

In principle, a nanometer light source can be as small as a single molecule or atom. At the same time, its position and scanning have to be defined in space as well as those of an STM tip (a randomly flying atom does not qualify). Existing designs for optical supertips (5) are based on the same principle as the green plant photosynthetic system. A submicrometer antenna collects the photons by absorption and transfers the excitation energy to a single active center. From there, the energy is either (i) radiated as a photon or (ii) transferred to the sample in an energy transfer process (Foerster-Dexter) (5). In either case, the result is generally affected by the nearby sample molecule: (i) the radiated excitation may be affected, for instance, by intermolecular spin-orbit coupling (Kasha effect) (5, 18); (ii) the energy transfer results in a fluorescence or phosphorescence typical of the sample molecule. In the latter case, only virtual photons are produced by the supertip; this gives an excitation transfer tip ("exciton tip"), and only sample luminescence is detected. The world's largest ordered molecules, "dendrimers" (19), have been used or synthesized for this purpose. Such a single molecule exhibits a 125 Å antenna with an active center of 10 Å or less. So far, only tips with aggregates of sample molecules have been used [Tan *et al.* (1)].

Many technical problems still have to be solved, from understanding the contrast mechanism to the control of photobleaching (a standard problem in fluorescence microscopy). However, the future looks bright. Reversible bleaching (5) could be the basis for the highest density optical memories (with the "bit" occupying only a single molecule). More realistic in the near term would be pixels on the order of 100 Å. The necessary high scanning speeds are presently limited by the probe intensity, but we expect much higher photon outputs to become possible. Indeed, subwavelength probes have already been turned into high-flux lasers, as just reported by Betzig *et al.* (1). Analytical chemistry is being driven to the extreme of imaging single dye molecules and reaching absolute ion detection limits of zeptomoles or less. Complex polymeric samples may finally be characterized on the molecular level by nanospectroscopy. Maybe most importantly, biosamples, including living cells, could be imaged down to the molecular level and analyzed spectroscopically or by chemical sensors. The intracellular molecular dynamics of organogenesis, metabolism, splitting, and chemical damage could be followed in vivo and in real time. At the same time, DNA could be sequenced in situ (18) or even manipulated by the right probe at the right location. Although much of this might sound like science fiction, some of the above-mentioned

achievements in near-field optics must have sounded like science fiction only a couple of years ago.

