

De Broglie's meter stick:

Making measurements with matter waves

Markus Arndt

Interfering atoms and molecules serve as exquisite rulers that sharpen our knowledge of physical laws, measure tiny forces, and reveal subtle properties of matter.

In 1923 Louis de Broglie proposed an idea that was as revolutionary as it was simple:¹ that one can “associate a periodical phenomenon with any isolated portion of matter or energy” and that a fixed observer will associate with that phenomenon a wave of wavelength $\lambda_{\text{dB}} = h/mv$ that scales with Planck's constant h , the object's mass m , and its velocity v .

De Broglie's waves came as a surprise because they represent states of matter that seem to defy classical logic. For instance, the textbook example of electron diffraction at a double slit can only be explained by assuming a delocalized quantum wave in free propagation—even though the electrons themselves are detected as localized clicks. Because every single particle can be described as a sum of two or more waves whose centers may be clearly separated, it is tempting to speak of the object as “being” in many places at once. The wave-particle duality is particularly conspicuous when observed with large molecules, which can be inspected under a microscope as localized composite particles before and after their wave-like evolution through the experiment.² Such observations have spurred interesting and ongoing debates about the meaning of words like reality, locality, space, and time.

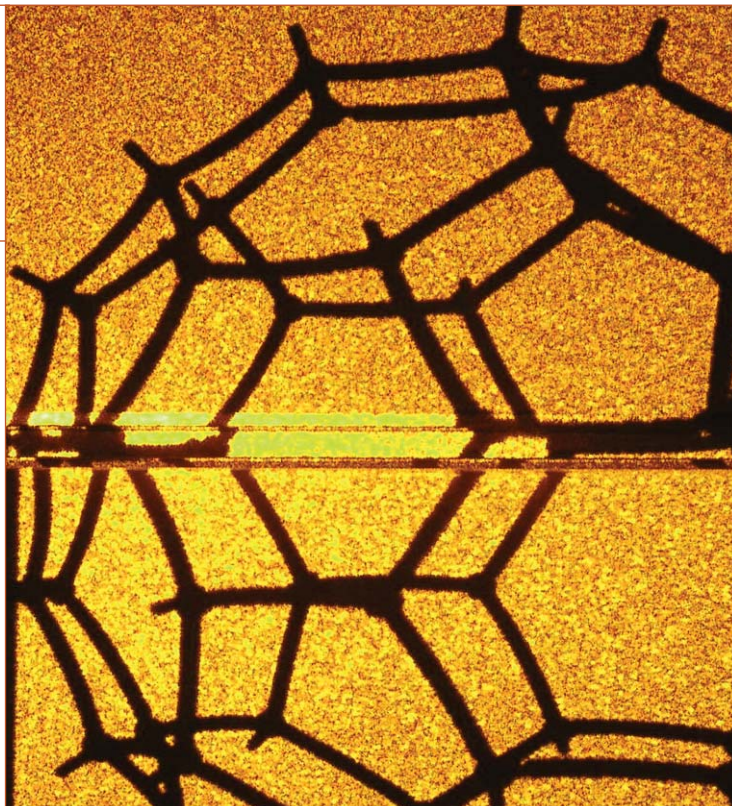
Independent of those philosophical puzzles, it's clear that the quantum wave nature of matter is firmly rooted in the Schrödinger equation, which has been perfectly confirmed in uncountable experiments. How, then, can we exploit the wave nature of matter in emergent quantum technologies? In recent decades various groups have devised strategies to use interfering atoms as fine rulers capable of

measuring minute fields, inertial forces, and the properties of atoms themselves. Lately the effort has been extended to molecules and macromolecules. Soon even large biomolecules may be studied in quantum-enhanced sensors. Eventually the sensitivity and accuracy of such matter-wave sensors is expected to exceed that of classical techniques. Here I focus on a question that's being asked by several groups across the globe: What kinds of matter-wave sensors are conceivable with atoms and molecules?

Setting up a quantum ruler

In quantum physics, both massless photons and massive particles are described by a wavefunction. For simplicity, let's restrict ourselves to a scalar function $\psi = \psi_0 \exp[i\phi(t)]$ with amplitude ψ_0 and time-dependent phase $\phi(t)$. The wave's phase—the position of its crests and troughs relative to some reference point in space and time—defines a natural ruler. Common detectors, however, are sensitive only to the intensity $|\psi|^2$. How can one then extract any phase?

