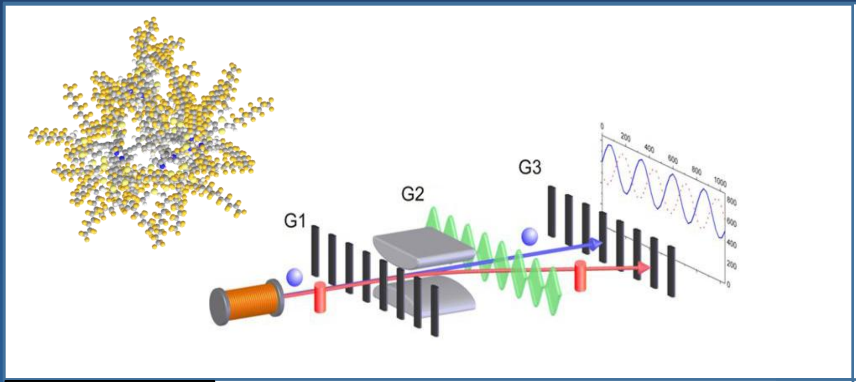
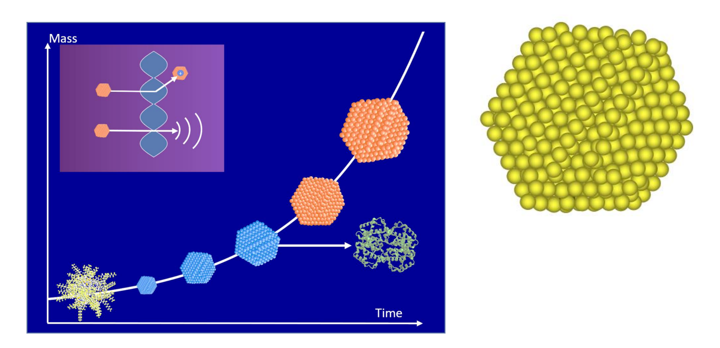
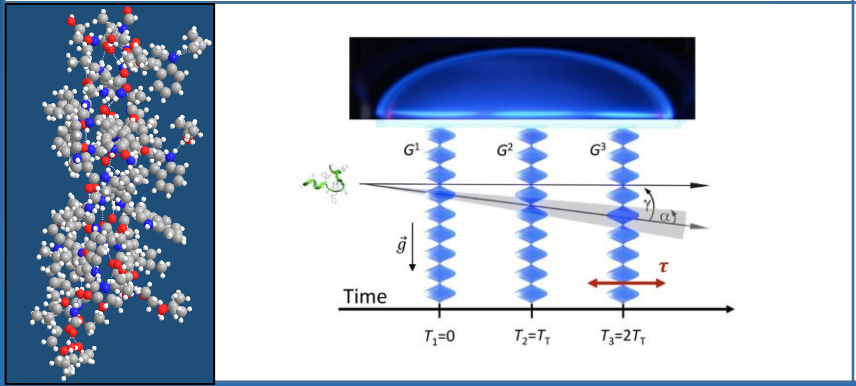
Universal matter-wave interferometry across the mass & complexity scales







Markus Arndt
Quantum Nanophysics Group
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Team & Funding

FWF

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ONR
Science & Technology

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International Partners

Matter-Waves
Klaus Hornberger, Univ. Duisburg-Essen
Christian Brand, Univ. of Ulm
Ben Stickler, Univ. of Ulm

Cluster Advise
Bernd von Issendorff, Univ. Freiburg
Vitaly Kresin, USC Los Angeles

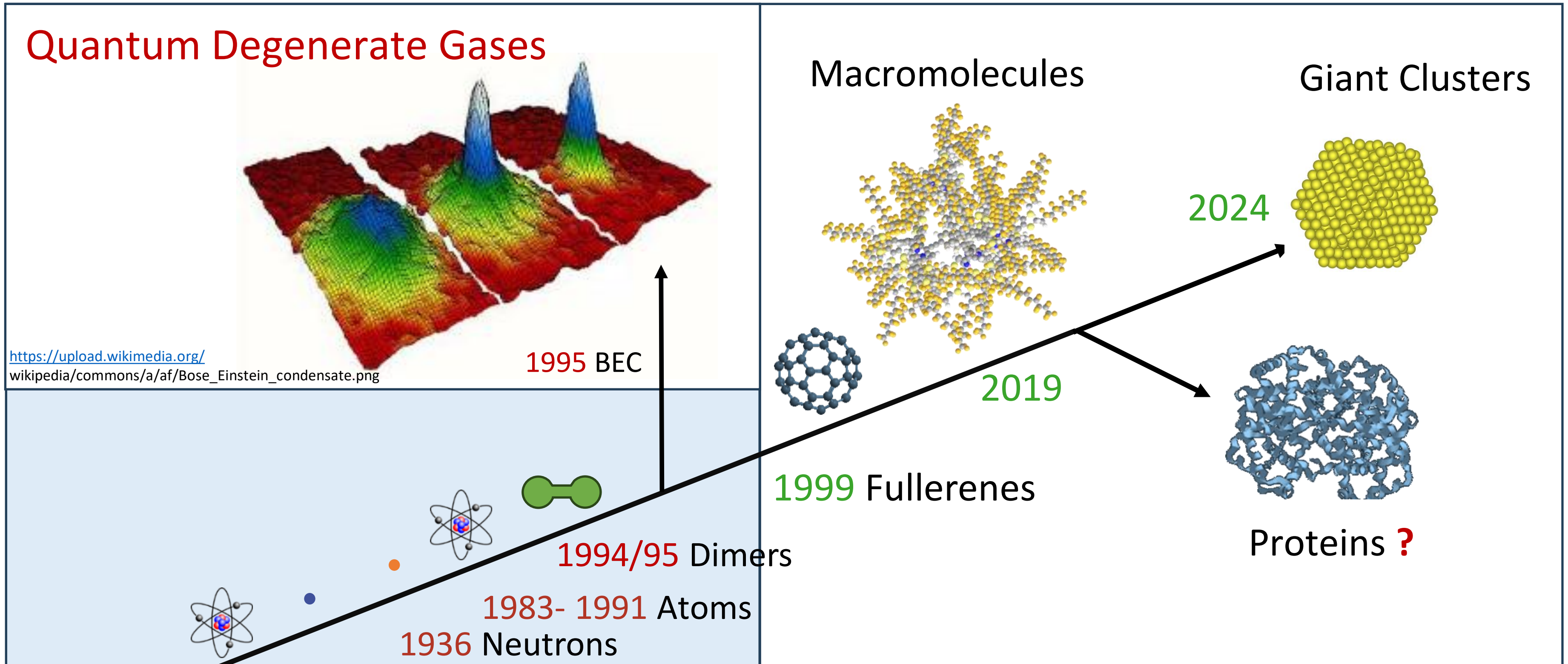
Nanofabrication
Anders Marlow, Univ. Melbourne

Superconducting Detectors
R. Gourgues, M. Castaneda, A. Fognini
Single Quantum, Delft

Synthetic Chemistry
Y. Hua, V. Köhler & M. Mayor, Univ. Basel

Ion Traps, Nanoparticles
Tracy Northup, Univ. Innsbruck
Roland Wester, Univ. Innsbruck
Uros Delic, Univ. Vienna

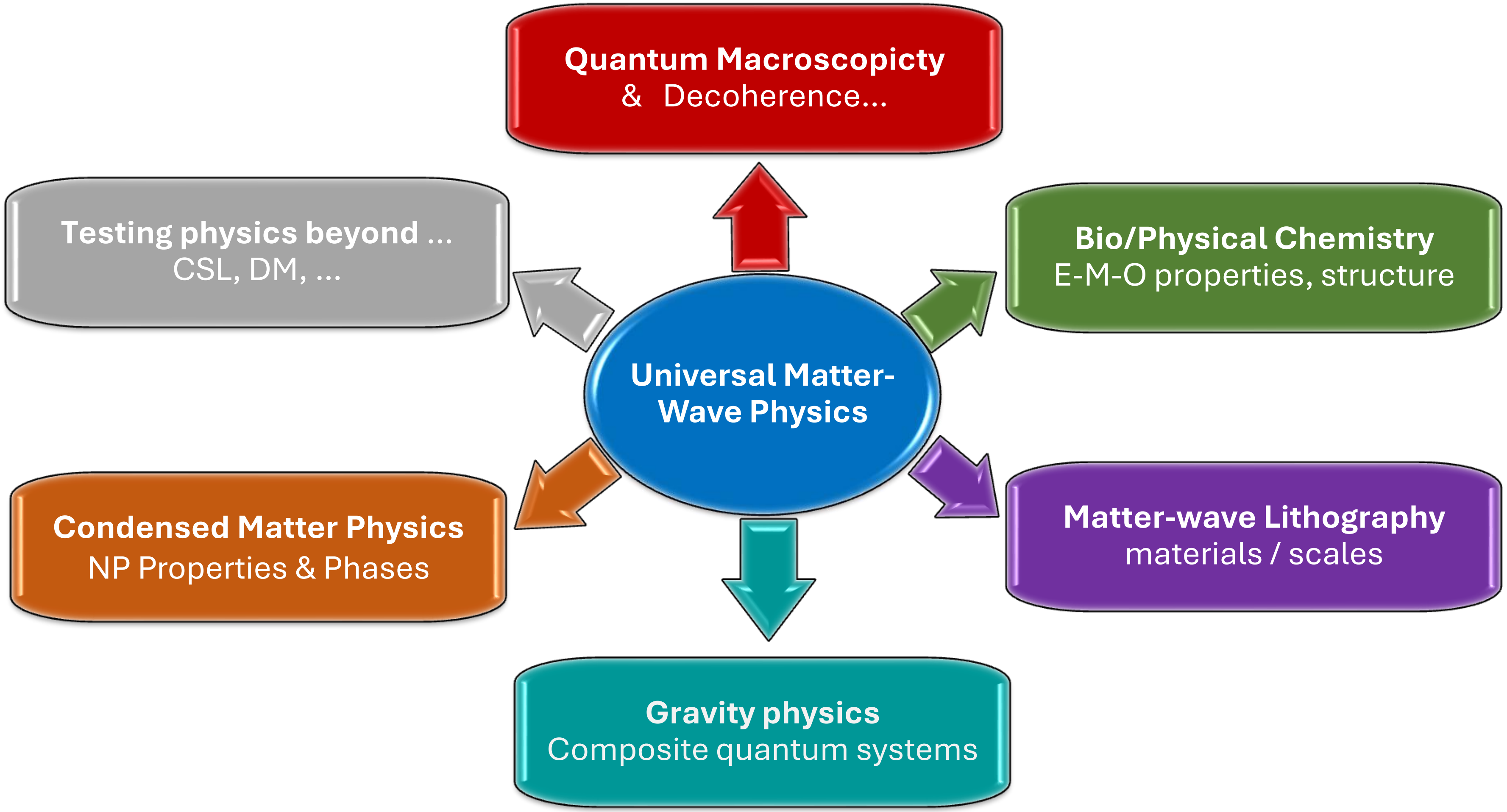
Quantum waves across mass & complexity scales



https://upload.wikimedia.org/wikipedia/commons/a/af/Bose_Einstein_condensate.png

- N atoms : $10^2 \rightarrow 10^6$?
- Warm/ Hot : 4 K .. 1000 K
- Binding : 1..10 eV
- λ_{dB} : 20 fm – 10 pm
- Delocalized : D > 0.1- 0.5 µm

Why Universal Matter-Wave Interferometry ?



Universal Matter-Wave Physics

Quantum Macroscopicty
& Decoherence...

Testing physics beyond ...
CSL, DM, ...