References and Notes

1. Papers presented at the Second Near-Field Optics Conference, Raleigh, North Carolina, 18 to 20 October 1993; Special issue in *Ultramicroscopy*, in press.
2. E. Betzig and R. Chichester, *Science* **262**, 1422 (1993).
3. A. Lewis *et al.*, in *Optical Microscopy for Biology*, B. Herman and K. Jacobson, Eds. (Wiley-Liss, New York, 1990), p. 615; A. Lewis and K. Lieberman, *Anal. Chem.* **63**, 625A (1991).
4. E. Betzig, J. K. Trautman, T. D. Harris, J. S. Weiner, R. L. Kostelak, *Science* **251**, 1468 (1991); E. Betzig and J. K. Trautman, *ibid.* **257**, 189 (1992).
5. R. Kopelman and W. Tan, in *Spectroscopic and Microscopic Imaging of the Chemical State*, M. D. Morris, Ed. (Dekker, New York, 1993), p. 227.
6. D. W. Pohl, in *Advances in Optical and Electron Microscopy*, C. J. R. Sheppard and T. Mulvey, Eds. (Academic Press, London, 1990), p. 243.
7. B. Hille, *Ionic Channels in Excitable Membranes* (Sinauer, Sunderland, MA, 1984).
8. W. E. Moerner and L. Kador, *Phys. Rev. Lett.* **62**, 2535 (1989); L. Kador, D. E. Horne, W. E. Moerner, *J. Phys. Chem.* **94**, 1237 (1990); see also the review of single-molecule detection techniques by Th. Basche and W. E. Moerner, *Angew. Chem. Int. Ed. Engl.* **32**, 457 (1993).
9. M. Orrit and J. Bernard, *Phys. Rev. Lett.* **65**, 2716 (1990); see also _____ and R. I. Personov, *J. Phys. Chem.* **97**, 10256 (1993).
10. E. B. Shera, N. K. Seitzinger, L. M. Davis, R. A. Keller, S. A. Soper, *Chem. Phys. Lett.* **174**, 553 (1990).
11. W. B. Whitten, J. M. Ramsey, S. Arnold, B. V. Bronk, *Anal. Chem.* **63**, 1027 (1991).
12. K. Lieberman, S. Harush, A. Lewis, R. Kopelman, *Science* **247**, 59 (1990).
13. A. Lewis and K. Lieberman, *Nature* **354**, 214 (1991).
14. R. Toledo-Crow, P. C. Yang, Y. Chen, M. Vaez-Iravani, *Appl. Phys. Lett.* **60**, 2957 (1992).
15. D. Birnbaum, S. Kook, R. Kopelman, *J. Phys. Chem.* **97**, 3091 (1993).
16. S. Smith *et al.*, *SPIE* **1858**, 81 (1993).
17. W. Tan, Z.-Y. Shi, S. Smith, D. Birnbaum, R. Kopelman, *Science* **258**, 778 (1992); W. Tan, Z.-Y. Shi, R. Kopelman, *Anal. Chem.* **64**, 2985 (1992).
18. R. Kopelman *et al.* in *Hum. Gen. Prog., Human Genome Program Reports (1991-1992)* (U. S. Department of Energy, Washington DC, 1992), p. 130.
19. Z. Xu and J. S. Moore, *Angew. Chem.* **32**, 1354 (1993); Z. Xu *et al.*, *Polymer Prepr. Am. Chem. Soc. Div. Polym.* **33**, 130 (1993).
20. We acknowledge the financial support from National Science Foundation grant DMR-9111622 and Department of Energy grant DE-FG02-90ER 61085.

Neutron Interferometry

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As quantum objects with both wave and particle properties, neutrons can exhibit familiar optical effects such as diffraction and interference. In the two decades since the first perfect-crystal neutron interferometer was tested by an Austrian-German group at our 250-kw TRIGA (Training Reactor, Isotopes General Atomic) reactor in Vienna (1), neutron interferometry has become a laboratory for fundamental tests of quantum mechanics. Neutrons are fermions of well-defined mass and are subject to strong electromagnetic and gravitational interactions, all of which cause measurable interference effects. When placed in a magnetic field, neutrons occupy two energy levels between which transitions can be induced by proper oscillating magnetic resonance fields; the existence of these energy levels increases the variety of quantum mechanical tests that can be performed.

To perform interferometry, separate but phase-coherent neutron beams are needed. Such beams can be produced by dynamical Laue-reflection of thermal neutrons in an appropriately shaped perfect silicon crystal. This is analogous to the Mach-Zehnder

type of interferometer used in light optics and to the Bonse-Hart interferometers (2) developed for x-rays; electron interferometry (3) and the recently developed atom interferometry (4, 5) use somewhat similar schemes. According to the complementarity principle of quantum mechanics, the neutron behaves purely as a wave inside the interferometer.

Symmetry dictates that the wavefunctions originating from beam paths I and II and composing the forward beam (0) behind the interferometer are equal in amplitude and phase because they are transmitted-reflected-reflected (TRR) and reflected-reflected-transmitted (RRT), respectively. Therefore, complete beam modulation is expected as a function of the phase shift between the beams:

$$I \propto |\psi_0|^2 + |\psi_0|^2 \propto 1 + \cos \chi \quad (1)$$

Different kinds of interactions can cause phase shifts between the coherent beams, which can be calculated as the path integral of the canonical momentum $\mathbf{k}_c$ along the interferometer loop, $\chi = \oint \mathbf{k}_c \cdot d\mathbf{s}$.

Neutron interference experiments belong to the domain of self-interference where, in nearly all cases, only one neutron is inside the interferometer, while the next one has yet to be born and is still contained

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in the fuel of the nuclear reactor. Although there is no interaction between different neutrons, they have a certain common history within the limits defined by, among other things, the neutron moderation process, their movement along neutron guide tubes, the monochromator crystal, and the particular interferometer setup. Any real interference pattern therefore contains single particle as well as ensemble properties and, hence, deviates slightly from the ideal interference pattern.