Interferometers provide the solution. They convert phase differences between two waves into in-



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tensity modulations and, in turn, into measurable detector clicks. Dozens of interferometer types have been developed in classical and quantum optics. The key idea is always the same: An incident beam is divided by a beamsplitter into at least two wavelets; those wavelets—steered by mirrors, gratings, waveguides, or even gravity—travel along different paths before being recombined with a second beamsplitter.

The sensitivity of the interferometer derives from the fact that different parts of the same wave explore different regions of spacetime. They may travel different distances or be exposed to different forces or potentials. As a result, two wavelets, $\psi_1 \exp[i\phi_1(t)]$ and $\psi_2 \exp[i\phi_2(t)]$, acquire different phases. In linear optics as well as in quantum mechanics, we have to add the amplitudes rather than the intensities when two wavelets are superposed. Provided the wavelets remain indistinguishable in all degrees of freedom, they interfere and modulate the detected intensity according to their phase difference: $|\psi_{\text{tot}}|^2 = |\psi_1|^2 + |\psi_2|^2 + 2\psi_1\psi_2\cos(\phi_1 - \phi_2)$. To ensure that intensity modulations of individual matter waves add constructively to form an interferogram—that is, to ensure that the contributions of individual particles don't wash each other out—the beam must remain spectrally and spatially coherent for the duration of the experiment.

In an optical interferometer, such as the Mach-Zehnder interferometer illustrated in figure 1a, variations in the difference between two paths can be measured to a fraction of the light's wavelength. The sensitivity is limited only by the signal-to-noise ratio and the overall stability of the setup. State-of-the-art optical interferometers that are used to hunt for gravitational waves can even detect length changes as small as 10^{-18} m.

Modern matter-wave interferometers cover de Broglie wavelengths ranging from 10^{-13} m for macromolecules to more than 10^{-6} m for ultracold atoms. That corresponds roughly to the range between x rays and IR radiation in optics. A key advantage of matter waves over photons is that they couple to a plethora of external perturbations. Atoms and molecules have mass and rich internal electromagnetic spectra. Molecules and clusters add vibrational, rotational, and conformational dynamics. Thus matter waves are particularly sensitive to inertial and electromagnetic forces or collisions with many kinds of particles. As a result, matter-wave interferometers realize both force transducer and ruler in the same element. But how does one split a matter wave in the first place?

Dividing the indivisible

The word “atom” derives from the ancient Greek for indivisible. The splitting of an atomic beam, however, must certainly be more than the random sorting of particles in one direction or the other. That possibility is excluded by the fact that interference is based on the superposition of wavelets with a well-defined phase. We can also rule out the idea that each atom is physically split in any naive sense; all experiments confirm that each particle always contributes with the entirety of its internal proper-

ties—mass, polarizability, and so forth—in all accessible places. Atomic beam-splitting is actually about dividing an individual atom's quantum wavefunction. We may, in particular, distinguish between wavefront and amplitude beamsplitters.

Wavefront beamsplitters modify the wavefunction as a function of its position. In quantum physics, position and momentum are related through a Fourier transform. A modulation of the wavefront—for instance, by a mechanical grating of period d , as shown in figure 2a—therefore imprints a coherent superposition of transverse momenta $\Delta p = n \cdot h/d$, which leads to diffraction peaks in the far field (at distances much greater than A^2/λ , where A is the width of the beam-constricting aperture) behind the grating. The peaks correspond to well-separated wavelets having fixed phase relations but traveling their own unique paths through spacetime; n is a natural number describing the diffraction order. To achieve a wide separation of the interferometer arms, the grating period should be small. Many atom and molecule interferometers use gratings with $100 \text{ nm} < d < 1 \text{ }\mu\text{m}$. Such gratings can be produced in nanofabrication laboratories^{2,3} and even occur naturally in the skeletons of nanoporous algae.²

Wavefront beamsplitters can also be realized with optical gratings (see figure 2b). Phase gratings couple the polarizability of matter to the electric

