# Real-time single-molecule imaging of quantum interference

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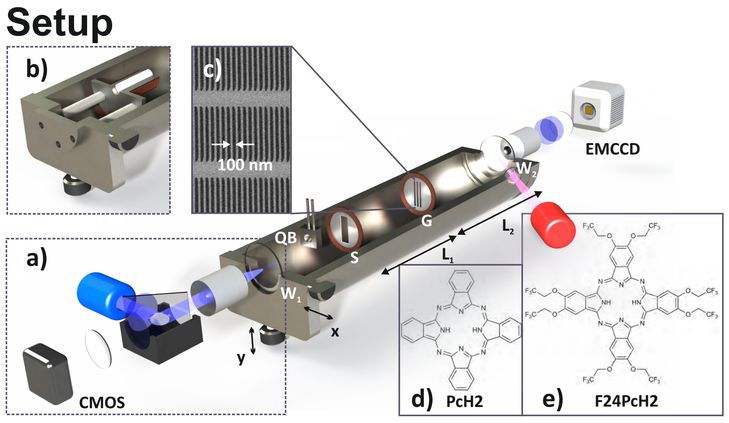
## Abstract / motivation

We demonstrate the combination of different nanotechnologies to realize the composition and imaging of quantum interference patterns from single dye molecules [2] .
The molecular beam is created using soft laser evaporation from a coated window. The molecular beam is diffracted at an ultra-thin nanomechanical grating and the resulting matter-wave diffraction pattern is collected on a quartz plate at the vacuum-air interface.
Fluorescence microscopy with single-molecule sensitivity allows us to determine their position with an accuracy of 10 nm and to record the build-up of the interference pattern in real-time.
Besides visualizing the wave-particle duality in a particularly intuitive way for massive organic compounds, the experiments now enable studies of the van der Waals interaction between complex molecules and the grating bars in a regime, where the transit time of the molecules through the diffractive element becomes comparable to their rotation period. Our experiments aim at a quantitative understanding of these potentials for gratings as thin as graphene, as well as at an extension to two-dimensional diffraction structures in preparation for molecular holography.


## Setup




Phthalocyanine derivatives

- **Molecular beam source:**
  Laser micro-evaporation from a dye coated window mounted onto a x/y-stage
  → Short heating times: minimally invasive, acting only on a restricted set of molecules
  → Small emission area -> ensures some degree of transverse coherence
- **Diffraction elements:**
  $SiN_x$ or carbon nanomasks, period d=100nm, slit width s=50..75 nm, thickness t = 10...200 nm.
- **Coherence preparation:**
  → Longitudinal coherence: gravitational velocity selection v=100..350 m/s, $\lambda_{dB}$ = 2..7 pm
  → Transversal coherence: laser microevaporation and/or collimation slit (collimation to 2 µrad!).
- **Detection:** Fluorescence microscopy with single molecule sensitivity
- **Avoiding decoherence:** The experiments are performed under high vacuum conditions ( ~ 5 × $10^{-8}$ mbar).

## Fluorescence microscopy

**Optical properties**
→ Excitation: 661 nm diode laser, 5.. 50 W/cm². The excitation light is steered not to enter the detection system.
→ Fluorescence: at ~700 - 720 nm, filtered & detected by a cooled EMCCD camera.

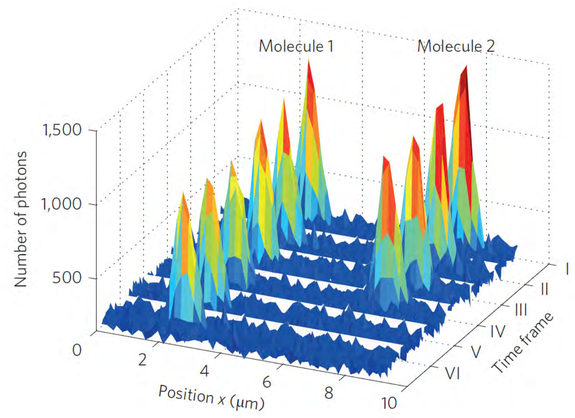
**Sample size:** We image an area of (400 µm)² with single-molecule sensitivity within a few seconds.