Bio/Physical Chemistry
E-M-O properties, structure

Condensed Matter Physics
NP Properties & Phases

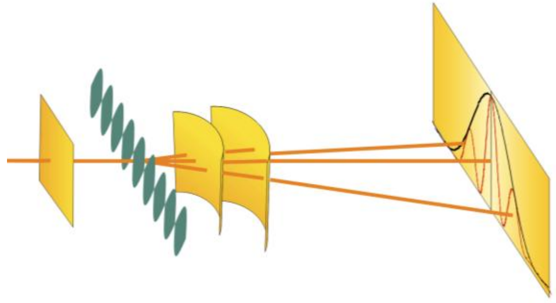
Matter-wave Lithography
materials / scales

Gravity physics
Composite quantum systems

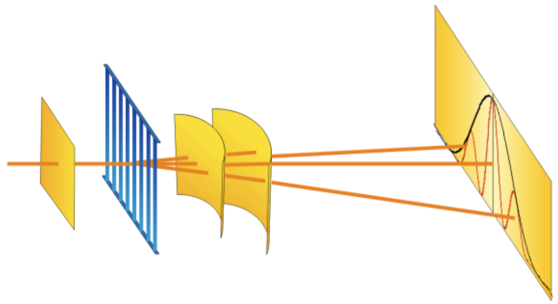
Beam Splitter Technologies

Pioneering experiments in modern atom interferometry

Wavefront Division

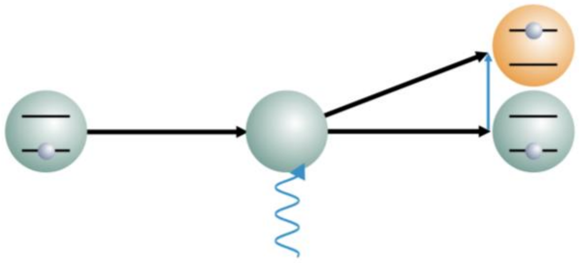


Ph. E. Moskowitz, Ph. L. Gould, S. R. Atlas, and **D. E. Pritchard**
Diffraction of an atomic beam by standing-wave radiation
Phys. Rev. Lett. 51, 370 - 373 (1983).

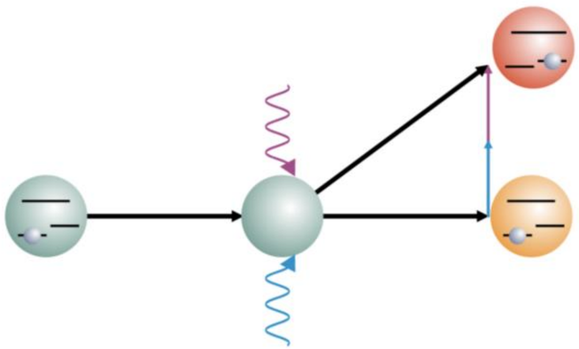


D. W. Keith, M. L. Schattenburg, Henry I. Smith, and **D. E. Pritchard**
Diffraction of atoms by a transmission grating
Phys. Rev. Lett. 61, 1580-1583 (1988).

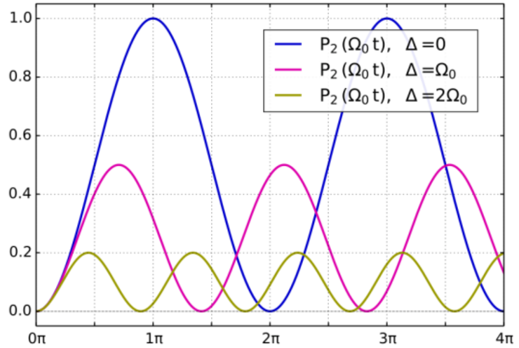
Amplitude Division



Ch. J. Bordé, *Atomic interferometry with internal state labelling*
Phys. Lett. A 140, 10-12 (1989) & J. Phys. Coll. 42, C8 (1981).



M. Kasevich and **S. Chu**
Atomic interferometry using stimulated raman transitions
Phys. Rev. Letters 67, 181 (1991).



50/50-splitter:

$$\int_0^\tau \frac{dE}{\hbar}\,dt = \frac{\pi}{2}$$

Superposition & Entanglement:

$$|\psi\rangle = |1\rangle|p_0\rangle + e^{i\phi}|2\rangle|p_0 + \hbar k\rangle$$

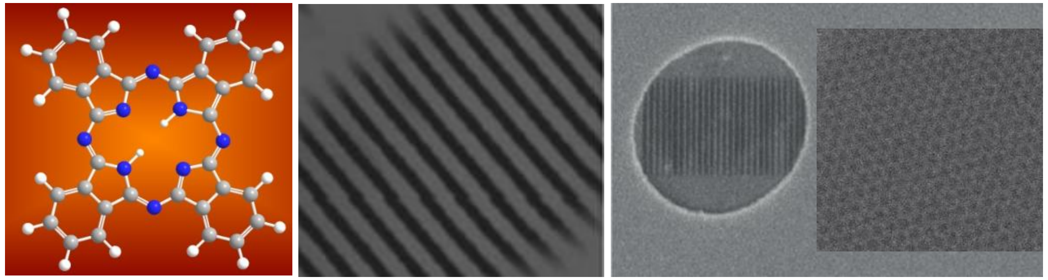
Works for

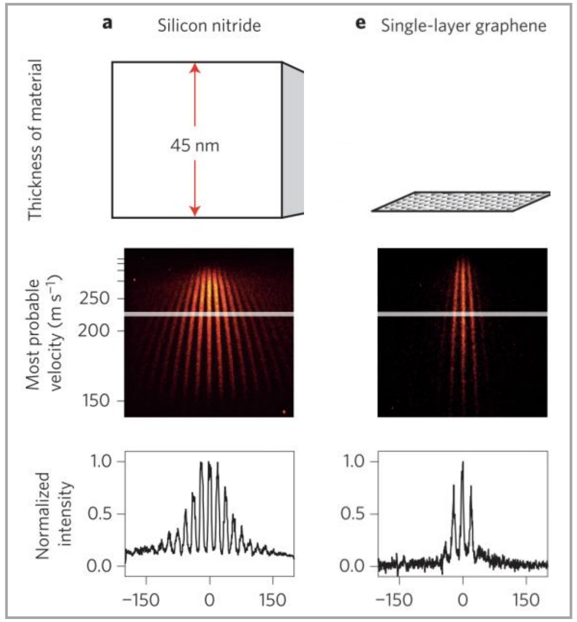
- 1-photon absorption
- 2-photon Raman transition

Nanomechanical beam splitting
Provides information about molecules

1. van der Waals forces

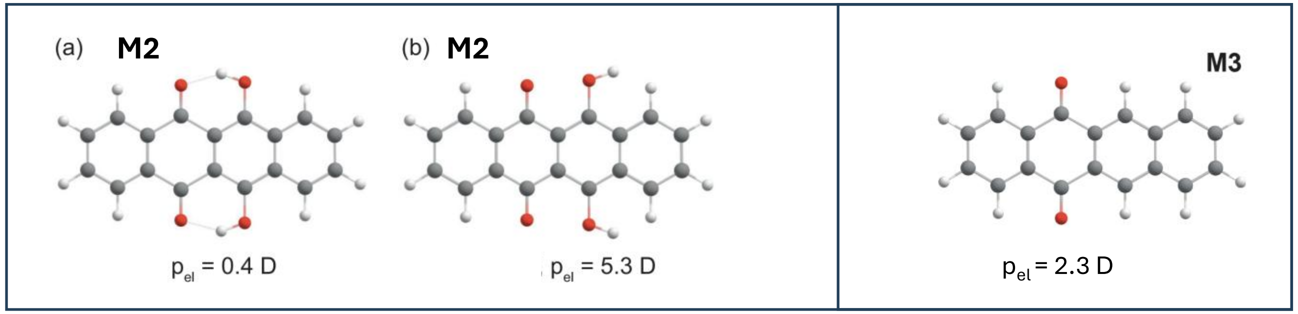
- The surface wall attraction causes a position and velocity dependent matter-wave phase shift because of residual charges in

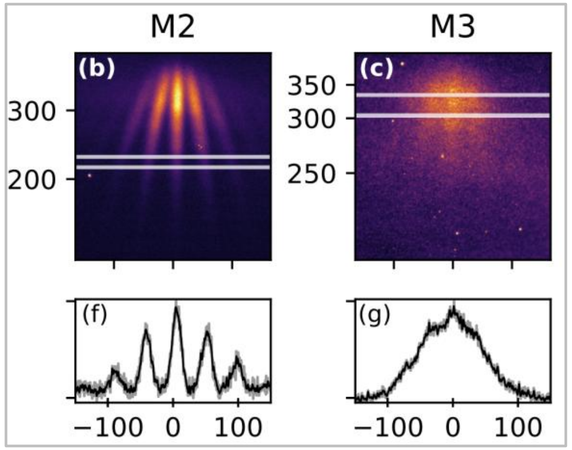




2. Electric dipole moments

- d_e causes dephasing with random charges in the grating
- Neon FIB deposits less charges than Ga FIB → higher contrast





$P_{el} \simeq 0$ D

$P_{el} \simeq 2.3$ D

M. Arndt, O. Nairz, J. Voss-Andreae, C. Keller, G. van der Zouw, A. Zeilinger, **Nature 401, 680-682 (1999).**

C. Brand, M. Sclafani, C. Knobloch, Y. Lilach, T. Juffmann, J. Kotakoski, C. Mangler, A. Winter, A. Turchanin, J. Meyer, O. Cheshnovsky, M. Arndt, **Nature Nano. 10, 845 (2015)**

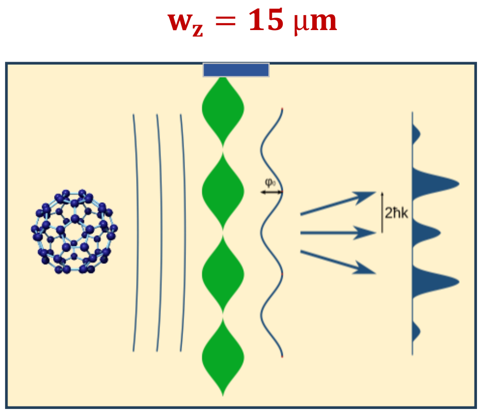
K. Simonović, R. Ferstl, A. Barlow, A. Shayeghi, C. Brand, and M. Arndt**, Phys. Rev. Res. 6, 033109 (2024).**

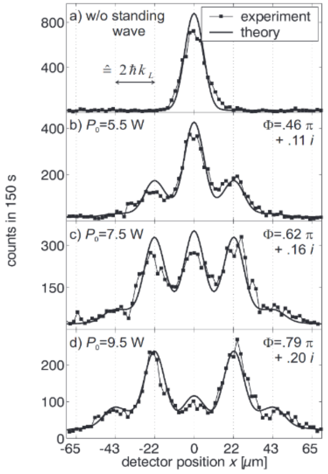
Optical Dipole Phase Grating → Universal !
Independent of internal molecular states

Kapitza-Dirac:

$$\varphi(x) = \frac{\alpha}{v_z \hbar} \int \frac{I(x,z)}{2c\varepsilon_0} dz \;=\; \sqrt{\frac{8}{\pi}} \cdot \frac{\alpha\, P}{\hbar \varepsilon_0 c\, w_y v_z} \cos^2\left(\frac{2\pi x}{\lambda}\right)$$

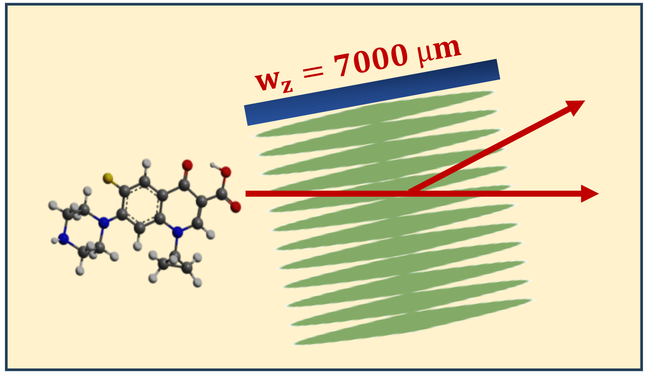
$$\psi_{\text{out}}(x) \propto \sum_{n=-\infty}^{n=\infty} J_n(\phi_0) e^{-i2nkx}$$

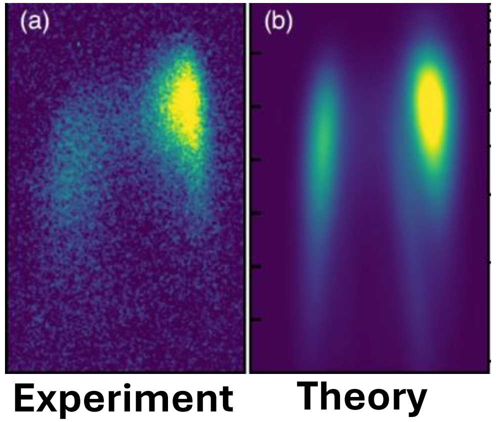




Bragg regime:

- Also for **polar molecules**
- Large : $\Delta p = 18\ \hbar k$
- Only **2 branches**



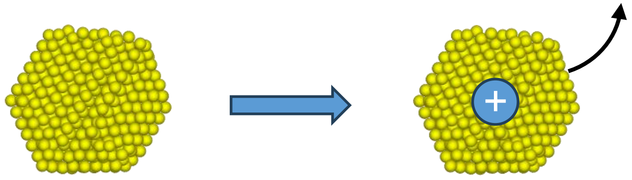


Experiment **Theory**

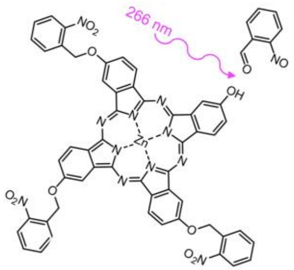
Nairz, O., Brezger, B., Arndt, M. & Zeilinger, A. Diffraction of complex molecules by structures made of light. **Phys. Rev. Lett. 87, 160401 (2001).**
C. Brand, F. Kialka, S. Troyer, C. Knobloch, K. Simonovic, B. A. Stickler, K. Hornberger, and M. Arndt, **Phys. Rev. Lett. 125, 033604 (2020).**

Photodepletion grating = Measurement-induced grating

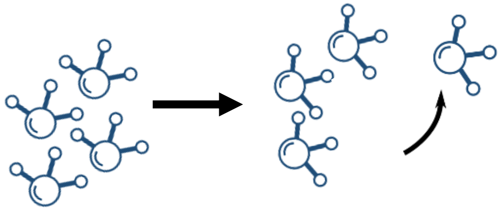
1. Photoionization of metal clusters



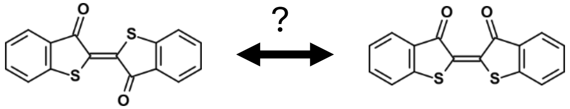
2. Photocleavage of covalent molecules

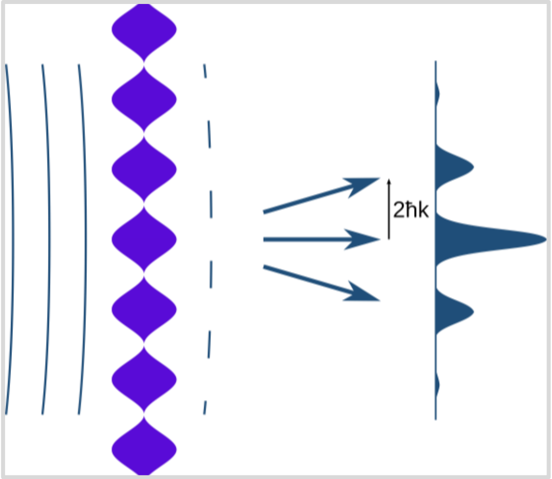


3. Cluster evaporation: atoms or molecules



4. Photoisomerization of molecules





- **Molecular beam depletion**
  at the anti-nodes of the standing light wave.
- **‘Interaction free’ beam splitting:**
  - **Measure** where the particles **are not**
  - **Particles are diffracted by “not having seen” the light.**
- **Momentum transfer is purely geometrical**

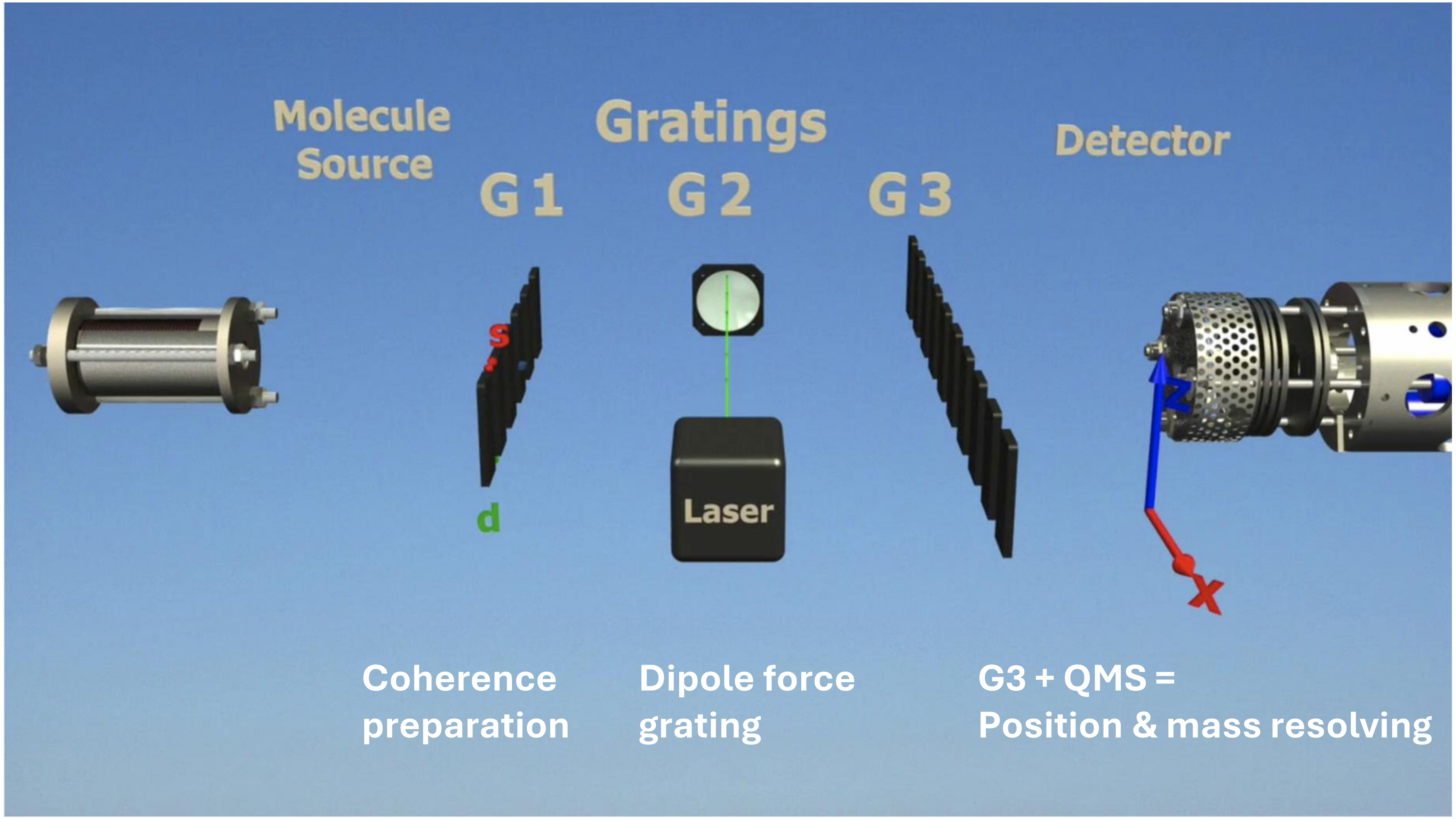
$$\Delta p = n \cdot 2\hbar k = n \cdot \frac{2h}{\lambda} = n \cdot \frac{h}{d}$$

E. Reiger, L. Hackermüller, M. Berninger, M. Arndt, *Exploration of gold nanoparticle beams for matter wave interferometry*, **Opt Commun 264, 326 (2006).**

K. Simonović, R. Ferstl, A. di Silvestro, M. Mayor, L. Martinetz, K. Hornberger, B. A. Stickler, C. Brand, M. Arndt, **under review 2024**

Matter-wave interferometry for measurements of molecular properties

Universal Matter-Wave Interferometry with complex molecules
KDTLI / LUMI 1.0



Coherence preparation

Dipole force grating

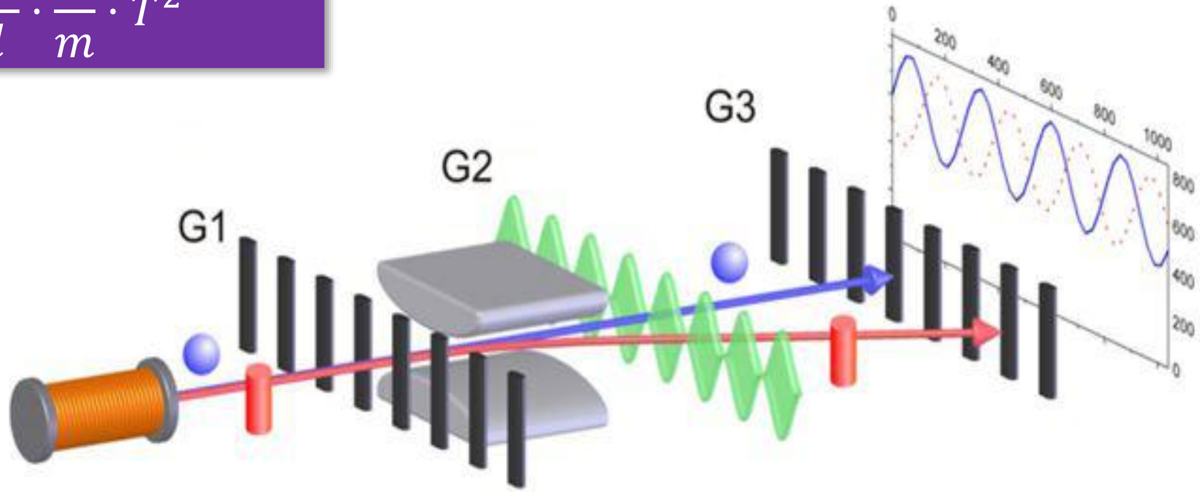
G3 + QMS = Position & mass resolving

First **L = 0.2 m** interferometer: S. Gerlich, L. Hackermüller, K. Hornberger, A. Stibor, H. Ulbricht, M. Gring, F. Goldfarb, T. Savas, M. Müri, M. Mayor, M. Arndt, **Nature Physics 3, 711 (2007).**
First **L = 2.0 m** interferometer: Y. Y. Fein, P. Geyer, P. Zwick, F. Kiałka, S. Pedalino, M. Mayor, S. Gerlich & M. Arndt, **Nature Physics 15, 1242 (2019).**

Matter-wave force sensing for Molecular Analysis

- **Quantum interferogram = molecular nano-ruler in free flight**
- **External forces shift the interference pattern**
- **We can resolve fringe shifts with < 2 nm resolution**

$$\Delta x \propto \frac{2\pi}{d} \cdot \frac{F}{m} \cdot T^2$$



Force sensitivity:
$< 10^{-26}$ N

Properties

Optical	$(\sigma_{abs}, \alpha_{opt})$
Magnetic	(χ_m, d_m)
Electric	(χ_{el}, d_{el})
Structural	(folding)
Dynamical	(photoisomers)

- Angew. Chem. Int. Ed. 47, 6195 (2008)
- Phys. Rev. Lett. 112, 250402 (2014).
- Angew. Chem. Int. Ed. 56, 10947 (2017)

- Phys. Rev. Lett. 121, 173002 (2018)
- Phys. Rev. X 10, 011014 (2020)
- Phys. Rev. Lett. 129, 123001 (2022)