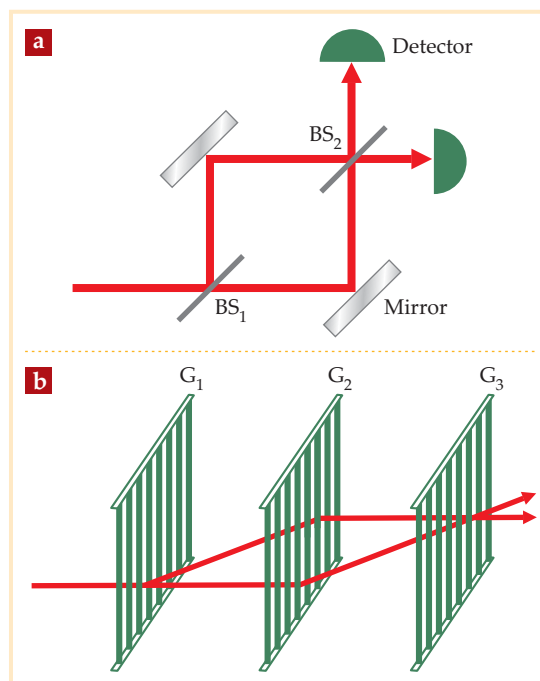


Figure 1. Interferometry's ingredients. (a) In an optical Mach-Zehnder interferometer, a beamsplitter (BS_1) divides an incident beam of light into a superposition of wavelets; mirrors steer the wavelets along two distinct paths toward a second beamsplitter (BS_2), and a pair of detectors records the intensity of the superposed wavelet fields. (b) In the matter-wave version of the interferometer, two nanoscale diffraction gratings (G_1 and G_3) act as the beamsplitters. A third grating (G_2) plays the part of the mirrors. The three-grating arrangement has become the basis for many atom and macromolecule interferometers.

field of a standing light wave to imprint a position-dependent phase onto the matter-wave field. Photodepletion gratings mask portions of the wavefunction—for instance, by transferring particles that pass through the grating's antinodes to undetectable states. Light masks may ionize atoms, dissociate molecular clusters, or change a particle's internal state to make the particle undetectable if it passes through an antinode of the light field. The effect closely resembles that of nanomechanical gratings.

A collimated particle source, a single grating, and a detecting screen can be interpreted as a full interferometer. If the source is sufficiently small or distant to the grating, a coherent wave field may evolve that covers transverse distances larger than the grating period d . The wavelets emerging from neighboring slits will then interfere on a screen in the far field, even without the help of additional beamsplitters and beam-steering elements. Modern precision experiments, however, typically include three or four optical elements to achieve larger beam separation, longer coherence times, and higher sensitivity to external perturbations.

In 1991 a group led by David Pritchard combined three equidistant, nanofabricated gratings, configured as shown in figure 1b, to realize a Mach-Zehnder interferometer for sodium atoms.³ The first grating (G_1) splits the wavefront into wavelets of various diffraction orders. A second (G_2) and third

(G_3) grating can be arranged to redirect and recombine those wavelets to obtain quantum interference. The intensity profiles of the two recombined beams behind G_3 are complementary and depend on the phase shift between the two arms of the interferometer. Additional interference pathways arise through higher diffraction orders. They can be spatially separated with a slit.

Well-collimated atomic beams have been split into arms separated by as much as 27 μm , sufficient to expose two wavelets to different electric fields or to regions of different gas pressure. That approach produced the most precise measurement of an atomic ground state polarizability to date and a sophisticated analysis of atom-atom scattering cross sections.³

Interfering incoherent beams

Mach-Zehnder interferometry can be generalized to many atoms in the periodic table and to some molecules. It requires, however, an intense and transversely coherent beam, which can be routinely achieved for many atoms but remains an open challenge for molecules and nanoparticles. Atomic beams are typically formed by evaporation or sublimation, sometimes at temperatures exceeding 1000 K. Massive molecules tend to be less volatile and would require still higher temperatures. Many molecules, atomic and molecular clusters, and nanoparticles, however, need to be studied at cool

