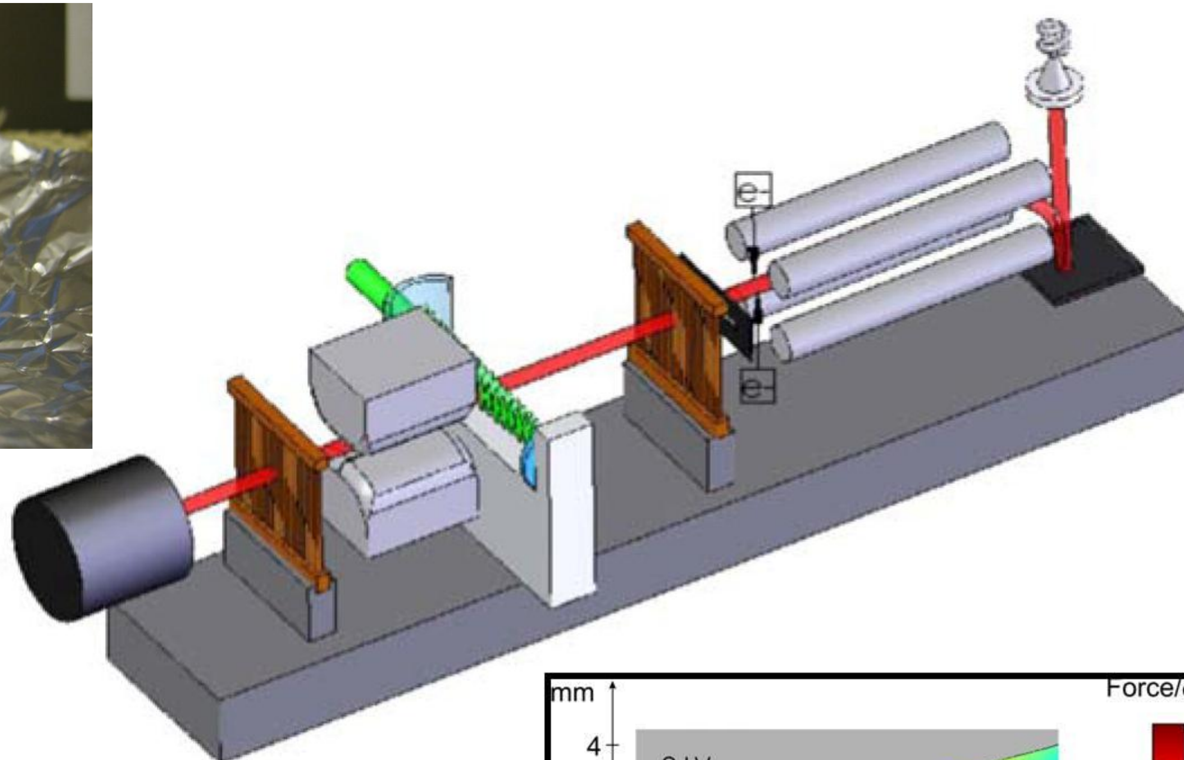
On the cover of:

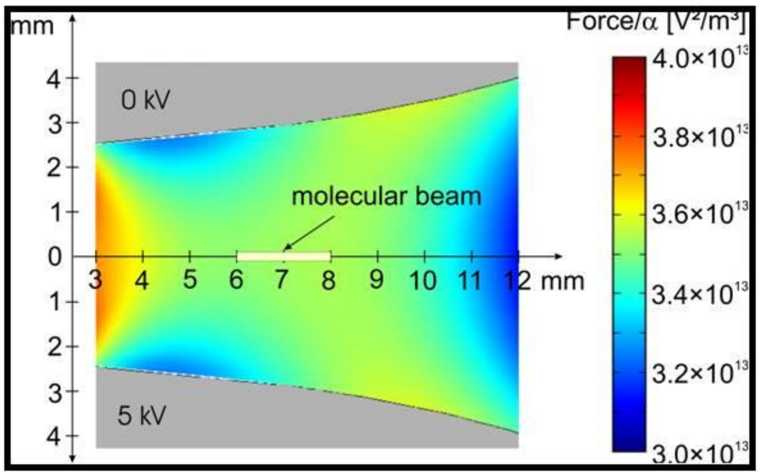


Gerlich et al., *Angew. Chem. Int. Ed.* **47**, 6195 (2008)