Soon after the successful operation of perfect-crystal interferometers, researchers were able to verify the 4π -symmetry of spinor wavefunctions, a feat achieved independently in Europe and the United States (6, 7). When a neutron travels through a magnetic field, its wavefunction is modified by the coupling between the field and the neutron's magnetic moment. This wavefunction has a period of 4π (and not 2π), which shows up in the interference pattern, in excellent agreement with the experimental observation. The 4π periodicity effect has been observed for unpolarized and polarized neutrons as well, which demonstrates the self-interference properties of this kind of experiment—the intrinsic feature of this phenomenon—and that single-particle properties and not only ensemble ones are described by the wave function (8).

In an extension of this kind of investigation, the quantum mechanical law of spin-superposition has been verified on a macroscopic scale by means of a polarized beam of incident neutrons split coherently into two beams, the polarization of one beam then being inverted. The wavefunction for the forward beam is then $\psi \propto \psi^I + \psi^{II} \propto (|z\rangle + e^{i\chi}| -z\rangle)$ and the final polarization is in the xy plane (that is, perpendicular to both states before superposition) (9). Wigner (10) pointed out that, in this case, a pure initial state in the $|z\rangle$ direction is transferred into a pure state in the $|x\rangle$ direction ($\chi = 0$), although in one beam path, no spin reversal ($|z\rangle$) occurs, and in the other one, a complete spin reversal ($| -z\rangle$) occurs.

If the spin reversal is accomplished by a Rabi-type resonance flipper, the total energy changes by the amount of the Zeeman energy $2\mu B_0 = \omega_L$, generating a time-dependent phase shift ($\omega_L t$) and causing the final polarization to rotate in the xy plane with the Larmor frequency ω_L , despite the absence of a magnetic field (11). A slight modification of this arrangement permitted a verification of the magnetic Josephson effect (12). In this case, Rabi flippers are operated in both beam paths but with slightly different Larmor resonance frequencies $\omega_L^{(1)}$ and $\omega_L^{(2)}$, owing to slightly different guide fields. This causes a temporal beam modulation in the form of a quantum beat effect, where the time-dependent phase

shift is driven by the magnetic potential

$$\Delta(t) = (\omega_L^{(1)} - \omega_L^{(2)})t = 2\mu\Delta B_0 t/\hbar \quad (2)$$

In a superconducting tunnel junction, by contrast, the phase shift is driven by the electric potential

$$\Delta_j(t) = 2eVt/\hbar \quad (3)$$

The observed period of the beam modulation and the energy sensitivity are connected by the quantum mechanical uncertainty relation. The whole theoretical treatment is closely connected to the Berry phase formalism (13) and to the scalar Aharonov-Bohm effect (14), which has been observed in neutron interference experiments recently (15). In all these cases, the canonical momentum—not the kinetic one ($\hbar k = mv$)—changes, and therefore, no classical force acts on the particle.

The coupling of the neutron to the gravitational field has also attracted interest as it offers a way of testing some of the connections between gravity and quantum fields. The effect of the Earth's gravitational field manifests itself when the interferometer crystal is rotated around a horizontal axis in such a way that one beam path experiences a higher gravitational potential than the other (16). The induced phase shift therefore contains terms of both gravitational and quantum mechanical origin. Neutron interferometry also permitted the observation of the Sagnac effect, which is the phase shift between two paths oriented in opposite directions about the Earth's rotation axis (17). In this case, the interferometer crystal was rotated around a vertical axis, and the phase shift then depends on the colatitude of the site of the experiment and the Earth's rotation frequency.