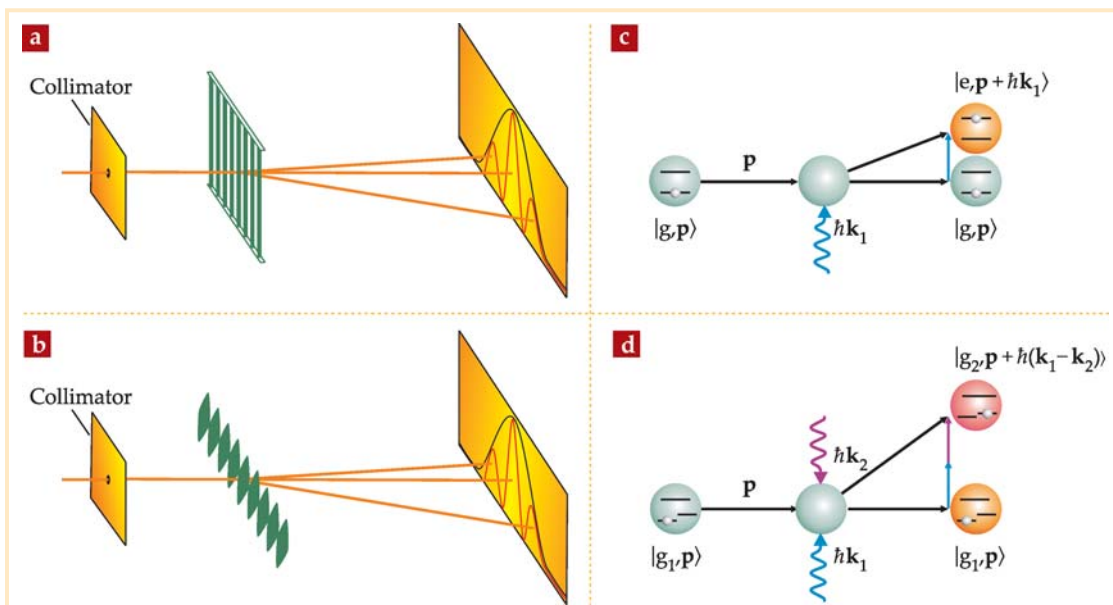


Figure 2. Splitting matter waves. (a) A nanomechanical diffraction mask splits an atomic or molecular beam by dividing the matter wavefront into beams of varying diffraction order. (Beams of diffraction order 0 and ± 1 are shown here.) (b) A beam can be similarly split using a standing light wave, which may remove parts of the matter wave via ionization or fragmentation or may modulate the wavefront's phase via dipole interaction. (c) Amplitude beamsplitters use laser pulses to create a coherent superposition between an atom's ground state $|g\rangle$ and a resonantly coupled excited state $|e\rangle$. Upon excitation, the momentum $\mathbf{p}$ of the atom is enhanced by the momentum $\hbar\mathbf{k}$ of the photon. The atom therefore ends up in an entangled state, where neither the internal nor the center-of-mass state is known but where the two are strictly correlated. (d) When the superposition is created between two hyperfine ground states $|g_1\rangle$ and $|g_2\rangle$ via a two-photon Raman transition, the apparatus is known as a Raman beamsplitter. Wider arm separations can be achieved with higher-order Raman transitions. (Panels c and d adapted from ref. 16.)