Measurement of susceptibilities



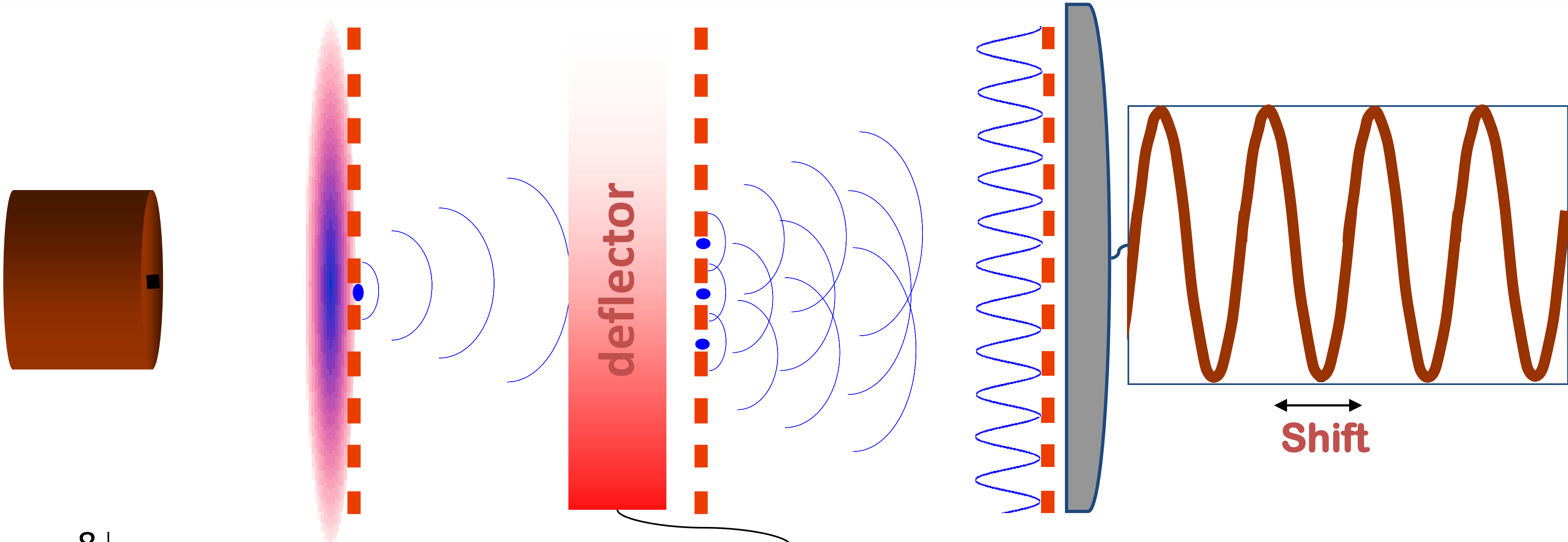


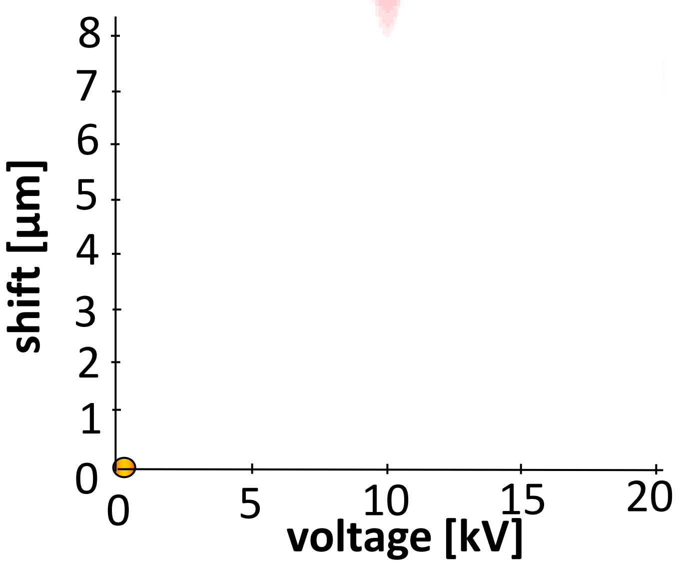


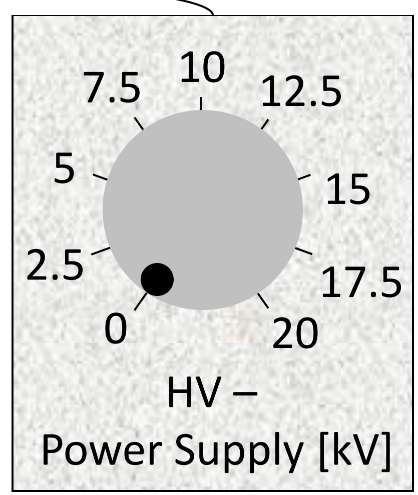
The **laser** interacts through the **optical polarizability**.