Matter-wave interferometry as a probe of Quantum Macroscopicity

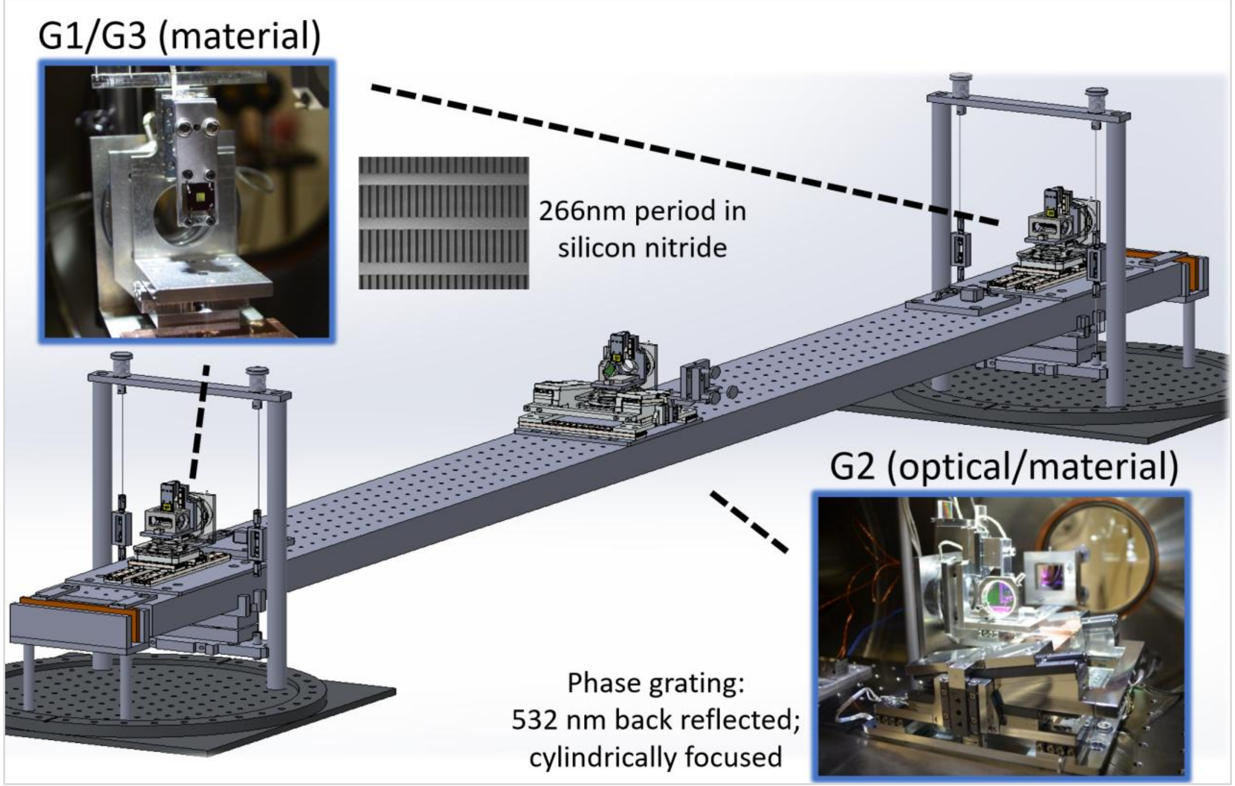
Long Base-line Universal Matter-Wave Interferometer, LUMI

Interference observed with:

- **Atoms:** alkalis, alkali earth
- **Molecules:** C_{60}, C_{70} , PAHs, radicals, 3-Peptides …

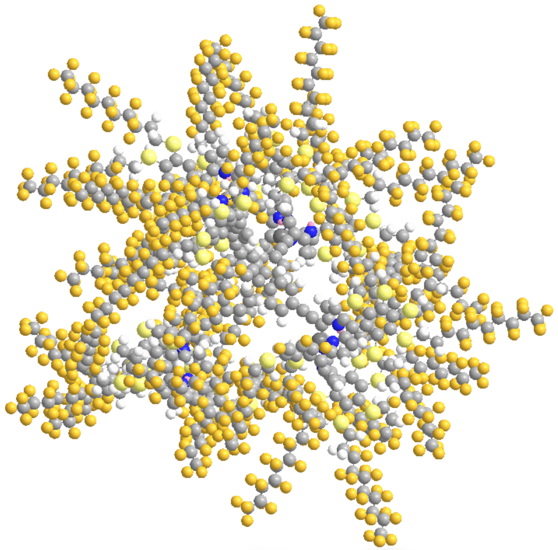
Design specifications:

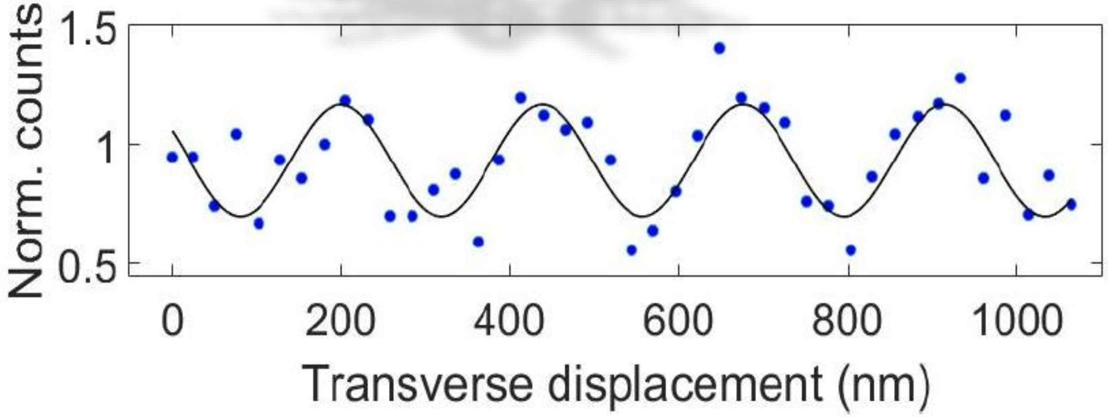
- **Wavelength** : $\lambda_{dB} < 50\text{fm}$
- **Mass** : $\mathrm{M} > 3 \times 10^4\ Da$
- **Force** : $F < 10^{-26}$ N



Y. Y. Fein, P. Geyer, P. Zwick, F. Kiałka, S. Pedalino, M. Mayor, S. Gerlich & M. Arndt,
Nature Physics 15, 1242 (2019).

Matter-wave Interference with Tailored Macromolecules





Functionalized Oligo-Porphyrines

- m ≃ 25‘000 – 28‘000 amu
- N ≃ 1800 - 2000 Atoms per molecule
- $C_{715}H_{260}F_{908}N_{16}S_{53}Zn_4$
- Velocity: $\mathrm{v} = 300$ m/s
- Molecular diameter: $D = 50$ Å
- Billions of structural Isomers
- De Broglie Wavelength $\lambda_{dB} = 50 - 60$ fm

Interference pattern in agreement with quantum theory

Y. Y. Fein, P. Geyer, P. Zwick, F. Kiałka, S. Pedalino, M. Mayor, S. Gerlich & M. Arndt **Nature Physics 15, 1242 (2019).**

Pushing the limits of Quantum Macroscopicity with Metal Clusters

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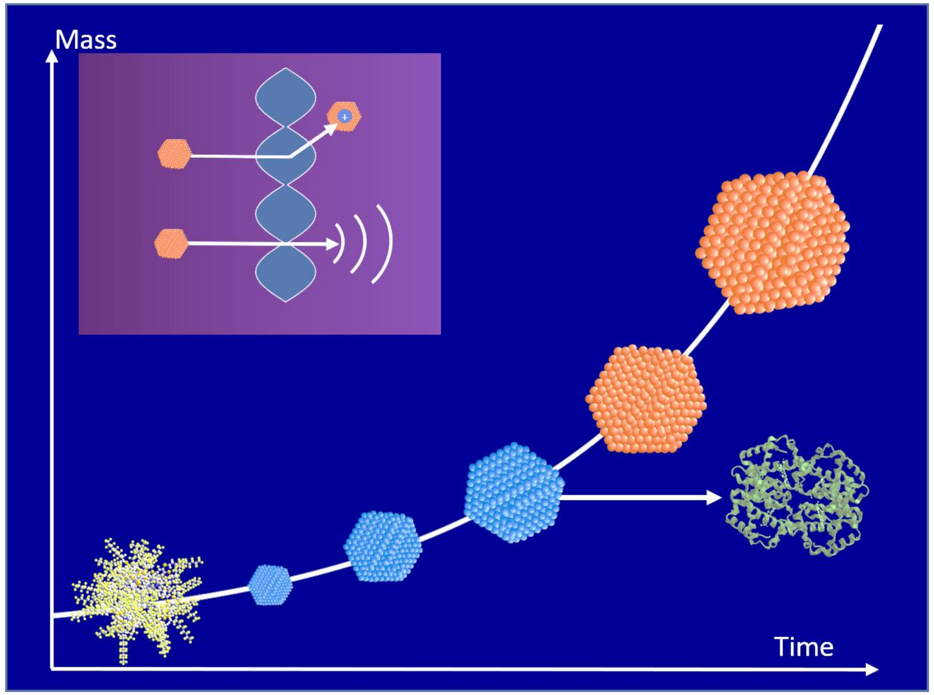
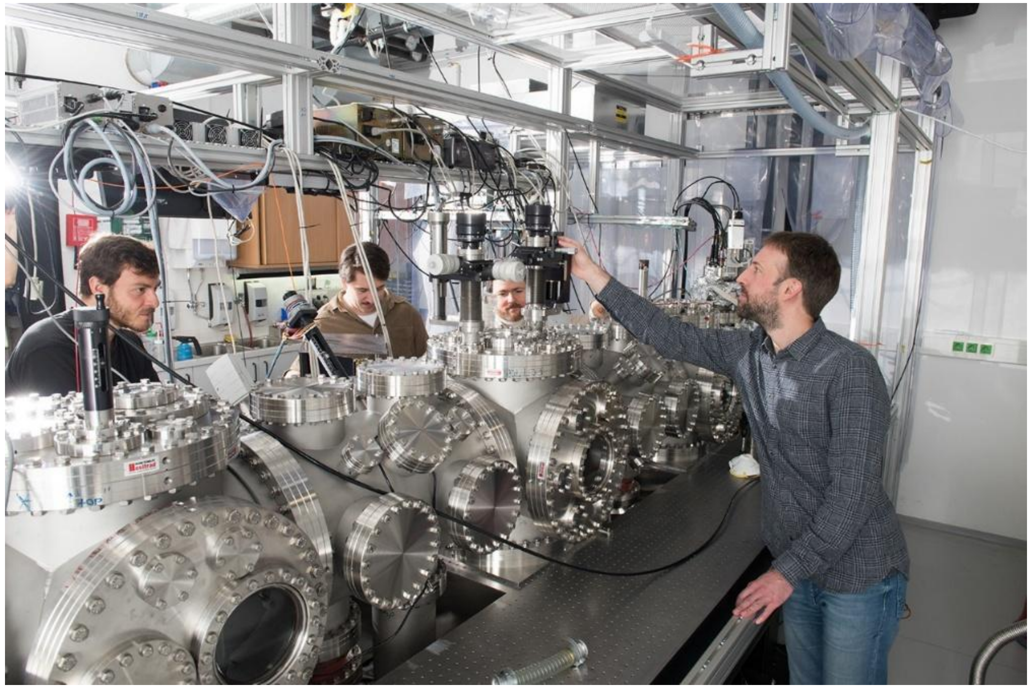




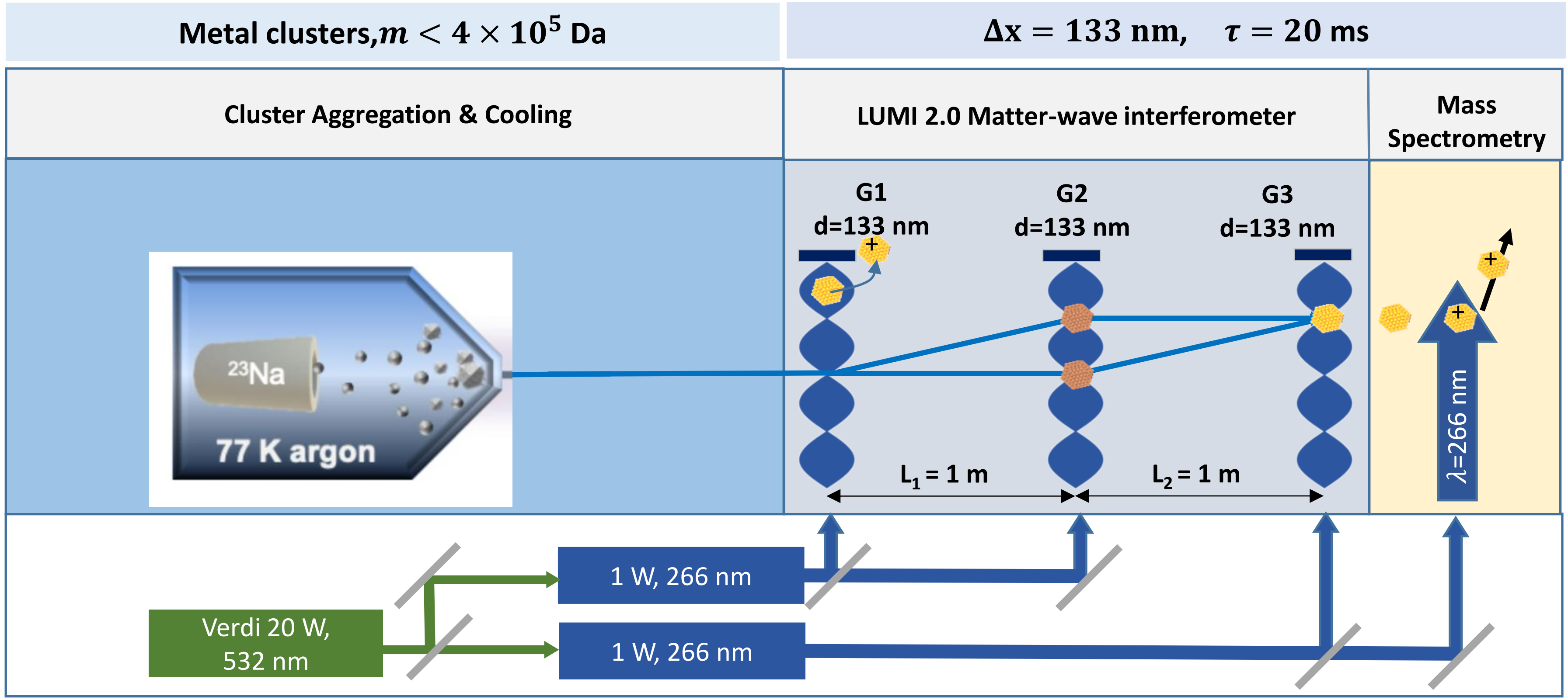
A vast new material class
whose quantum wave nature has never been exploited.