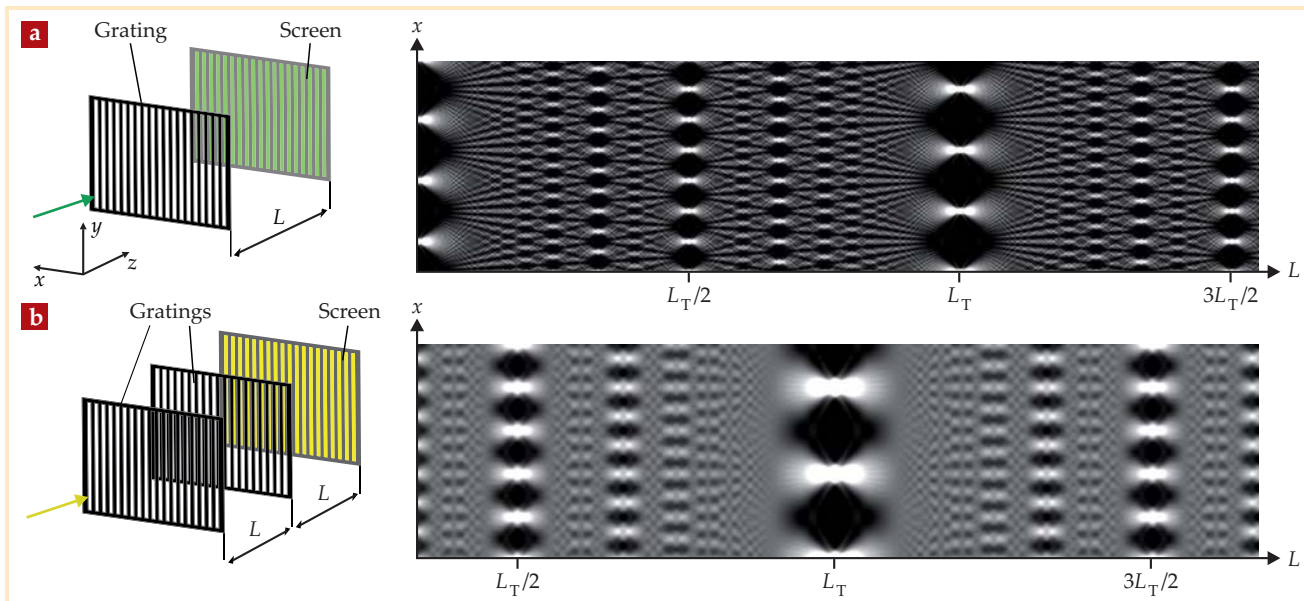


Figure 3. Talbot-Lau interferometry. (a) Coherent radiation of wavelength λ that's diffracted by a transmission grating of period d produces self-images of the grating's intensity distribution at multiples of the Talbot distance $L_T = d^2/\lambda$. Such a self-image is visible in the interference pattern at right, collected by scanning a detection screen along the z direction. (Fractional images of the grating occur at fractions of the Talbot distance.) (b) To achieve the same effect for monochromatic but spatially incoherent radiation, one can introduce a second, identical grating. Although the first grating does not produce visible interference fringes, it creates coherent illumination for the second grating, which then images the first onto a detection screen. The pattern shows interferograms for symmetric arrangements in which the spacing L between the two gratings is equal to that between the second grating and the detection screen. (Adapted from ref. 16.)

temperatures: Biomolecules usually denature at temperatures in excess of 330 K, nanoparticles can decompose when heated, and clusters often only form by aggregation at temperatures of a few kelvin. Various methods developed for mass spectrometry have proven potent for generating charged or fast neutral beams of macromolecules or clusters, but methods for forming intense, directed, and neutral particle beams of uniform mass, low velocity, and low temperature still require extensive development.

In 1997 John Clauser suggested a scheme that could substantially increase the signal even for a weak, spatially incoherent beam source. Known as a Talbot-Lau interferometer,⁴ it is based on William Henry Fox Talbot's observation, nearly two centuries ago, that spatially coherent light can image periodic structures even in the absence of lenses. As shown in figure 3a, coherent light passing through a grating produces self-images of the grating at multiples of the Talbot distance $L_T = d^2/\lambda$. At first glance, that seems to contradict our textbook knowledge that the intensity pattern behind a grating peaks at $\sin \theta = n\lambda/d$. The discrepancy is explained by the fact that Talbot's observation holds only in the near field behind the grating ($z \ll A^2/\lambda$); the textbook equations are given for the far field ($z \gg A^2/\lambda$).

So far, it seems we've gained nothing; we still need coherent light. However, the physics that leads to diffraction under coherent illumination also creates coherence under incoherent illumination. That result from light optics also holds for matter waves. If each slit is sufficiently narrow, the momentum of

each incident wave expands coherently by virtue of quantum physics.

In 1948 Ernst Lau suggested exploiting that finding in the optics setting by combining two gratings: Diffraction at any one slit in the first grating, G_1 , will produce wavelets that expand sufficiently to overlap several slits in G_2 . Multipath interference behind G_2 then creates a self-image of G_1 at a certain distance farther downstream. If the spacing between the gratings is chosen properly, typically on the order of the Talbot distance, the interferograms caused by all the wavelets emerging from G_1 overlap at the same distance behind G_2 and add constructively to form an interference pattern such as that shown in figure 3b. The pattern can be recorded on a screen, but for practical reasons, it's often detected behind a third grating.

The above strategy has been realized with various beamsplitters for clusters and large molecules. The version in which all three gratings are mechanical is known as a Talbot-Lau interferometer; when the central grating is replaced by an optical phase grating, it is called a Kapitza-Dirac-Talbot-Lau (KDTL) interferometer.⁵ The all-optical version, which uses three pulsed photo-depletion gratings, has been established as the optical time-domain ionizing matter-wave (OTIMA) interferometer.⁶