The **static field gradient** (homogeneous force field) interacts through the ***static* polarizability** & the **electric dipole moment**

Applying external electric fields







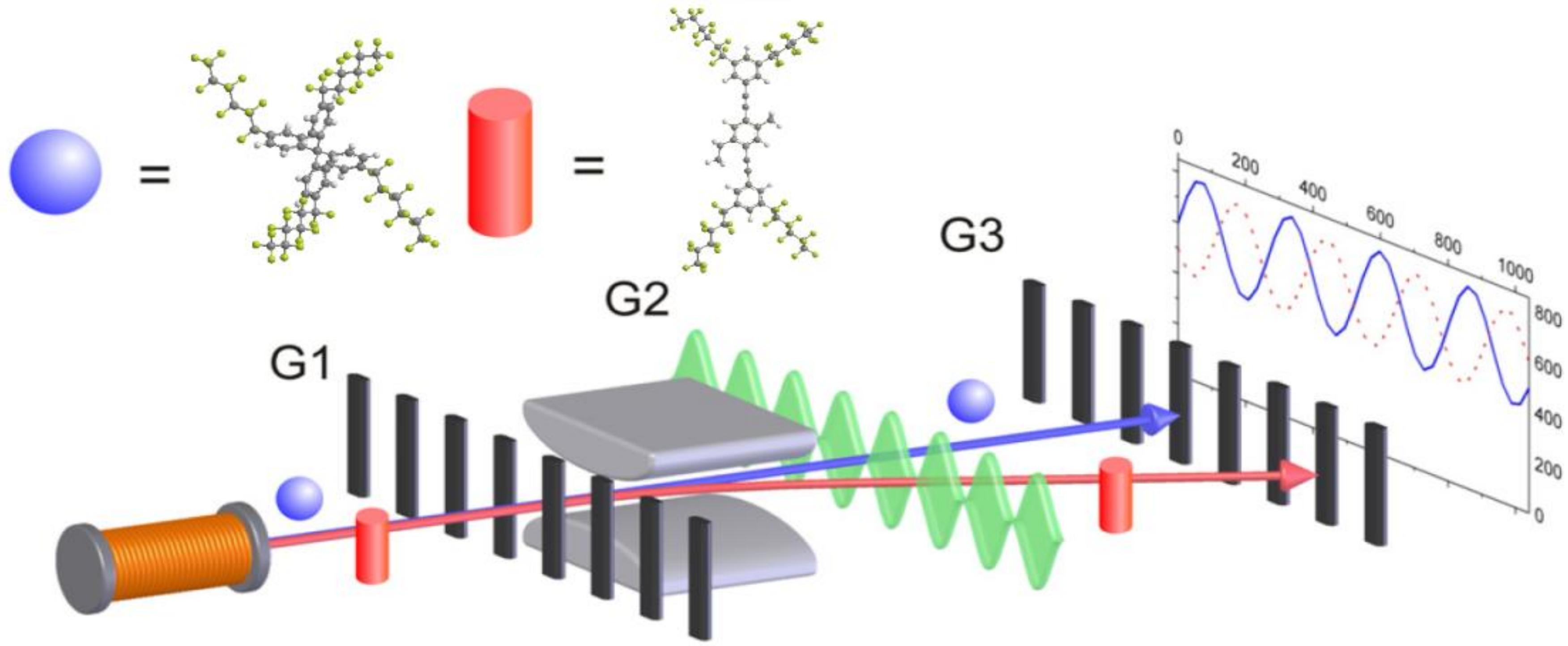
$$x_d \propto \frac{\alpha}{m} \frac{(E\nabla)E}{v^2}$$