F. Kiałka, Y.Y. Fein, S. Pedalino, S. Gerlich, and M. Arndt, **AVS Quantum Science 4, 020502 (2022).**

S. Pedalino, B. Ramirez Galindo, T. Sousa, Y.Y. Fein, P. Geyer, S. Gerlich, and M. Arndt
Proc. SPIE 12477, Quantum Sensing, Imaging and Precision Metrology, 124470K (2023)

GORDON AND BETTY MOORE FOUNDATION

The MUSCLE experiment: Multi-Scale Cluster interference



Metal clusters, $m < 4 \times 10^5$ Da

$\Delta x = 133$ nm, $\tau = 20$ ms

Cluster Aggregation & Cooling

LUMI 2.0 Matter-wave interferometer

Mass Spectrometry

G1
d=133 nm

G2
d=133 nm

G3
d=133 nm

$L_1 = 1$ m

$L_2 = 1$ m

λ=266 nm

Verdi 20 W,
532 nm

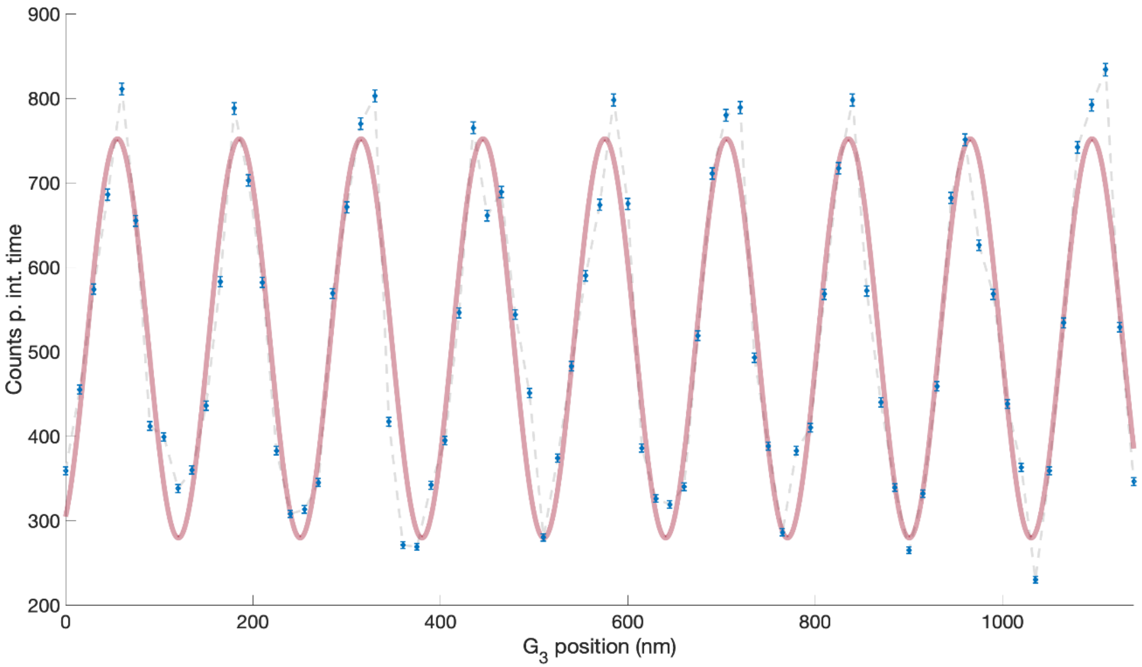
1 W, 266 nm

1 W, 266 nm

F. Kiałka et al, *Concepts for long-baseline high-mass matter-wave interferometry*, **Phys. Scr. 94, 034001 (2018).**

F. Kiałka, Y.Y. Fein, S. Pedalino, S. Gerlich, and M. Arndt, *A roadmap for universal high-mass matter-wave interferometry* **(2021).**

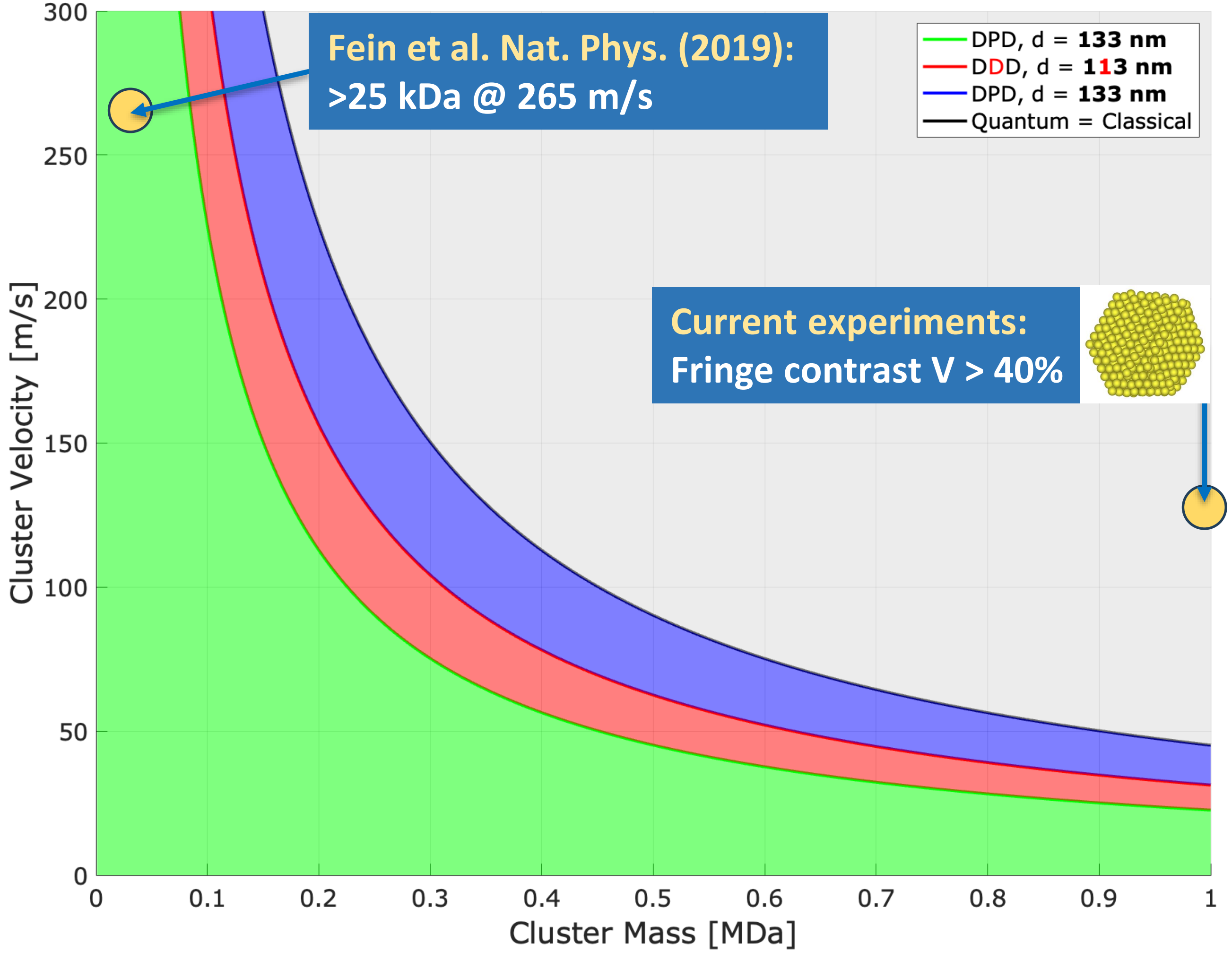
Observation of high contrast
Sodium Cluster Fringes @ „1 MDa“



At such high masses, classical & quantum patterns are identical: **Correspondence Principle**

But the high contrast allows **excluding many decoherence / dephasing processes**

Mass-Velocity boundaries of Talbot Lau interferometry



Fein et al. Nat. Phys. (2019):
>25 kDa @ 265 m/s

Current experiments:
Fringe contrast V > 40%

Towards Quantum interference of Proteins & Viroids

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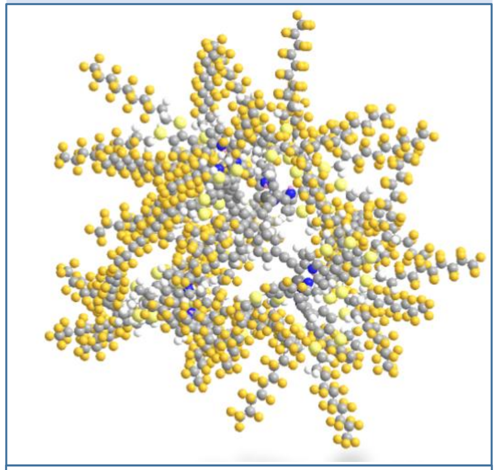
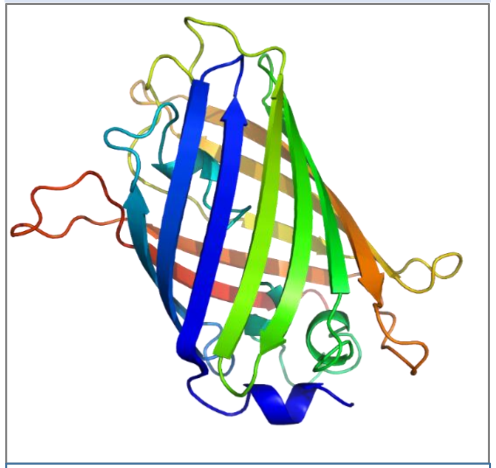
Quantum waves observed !



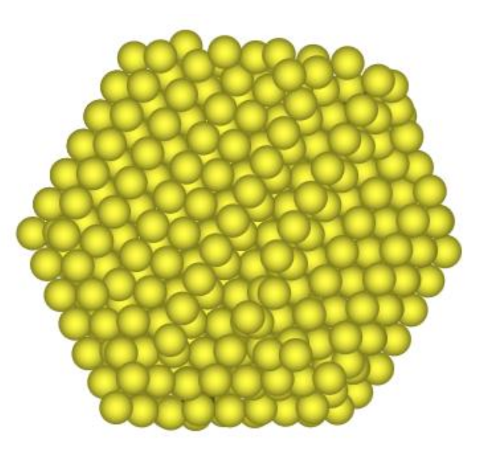
Perfluoroalkylated Oligoporphyrin (25-28 kDa)

Open Research !



Green Fluorescent Protein (27 kDa)

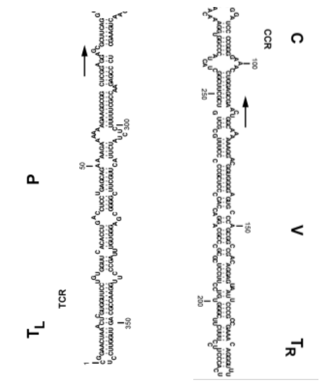
Consistent with Quantum waves !
To be confirmed



Sodium Cluster

(> 200 kDa)

Open Research !