Entanglement-based beamsplitting

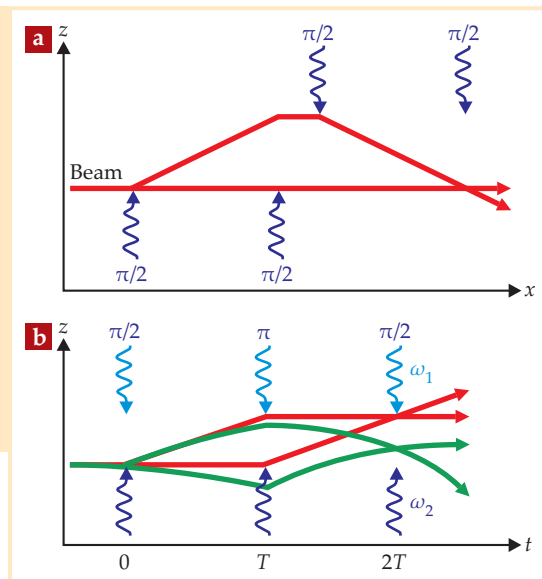
Amplitude beamsplitters divide the center-of-mass wavefunction of particles independent of their lateral position. A particularly important implementation,

Figure 4. Amplitude beamsplitters. (a) Pulsed laser beams at fixed positions and oriented orthogonal to an atom beam can form what's known as a Ramsey–Bordé interferometer: A $\pi/2$ pulse—which places the beam in a superposition of entangled electronic and momentum states, as depicted in figure 2c—splits the beam into two arms; two successive $\pi/2$ pulses steer the arms back together; and a fourth $\pi/2$ pulse recombines the arms to produce the interference pattern. **(b)** An interferometer can also be realized with a three-pulse, $\pi/2$ – π – $\pi/2$ sequence, as shown here for the case in which Raman pulses—counterpropagating beams whose frequency difference, $\omega_2 - \omega_1$, is tuned to a hyperfine transition—are applied to an atomic cloud at intervals of time T . In the presence of gravity, the interferometer can be used to infer the acceleration due to gravity based on the relative phase of the interferometer arms at time $2T$. (Red and green paths indicate atom-cloud trajectories in the absence and in the presence of gravity, respectively.)

illustrated in figure 2c, is based on the entanglement between the internal and the external states of atoms: When resonant light irradiates an atom having an effective two-level energy system, the light couples the ground state $|g\rangle$ to the excited state $|e\rangle$ and induces population oscillations between them. The oscillations occur at the Rabi frequency $\Omega_R = \mathbf{d} \cdot \mathbf{E}/\hbar$, where $\mathbf{d}$ is the dipole moment between $|g\rangle$ and $|e\rangle$, $\mathbf{E}$ is the light's electric field, and $\hbar$ is the reduced Planck's constant. If the interaction time τ is chosen to be $\int_0^\tau \Omega_R dt = \pi/2$, an atom initially in state $|g\rangle$ is transferred into a coherent superposition of $|g\rangle$ and $|e\rangle$. At the same time, the atom is transferred from its initial momentum state $|\mathbf{p}\rangle$ to the coherent superposition of $|\mathbf{p}\rangle$ and $|\mathbf{p} + \hbar\mathbf{k}\rangle$, where $\mathbf{k}$ is the photon wavevector. Because there is a strict correlation between the two processes—the photon absorption that electronically excites the atom also gives it a momentum kick—the atom ends up in an entangled state $|\psi\rangle \propto |g, \mathbf{p}\rangle - \exp(i\phi)|e, \mathbf{p} + \hbar\mathbf{k}\rangle$, and the wavefunction can no longer be factorized into its internal and external degrees of freedom.⁷

Sequences of $\pi/2$ pulses were originally used in high-resolution spectroscopy to determine transition frequencies in atoms and small molecules. In 1989 Christian Bordé realized that the method, illustrated in figure 4a, also implements a matter-wave interferometer, now often referred to as a Ramsey–Bordé interferometer.⁸ During a first pulse, an atom may absorb a photon and receive the accompanying momentum kick, or it may ignore the incident light. A second $\pi/2$ pulse hits the atom from the same side; if the atom is still in its ground state, it may stay put, as shown in the figure, or get electronically excited. If the atom is already in the excited state, it may ignore the arriving light or it may again receive the recoil of one photon, this time via stimulated emission in the direction opposite the beam source. A third and fourth pulse repeat the processes, except with inverted signs of momentum transfer, to form a closed interferometer.