Berninger et al., *Phys. Rev. A.* **76**, 013607 (2007).

De Broglie interference is sensitive to the molecular structure: Identification of constitutional isomers

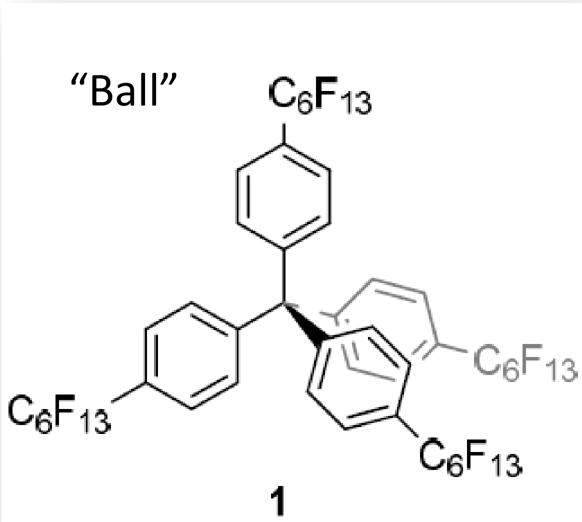
Identical chemical sum formula & mass,

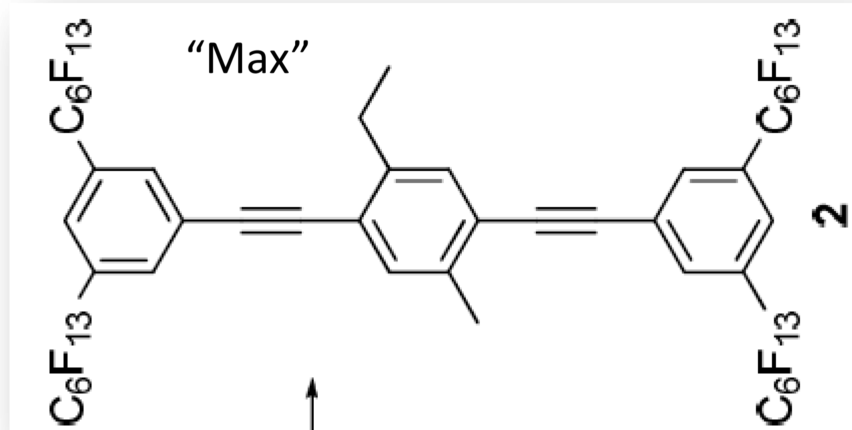
<u>but</u> different structure and different electric properties



The molecules couple to a conservative potential, without leaving any trace or position information