**Bleaching:** Phthalocyanines emit about $10^5$ detected photons before they bleach or redesorb.

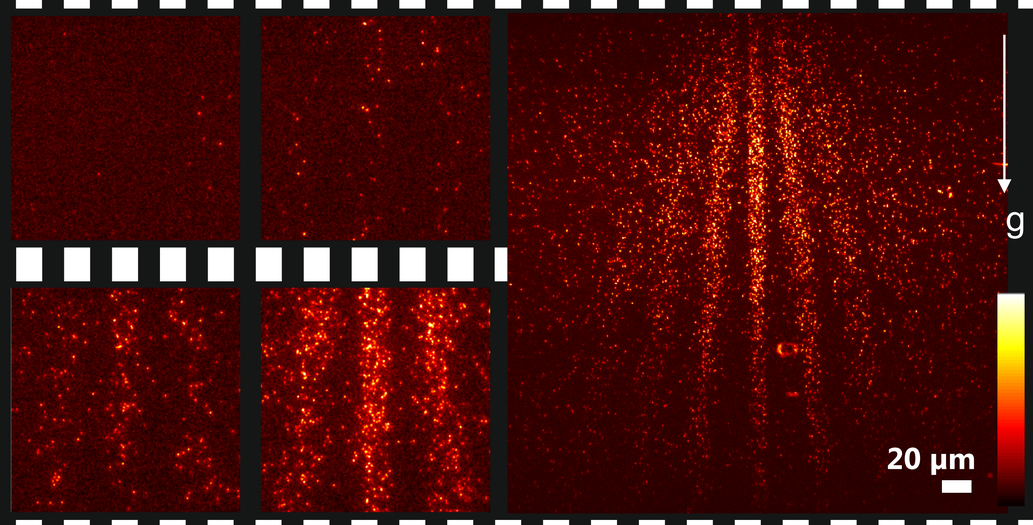
**Position accuracy:** A high signal-to-noise ratio allows determining the position of each molecule with an accuracy of about 10 nm.

**Sample Cleaning:** In situ plasma cleaning allows reusing the quartz screen hundreds of times.

**Figure (right panel):** Two phthalocyanine molecules are monitored over 6 subsequent time frames.
The stepwise bleaching of molecule 2 indicates that we image single emitters and not aggregates of them.



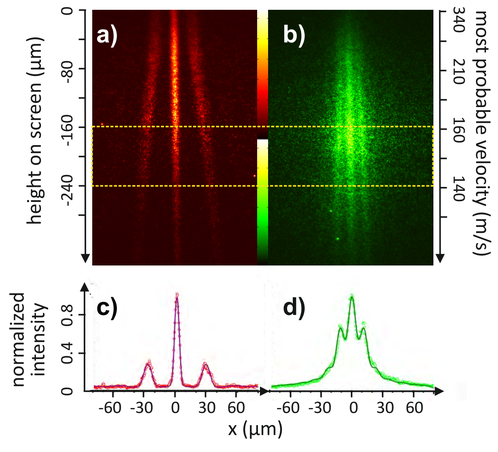

Watch the video on youtube!




Selected frames of the video show the gradual build-up of a far-field diffraction pattern from single phthalocyanine molecules.

Slower molecules land further down on the detection screen. Since their de Broglie wavelength is larger, the separation of the interference peaks also gets larger.

The recording of this movie took about 90 min. With the molecular beam source at full power we can get a comparable result within less than one minute.




**Far-field diffraction of larger molecules** [2,4]

- Phthalocyanine PcH2 (red):
  Highly fluorescent dye, thermally stable
- Perfluoro-alkylated phthalocyanines (green):
  Higher masses, high volatility,
  optical properties similar to PcH2.

**Future:**
- Tagging of larger molecules with dyes.
- Far-field diffraction at optical gratings

**Van der Waals interaction between the molecules and the grating** [2,6].

**Attractive vdW potential**
→ phase term in grating transmission function
→ enhances higher order interference fringes

**Experimental preconditions**
→ improved longitudinal coherence
→ homogeneous excitation of the chromophores

**Vary:**
velocity, grating thickness, slit width, material,...

**Theoretical description**
A new theoretical description in cooperation with Stefan Scheel and Stefan Y. Buhmann shall incorporate the approach to and the departure from the grating.
This might be essential for gratings as thin as 5-10nm.

## Outlook / References

**Diffraction of organic molecules at porous nanostructures.**

**Motivation:**

Like in neutron diffraction, it should be possible to get information about diffraction masks by analyzing the interference pattern in the far-field.

Unlike neutrons, molecules interact more strongly with the transmission mask, which can lead to additional information on the object.

Unlike electrons, molecules have a kinetic energy orders of magnitude smaller at comparable deBroglie wavelengths.

We want to study, whether these features of molecular matter-waves can be exploited in a molecular coherent diffractive imaging scheme.


[1] Arndt et al., Nature 401, 680- 682 (1999).
[2] Juffmann et al., Nature Nanotechnology 7, 297–300 (2012).




[3] Juffmann et al., PRL 103, 263601 (2009)
[4] Juffmann et al., Found. Phys. 42, 98-110 (2012).
[5] Hornberger et al., Rev. Mod. Phys. 84, 157-173 (2012).
[6] Grisenti et al, PRL 83, 1755-1758 (1999).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