In 1991 Mark Kasevich and Steven Chu realized an entanglement-based atom interferometer by coupling two hyperfine ground states via two-photon Raman transitions (see figure 2d). That leads to a boost in precision, because hyperfine ground states do not decay during the time scale of the experiment. Moreover, the momentum transfer using two-



photon transitions is almost double that of single-photon beamsplitters.⁹ The researchers also simplified the pulse sequence: As illustrated in figure 4b, three pulses—a $\pi/2$ pulse, followed by a π pulse that interchanges the momenta of the two wave components, followed by a second $\pi/2$ pulse—suffice to form a Mach–Zehnder interferometer, now often referred to as a Raman interferometer. Entanglement-based beamsplitters have become the basis for the most sensitive and most accurate matter-wave sensors to date.^{10,11}

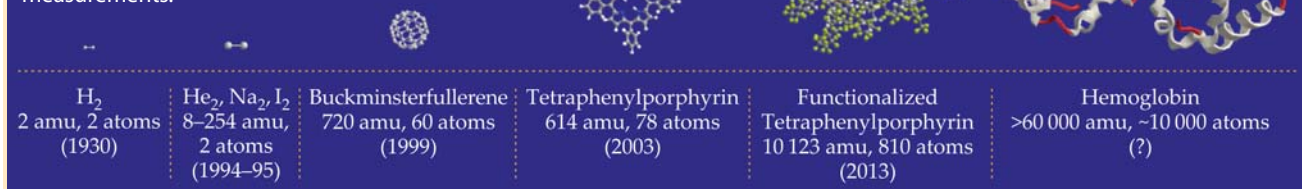
Atoms as quantum sensors

Atom interferometers are particularly useful for measuring inertial forces, such as Earth's gravity or rotation; in ultrahigh vacuum, atoms are essentially unperturbed test masses. In three-beamsplitter interferometers like the ones shown in figures 1b and 4b, wavelets traveling along the two arms acquire a relative phase shift $\Delta\phi = \Delta p \cdot a \cdot T^2/\hbar$, where a is the acceleration, Δp the momentum transfer in each beamsplitter, and T the time of flight between successive beamsplitters. Typical experiments are optimized to detect either Earth's gravitational acceleration, $a_g = g \approx 9.81 \text{ m/s}^2$, or the rotational acceleration, $\mathbf{a}_c = 2\mathbf{v} \times \boldsymbol{\Omega}_E$, due to the Coriolis effect, where $\mathbf{v}$ is the atom's velocity and $\boldsymbol{\Omega}_E$ is the vector of Earth's angular frequency.

In 1999 Achim Peters and colleagues used Raman interferometry in a fountain configuration¹²—in which a cold cloud of atoms is launched against gravity and returns in free fall—to measure g with an accuracy of $\Delta g/g = 3 \times 10^{-9}$. Recently, Susannah Dickerson and colleagues in Kasevich's lab have developed a 10-m fountain¹¹ that could potentially measure gravitational acceleration with a sensitivity of $3 \times 10^{-11} \text{ g}/\sqrt{\text{Hz}}$.

When two atom gravimeters are combined, they measure gravity gradients. Guglielmo Tino's group recently used that approach¹³ to measure Newton's gravitational constant G in the presence of a moving test mass with a statistical uncertainty of $0.011 \times 10^{-11} \text{ m}^3 \text{ kg}^{-1} \text{ s}^{-2}$.

Figure 5. Matter-wave diffraction started in 1930 with diatomic particles, but it didn't gain momentum until the early 1990s. Since then the technique has been extended to progressively larger and more complex molecules; some are depicted here along with their mass in atomic mass units, the number of atoms they comprise, and the year they were first successfully used in interference experiments. To date, a functionalized tetraphenylporphyrin, synthesized by Marcel Mayor and colleagues, is the most massive object for which matter-wave interference has been seen. In the future, bionanomatter such as hemoglobin proteins will likely be studied using matter-wave-enhanced measurements.



Atom gravimeters will be important for environmental monitoring applications: Earthquakes and volcanoes may give telltale warnings in the form of weak local accelerations. Earthbound and satellite-based inertial sensors may permit a more precise determination of the geoid—Earth's gravitational equipotential surface—and thereby provide climatologists with information about changes of water tables and glaciers.

Quantum gravimeters with a sensitivity of 1 part in 10^8 or better can assist in the prospection for deeply buried natural resources. Closer to the surface, small caves or sewage pipes might be detectable via quantum gravity gradiometry.

Atomic inertial sensors are also of interest for navigation systems. Global positioning systems rely on satellite signals, which are inaccessible in deep water and may be jammed by adversaries in the case of a military conflict. In the future, inertial navigation systems that combine gravitational and rotational accelerometers may enable precise positioning by integrating all accelerations along a path. Only the occasional correction with an external control signal would be necessary. Many of those applications are expected to flourish as soon as devices become compact and portable.