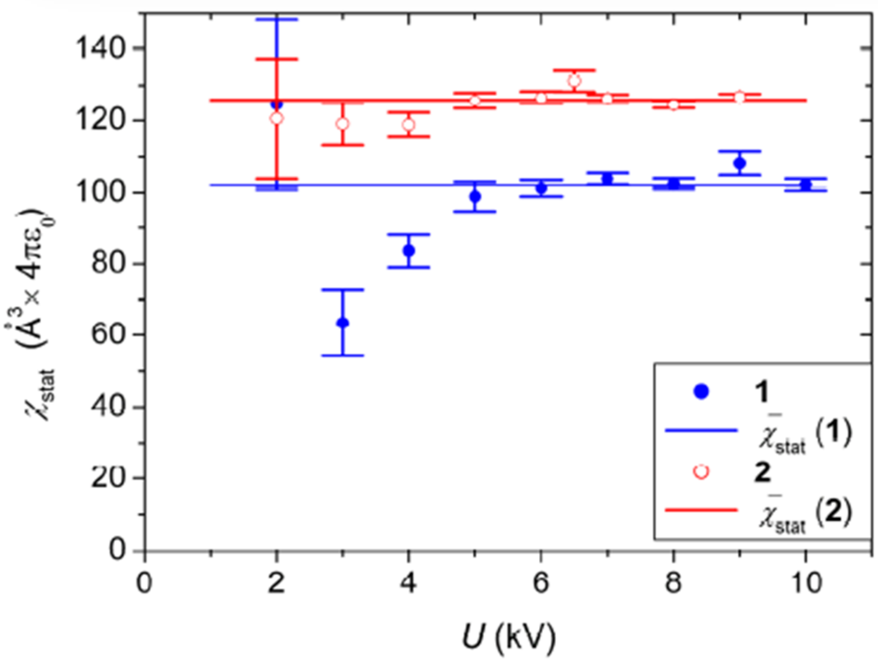
Interferometrically determined difference in susceptibility between the two isomers





$$\chi_{Ball} = 102 \pm 0.8 \text{Å}^3 \times 4\pi\varepsilon_0$$

$$\chi_{Max} = 126 \pm 0.5 \text{Å}^3 \times 4\pi\varepsilon_0$$

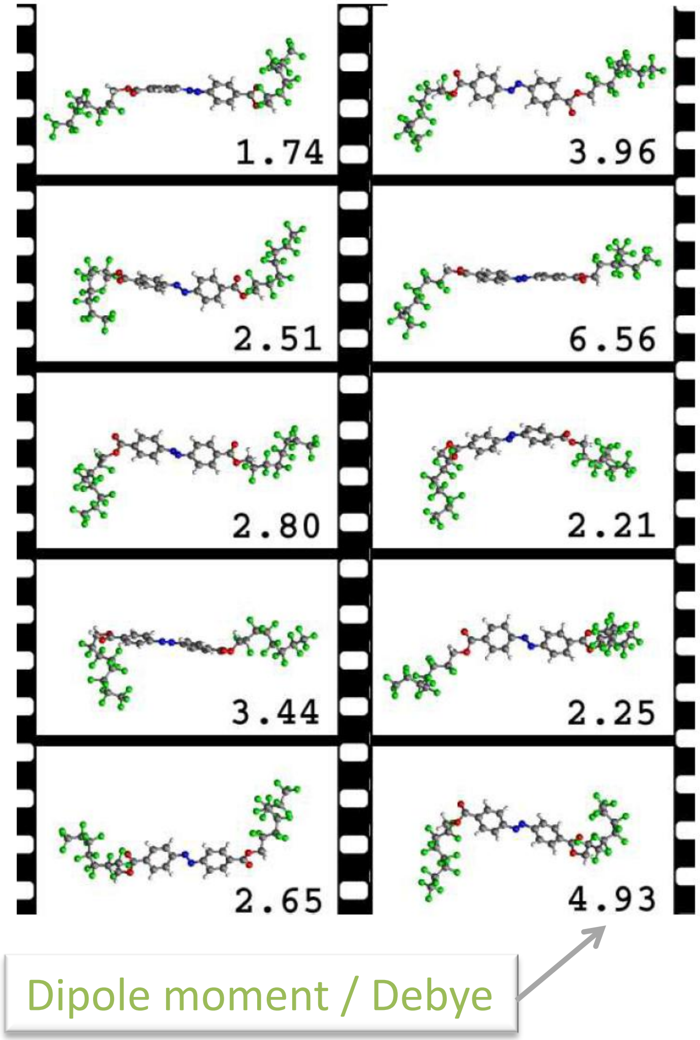


De Broglie interference is sensitive to:
Molecular dynamics & thermally activated conformation-oscillations