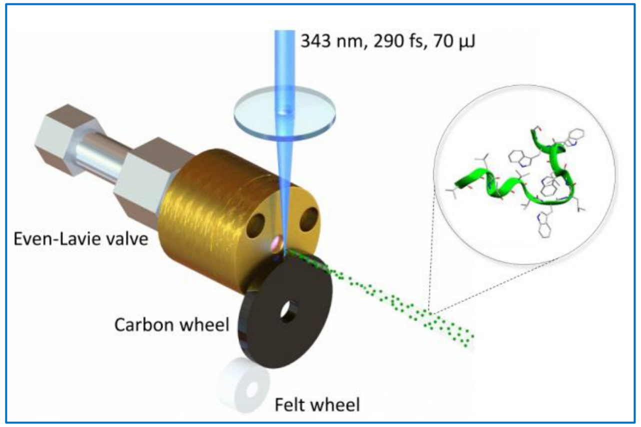


Family of Viroids (< 100 kDa)

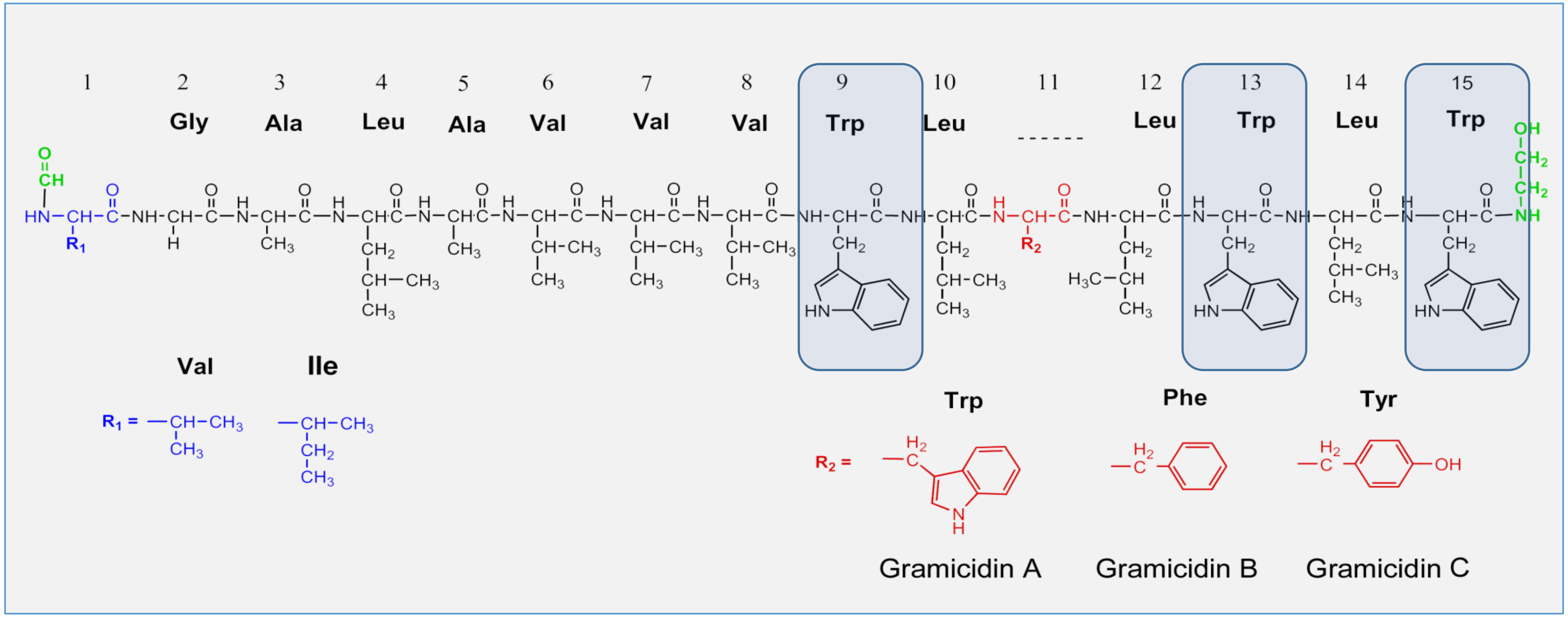
Biological nanomaterials represent a vast class of new materials whose quantum wave nature has not yet been exploited.
Greatest challenge: efficients sources !

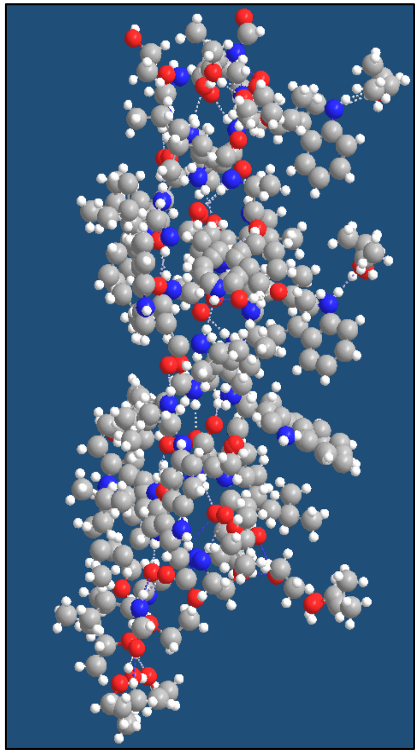
M. Arndt, De Broglie’s meter stick: Making measurements with matter waves, **Physics Today 67 30-36 (2014)**

Molecular beams of antibiotic polypeptides: **Gramicidin A1**

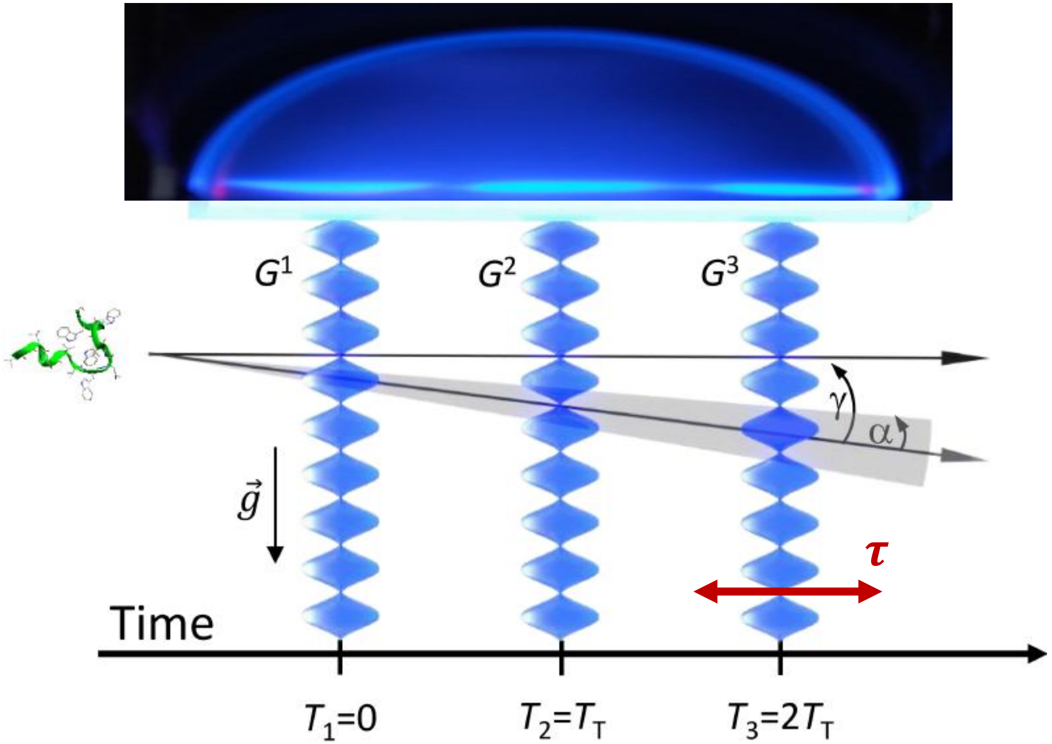


- Ultra-fast, intense laser light desorbs the peptides into an Argon jet
- The supersonic expansion cools the molecules and carries them to the interferometer
- Tryptophan residues are essential:
  among all amino acids only Tryptophan can be single-photon ionized
  by VUV radiation at 157 nm



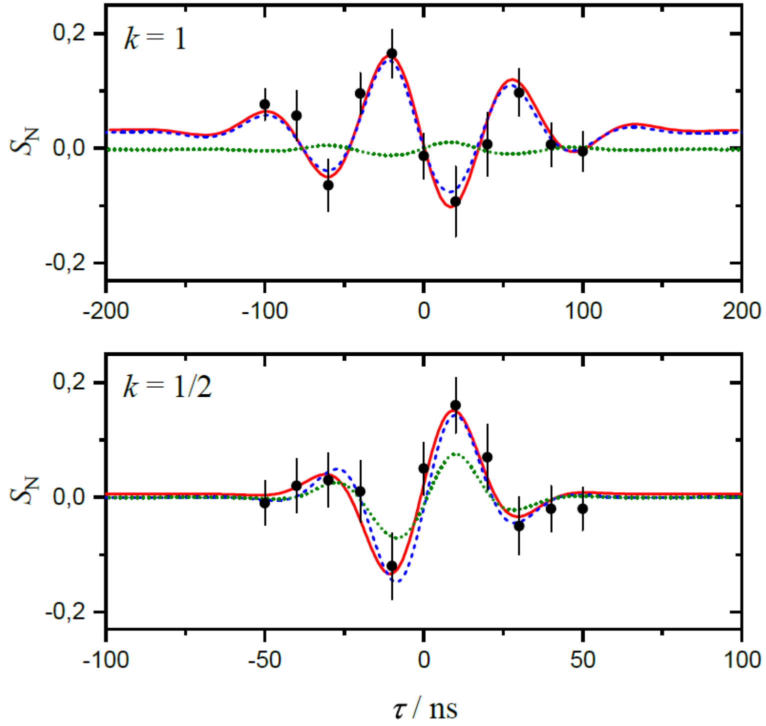


Quantum interference of the
Antibiotic polypeptide Gramicidin A



3 laser pulses of $\lambda_L = 157\ nm$ hit the molecules separated in time by $\tau \simeq T = m\, d^2/h$

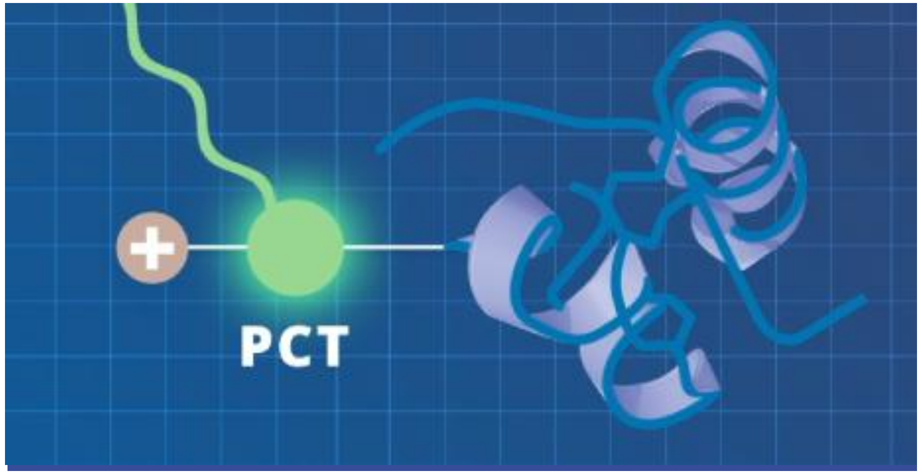
Interferometer transmission vs. pulse separation time τ