Ultrasensitive atom gravimeters will set new bounds on deviations from the weak equivalence principle—the conjecture that an object's freefall trajectory is independent of its composition and structure—on violations of local Lorentz invariance, on the gravitational redshift, and on the postulated existence of a “fifth force.” They may also serve as detectors for low-frequency gravitational waves.

Whereas inertial sensing uses atoms as test masses to measure external fields, precision measurements of the photon recoil in a Raman beam-splitter allow extraction of the ratio of h to atomic mass.¹⁴ That ratio, along with precise mass measurements, yielded the fine structure constant $\alpha^{-1} = 137.035\,999\,037$ with a relative uncertainty of 6.6×10^{-10} .

Molecules as quantum sensors

In principle, a molecule interferometer can detect both inertial forces and relativistic effects, just as an atom interferometer can. An interesting test of the

equivalence principle, for instance, would be to compare matter of vastly different composition or to systematically explore the mass dependence of phase shifts. Almost certainly, those studies will happen in the future.

For now, however, atom interferometry continues to lead the way in sensing, because hundreds of research groups have spent more than three decades on the development of cold and ultracold atom technologies. The development of intense sources of neutral macromolecules and highly efficient detectors is still an open challenge.

In my group at the University of Vienna, we've chosen to focus our molecular interferometry efforts on questions that no other experiment could address with the same precision and scope. On the one hand are tests of the linearity of quantum mechanics in the limit of ultrahigh masses. On the other hand is the study of delocalized particles in well-defined fields, which yields information about the particles' internal properties. In KDTL and OTIMA interferometry, the particles may be atoms, molecules, or even clusters of molecules.

Molecular-beam deflection, in which molecules' internal properties are inferred from changes to their trajectory in an applied external field, is a well-established method in physical chemistry. In contrast to classical machines, which typically realize molecular beam widths around 100 μm , our quantum interferometers can create molecular nanopatterns with periods as small as 80 nm. Beam shifts can therefore be seen with more than a thousand-fold increase in position sensitivity.

In 2007 we used a Talbot-Lau interferometer to determine the static polarizability of the fullerenes carbon-60 and carbon-70. With KDTL interferometry, it is now possible to study the influence of permanent and even dynamic electric dipole moments in a large variety of organic molecules.⁵ It will be possible to explore the magnetic world of molecules and clusters: aromaticity, magnetic dipole moments, and, eventually, phase transitions in clusters.

Most recently we have explored the possibility of using molecule interferometers to measure absolute optical absorption cross sections: The recoil imparted on each molecule due to the absorption of a single photon is sufficient to noticeably shift the

interference pattern.⁸ Such measurements can be used to analyze very dilute samples of species having unknown vapor pressure. Applied to medium-sized biomolecules with and without hydration shells, the technology could, in the future, uncover effects of solvation on structure, conformation, and photo-induced conformation changes.

Quantum interferometry with complex nanoparticles is still vast uncharted territory. Thousands of quantum objects, dozens of particle properties, and many new manipulation techniques remain to be explored. Often those quantities are intrinsically more dynamic in complex, warm molecules than they would be in static atoms.

Ongoing high-mass interference experiments at the University of Vienna allow handling of de Broglie wavelengths as small as 200 fm. Taking that as the current standard, a source that delivers nanoparticles of 10^6 atomic mass units (1.66×10^{-24} g) at a velocity of 1 m/s would be suitable for interferometry experiments. Various new sources are being developed to prepare tailor-made molecules and nanoparticles of 10^7 amu or more for interferometry experiments.

Quantum interferometry with biological nanomaterials is a fascinating short-term goal. Quantum physics with proteins or DNA having masses in the range 10 000–100 000 amu would open a new field of research and technology at the interface between quantum optics, physical chemistry, and biomolecular physics. The goal is challenging but not beyond reach: With the successful quantum interference last

year of functionalized organic molecules composed of 810 atoms (see figure 5), matter-wave interferometry has already crossed the 10 000-amu threshold.¹⁵

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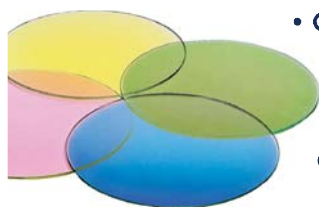
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