- Azobenzene derivative
  → no static dipole moment
- But at 500 K:
  → many different conformations

- Surprise 1:
  High-contrast de Broglie interference
- Surprise 2:
  Dynamic dipole moments μ measurable

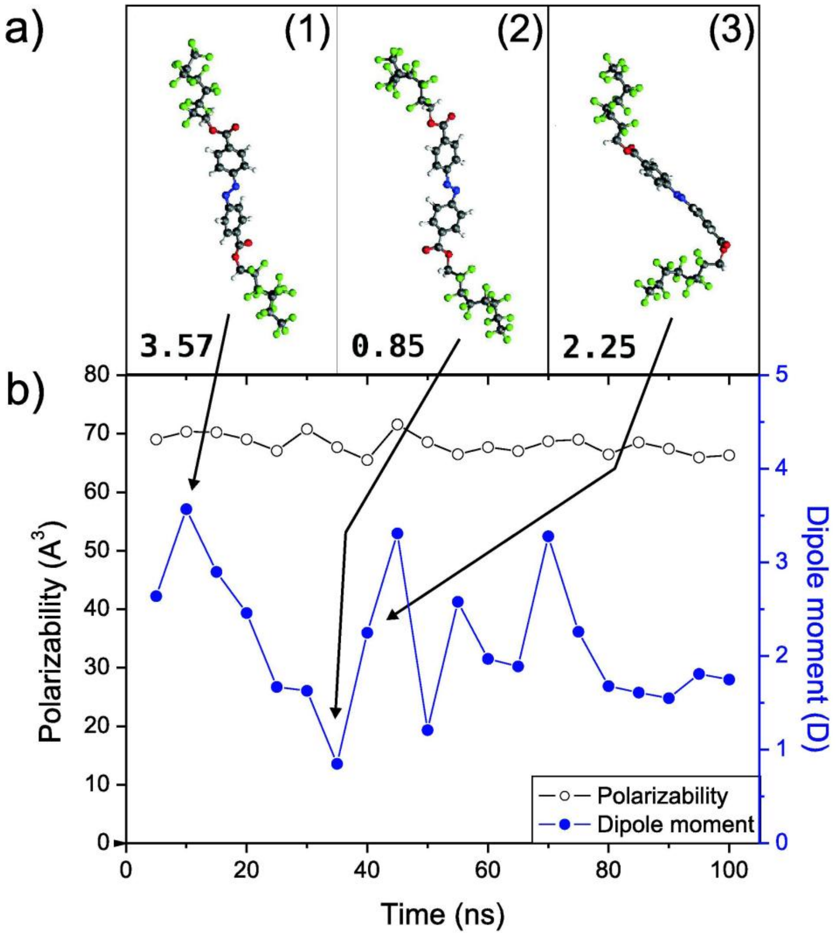
$$\chi = \alpha_{stat} + \frac{1}{3}\frac{\langle \mu_z^2 \rangle}{k_b T}$$



Phys. Rev. A , Rapid comm. (2010)