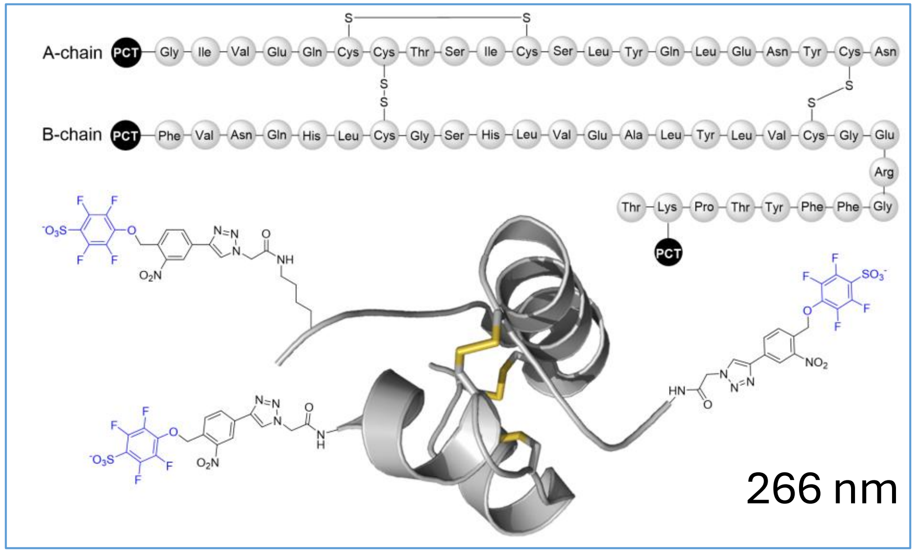


Black dots = Experiment
Red line = Quantum model (fits to experiment)
Green dotted line = Classical model (no fit)

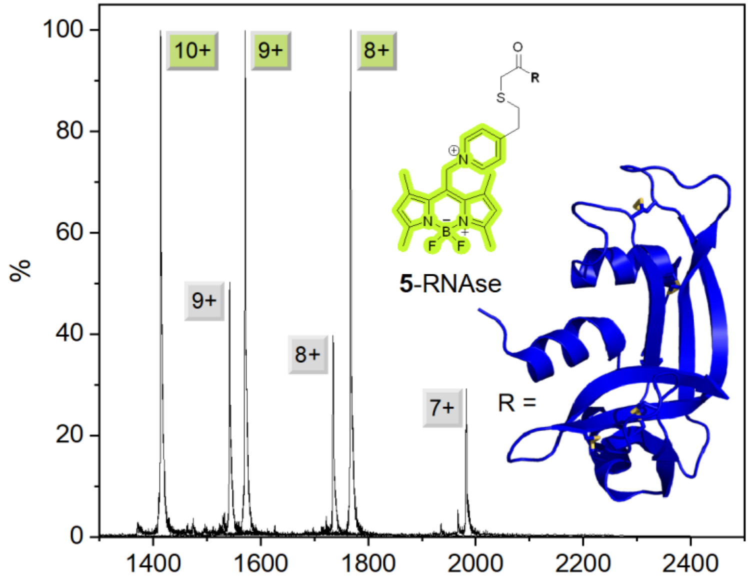
A. Shayeghi, P. Rieser, G. Richter, U. Sezer, J.H. Rodewald, P. Geyer, T.J. Martinez, and M. Arndt, *Matter-wave interference of a native polypeptide,* **Nature Communs. 11, 1447 (2020).**

Single-photon charge control of proteins:
Neutralization, beam splitting, Ionization (in progress)





- **Insulin** tagged with Benzonitrile ether: **cleaves at 266 nm**
- **RNAse** tagged with Bodipy pyridinium : **cleaves at 532 nm**



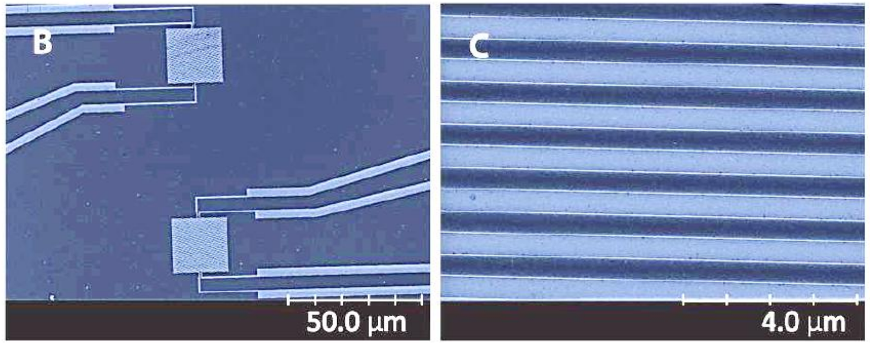
J. Schätti et al. Neutralization of Insulin
Chem. Commun. (Camb) 55, 12507-12510 (2019).

Y. Hua et al.
JACS Au 3, 2790–2799 (2023).

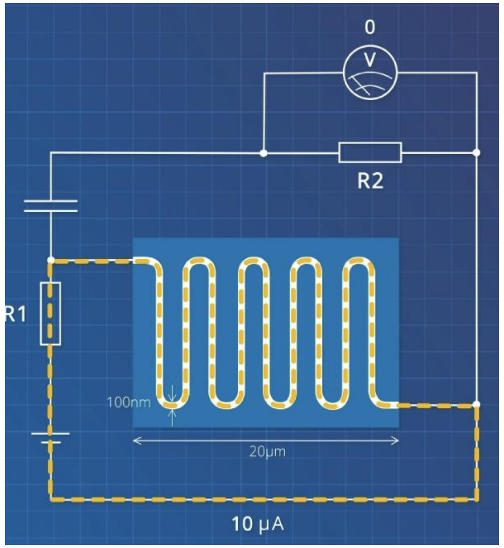
Highly sensitive Single Protein Detection:
Superconducting Nanowire Detectors (SNWD)

Nanowire Detectors (Single Quantum/Delft)

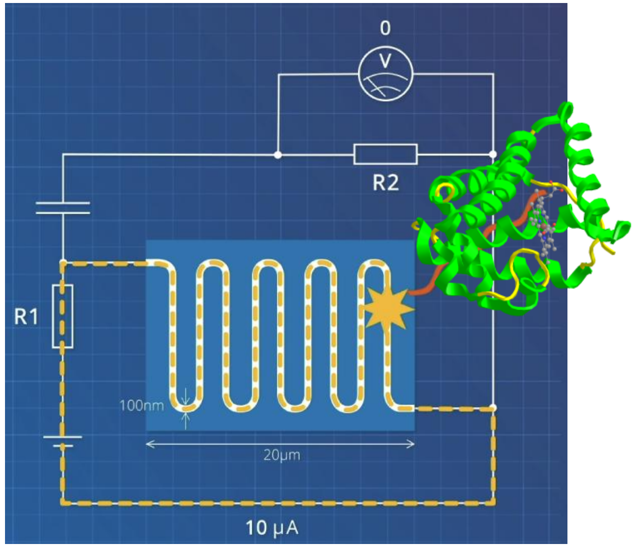
- NbTiN meander
- W =100 nm wide (500 nm)
- H = 10 nm
- A = 20 × 20 μm^2 (200 × 200 μm^2)



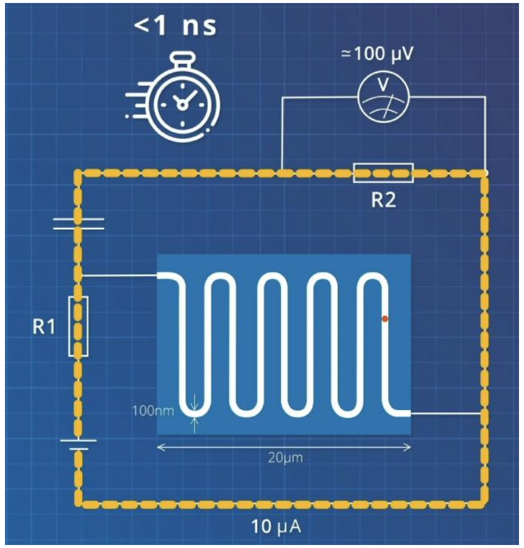
Supercurrent



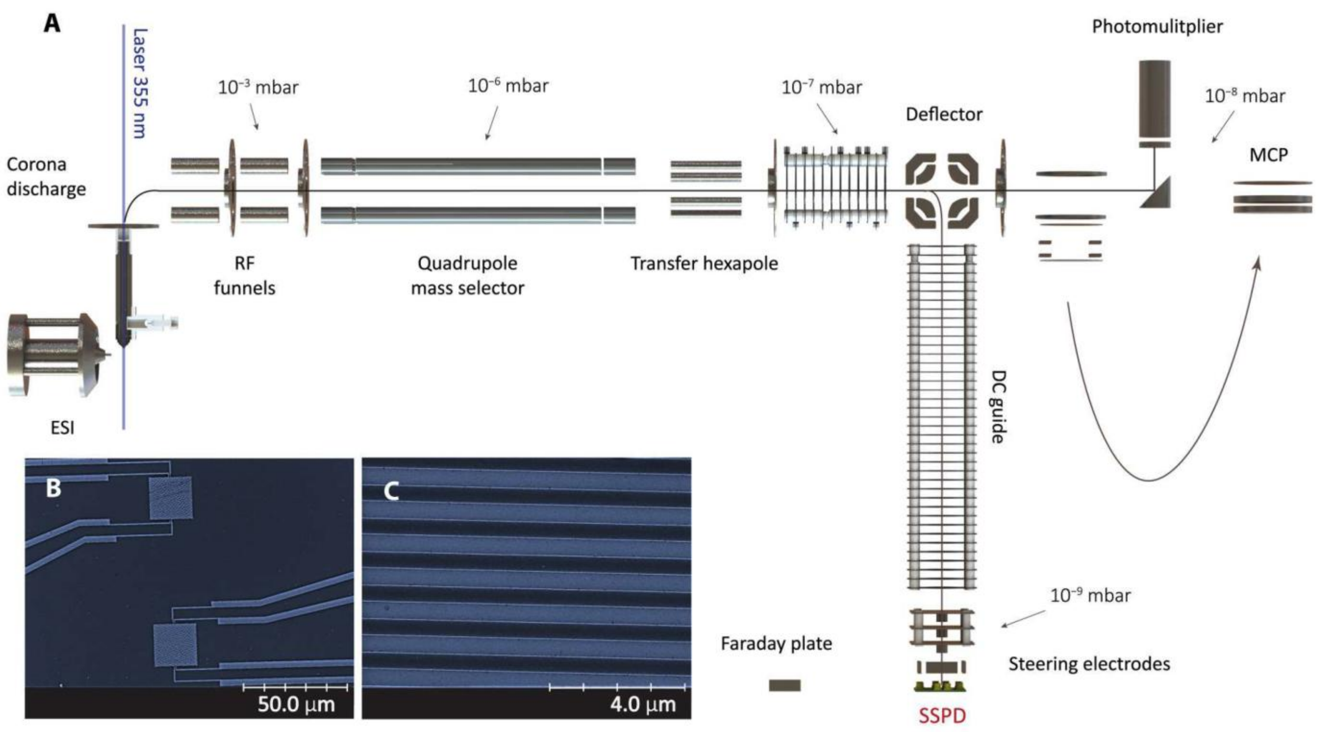
Protein impact

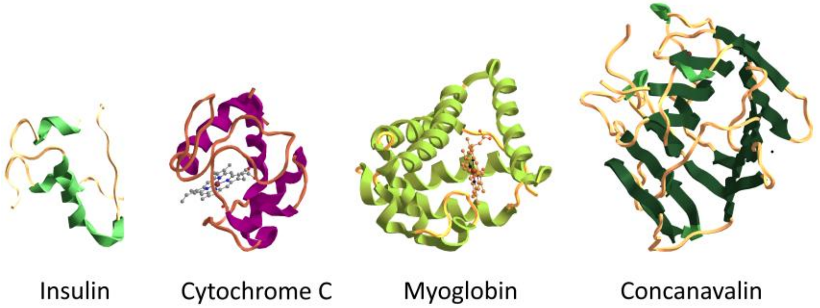


Phase transition reroutes the current



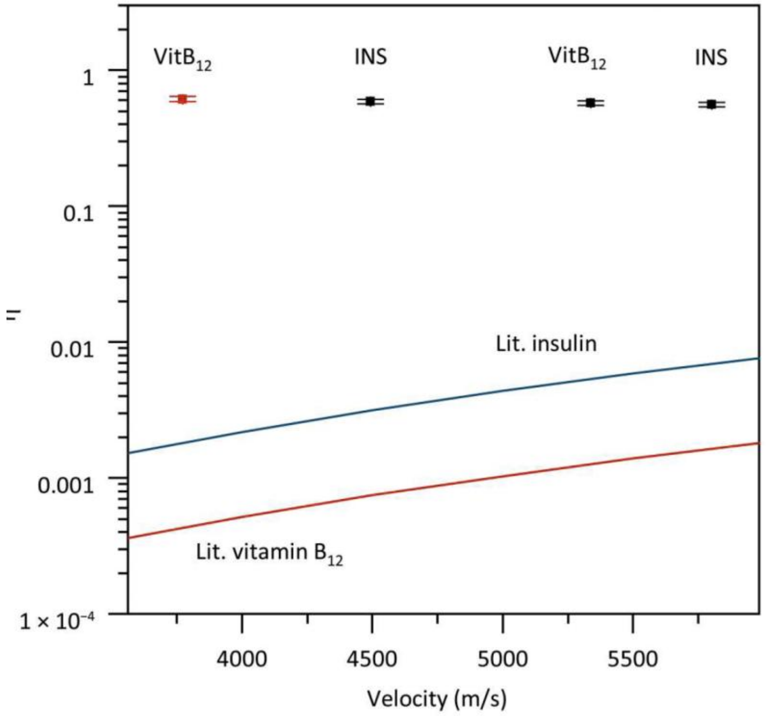
Quantum Enhanced Detection of Proteins
SSPDs in QMS mass spectrometry