Many neutron interference experiments have been performed to investigate the analogy between light and matter wave optics and to establish neutron quantum optics as a tool for both fundamental and applied research. In a matter-wave interferometer, the coherence function for the two beams (after splitting) is defined by the autocorrelation function of the overlapping wave functions as $\Gamma(\Delta) = \langle \psi^*(0) \cdot \psi(\Delta) \rangle$. The spatial shift Δ of the overlapping wavefunctions is nonzero only when the kinetic momentum in one limb of the interferometer has changed ($\chi = \Delta \cdot k$). In such experiments, one is able to observe high-order (up to 1000) interferences, but they are damped because of the finite coherence lengths of the beams. Recent experiments have demonstrated that interference phenomena can be restored by means of a post-selection element that narrows the spectral distribution after the two beams have recombined (18). In this case, the interference pattern disappears, but in momentum space, a significant spectral modulation ap-

pears (19). This arises from a superposition of two macroscopically distinguishable states (in essence, stationary Schrödinger cat-like states), separated in ordinary space but oscillatory in momentum space and apparently extremely fragile and sensitive to any kind of dephasing effects. Such experiments are closely connected to the recent progress in quantum measurement theory (20). These states exhibit, for certain parameters, squeezing phenomena which can be further enhanced by multiplate systems.

Experiments in neutron interferometry have shown not only that expectation values are measurable quantities but that several properties of single-particle wavefunctions can be measured too. A detailed analysis also shows that the entire history of each particle is stored in its wavefunction and becomes measurable if experiments with sufficiently high resolution are feasible. Neutron interferometry has contributed substantially to the understanding of the wave-particle duality of various topological phenomena and has pushed the sensitivity of such experiments to the quantum limit. The experimental tests have shown that quantum theory is still a lively theory, and one may better appreciate the pioneering work of the founders who created it with so little experimental evidence. Although this work provides much more direct evidence, even on macroscopic scales, the interpretation of quantum mechanics in many cases still goes beyond human intuition.

References and Notes

1. H. Rauch, W. Treimer, U. Bonse, *Phys. Lett. A* **47**, 369 (1972).
2. U. Bonse and M. Hart, *Appl. Phys. Lett.* **6**, 155 (1965).
3. G. Möllenstedt and H. Düker, *Naturwissenschaften* **42**, 41 (1954).
4. O. Carnal and J. Mlynek, *Phys. Rev. Lett.* **66**, 2689 (1991).
5. D. W. Keith, C. Ekstrom, Q. A. Turchette, D. E. Pritchard, *ibid.*, p. 2692.
6. H. Rauch, *et al.*, *Phys. Rev. A* **54**, 425 (1975).
7. S. A. Werner, R. Colella, A. W. Overhauser, C. F. Eagen, *Phys. Rev. Lett.* **35**, 1053 (1975).
8. Y. Aharonov, J. Anandan, L. Vaidman, *Phys. Rev. A* **47**, 4616 (1993).
9. J. Summhammer, G. Badurek, H. Rauch, U. Koschko, A. Zeilinger, *ibid.* **27**, 2523 (1983).
10. E. P. Wigner, *Am. J. Phys.* **31**, 6 (1963).
11. G. Badurek, H. Rauch, J. Summhammer, *Phys. Rev. Lett.* **51**, 1015 (1983).
12. G. Badurek, H. Rauch, D. Tuppinger, *Phys. Rev. A* **34**, 2600 (1986).
13. M. V. Berry, *Proc. R. Soc. London Ser. A* **392**, 45 (1984).
14. Y. Aharonov and D. Bohm, *Phys. Rev.* **115**, 485 (1959).
15. B. E. Allman *et al.*, *Phys. Rev. Lett.* **68**, 2409 (1992).
16. R. Colella, A. W. Overhauser, S. A. Werner, *ibid.* **34**, 1472 (1975).
17. J. L. Staudenmann, S. A. Werner, R. Colella, A. W. Overhauser, *Phys. Rev. A* **21**, 1419 (1980).
18. H. Kaiser, R. Clothier, S. A. Werner, H. Rauch, H. Wöhlitsch, *ibid.* **45**, 31 (1992).
19. D. L. Jacobson, S. A. Werner, H. Rauch, *ibid.*, in press.
20. W. H. Zurek, *Phys. Today* **44** (no. 10), 36 (1991).