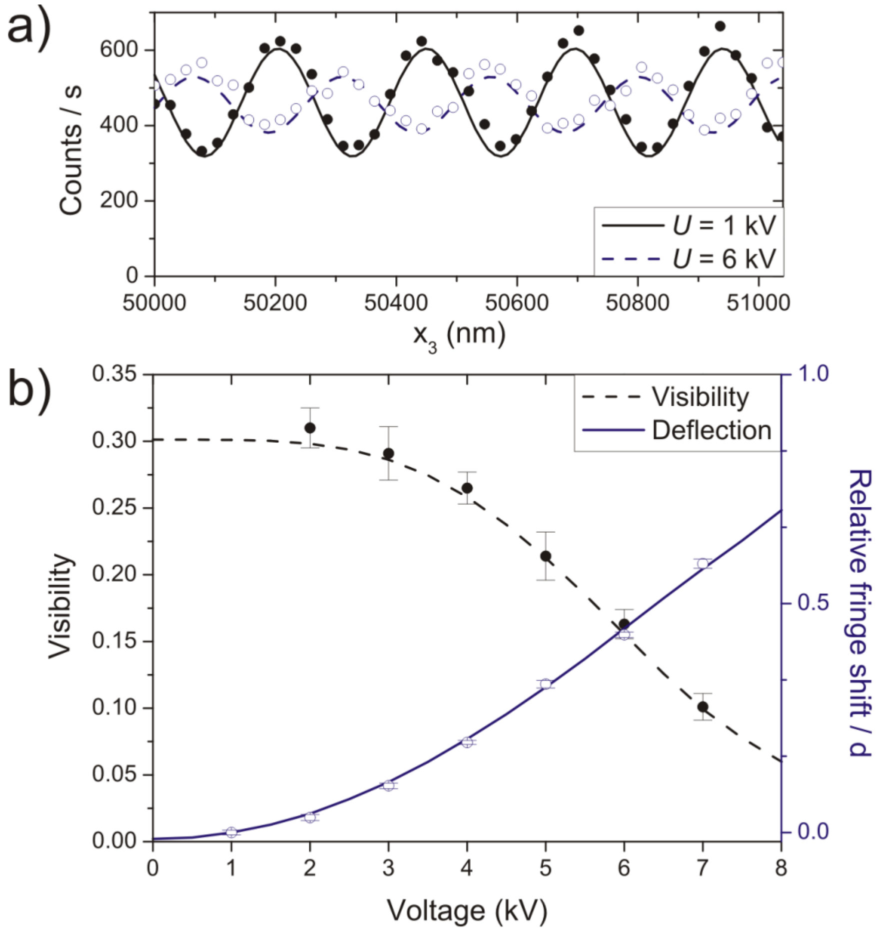
Total electric susceptibility χ reveals fluctuating dipole moments

Theory



$$\chi = 92.8 \pm 0.8 \pm 7.6\ \text{Å}^3 \times 4\pi\varepsilon_0$$

Experiment



$$\chi = 95 \pm 3 \pm 8\ \text{Å}^3 \times 4\pi\varepsilon_0$$

Limitations in the lab

- **Limitations using the time-domain TLI with grating period d=80 nm**

- Passage time through interferometer: 2T ~ 32 ns/amu
  - Gravity: fall distance ~ $mass^2$, particles > 10^6 amu fall out of focus!
  - Coriolis force: proper setup orientation ➔ no transverse deflection
  - Velocity: for 10^6 amu v < 1 m/s to stay in mirror region,
    ➔ slowing/cooling or larger mirrors needed
- Size: gold cluster radius ~ 3 nm @ 10^6 amu ➔ still small enough
- Relevant decoherence/dephasing rates @ 10^6 amu:
  - Collisions with rest gas: residual pressure < 10^{-10} mbar ➔ OK @ T=300 K
  - Thermal radiation: Rayleigh scattering & absorption/emission still OK @ T=300 K
  - Mirror stability: vibrations & thermal drifts < 20 nm over integration time

Gravity sets mass limit in the lab to 10^6 amu

Many fruitful collaborations

Interferometry

- Lucia Hackermüller → now Nottingham
- Hendrik Ulbricht → now Southampton

Molecular Synthesis & Analysis

- Marcel Mayor, Univ. Basel
- Jens Tüxen, Univ. Basel

Molecular simulations

- Nikos Doltsinis, Kings College London
- Marcus Böckmann, Univ. Bochum

Theory

- Klaus Hornberger, Dresden

New ideas on mesoscopic coherence experiments

- Herbert Gleiter, Horst Hahn, KIT Karlsruhe

The Vienna Quantum Nanophysics Team 2010

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