At $E_{impact} < 100$ eV:

- **Quantum yield : $\eta \simeq 60\ \%$**
- **Area normalized: $\eta \simeq 100\%$**
- **Improve over MCP: 100 – 1000 x**



M. Strauß, A. Shayeghi, M. F. X. Mauser, P. Geyer, T. Kostersitz, J. Salapa, O. Dobrovolskiy, S. Daly, J. Commandeur, Y. Hua, V. Köhler, M. Mayor, J. Benserhir, C. Bruschini, E. Charbon, M. Castaneda, M. Gevers, R. Gourgues, N. Kalhor, A. Fognini, M. Arndt *Highly sensitive single-molecule detection of macromolecule ion beams*, ***Science Advances 9, eadj2801 (2023).***

SSPD sensitivity to the molecular impact energy

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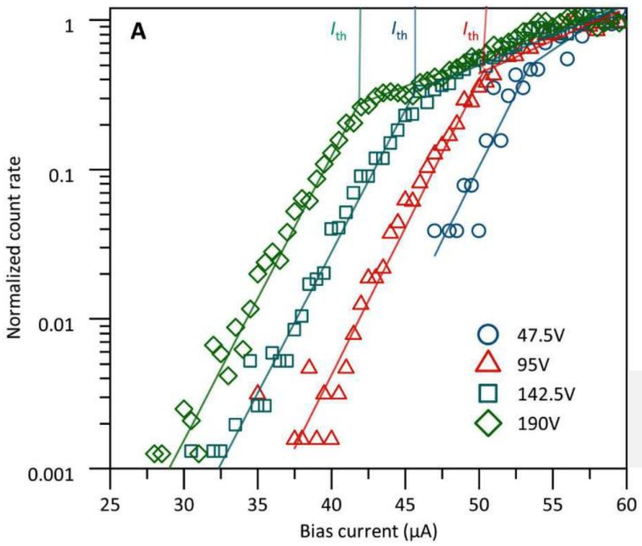
Superconducting nanowires can discriminate

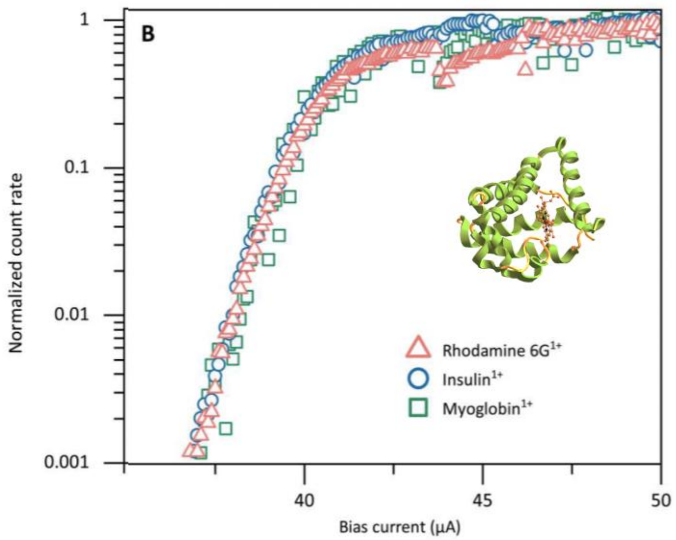
- **Different charge states**
  of the same mass
  in a fixed acceleration potential
- **Different impact energies**
  of the same protein



Impact energy

SSPDs are insensitive (at $E_{impact} = \text{const}$) to

- **Differences in momentum**
- **Molecular structure :**
  - Rh6G (0.5 kDa)
  - Insulin (5.7 kDa)
  - Myoglobin (17 kDa)



M. Strauß, A. Shayeghi, M. F. X. Mauser, P. Geyer, T. Kostersitz, J. Salapa, O. Dobrovolskiy, S. Daly, J. Commandeur, Y. Hua, V. Köhler, M. Mayor, J. Benserhir, C. Bruschini, E. Charbon, M. Castaneda, M. Gevers, R. Gourgues, N. Kalhor, A. Fognini, M. Arndt *Highly sensitive single-molecule detection of macromolecule ion beams,* ***Science Advances 9, eadj2801 (2023).***

Quantum enhanced detection:
Neutral organic molecules at low energy (3-6 eV)

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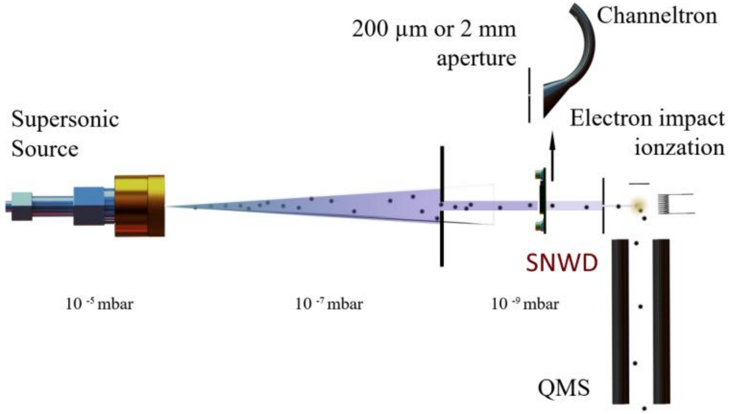
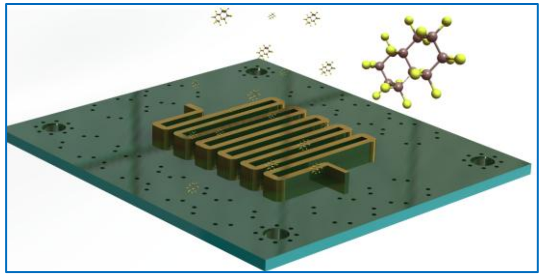
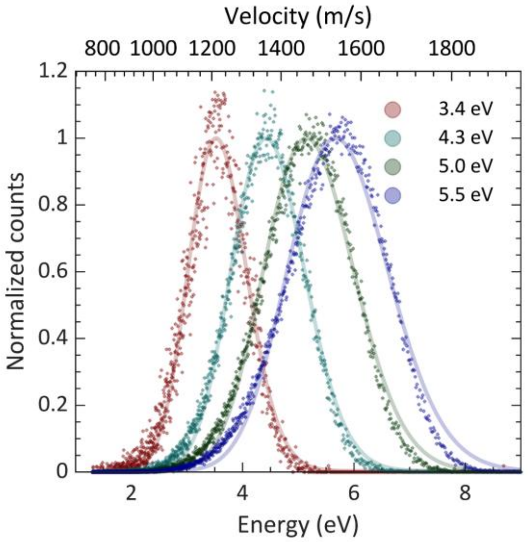


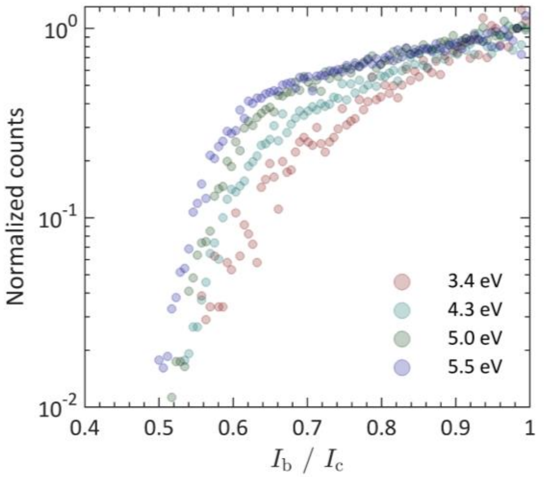


M. Strauß, R. Gourgues, M. Mauser, L. Kulman, M. Castaneda,
A. Fognini, A. Shayeghi, P. Geyer & M. Arndt (2024)

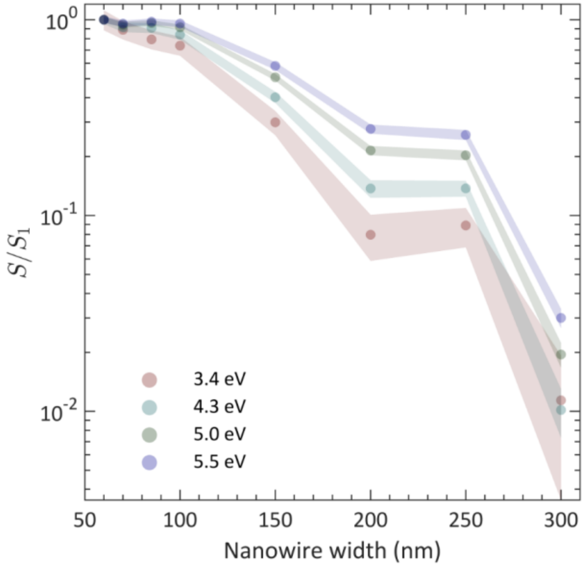
SSPD detection of low kinetic energy



Energy sensitivity of SSPD threshold



Quantum yield η : dependence on wire width



Future prospects in Vienna:
Universal matter-wave interferometry

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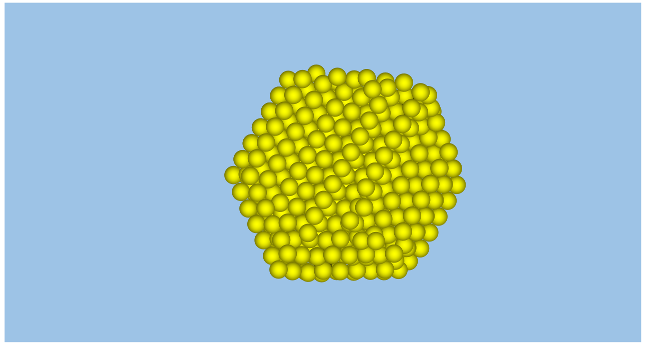
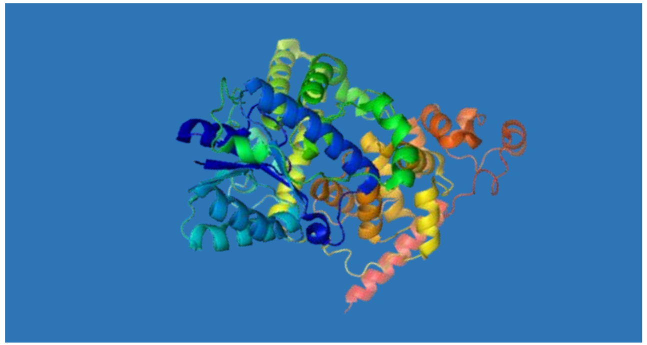
Metal Clusters

- Pushing Quantum Macroscopicity
- Phase transitions ?
- Super-resolution lithography



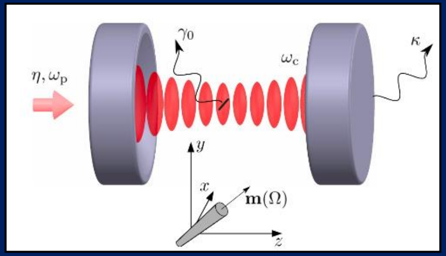
Proteins

- Quantum states of bio-matter
- Single-photon recoil spectroscopy
- Collisional decoherence: Folding



Cooling of NPs, Viruses & Bacteria

- Trapping
- Optomechanical cooling
- Rotational Quantum States



Thank you for your enthusiasm for physics ☺