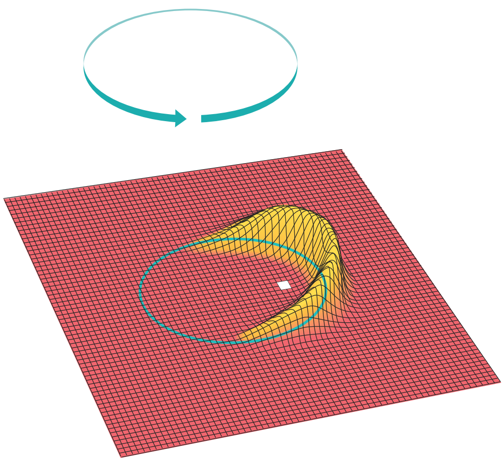
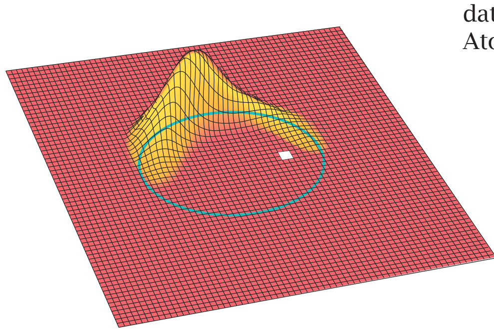
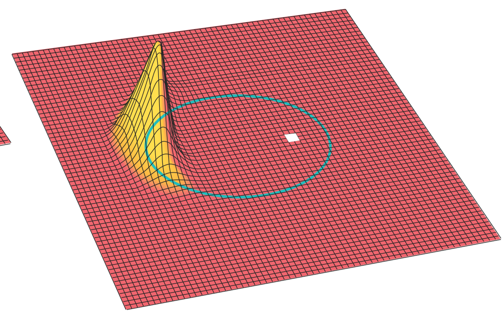
The Classical Limit of an Atom

By creating ultralarge atoms, physicists hope to study how the odd physics of the quantum world becomes the classical mechanics of everyday experience

by Michael Nauenberg, Carlos Stroud and John Yeazell

CLASSICAL ORBITAL MOTION can emerge from a quantum-mechanical object called a wave packet, which defines the probable location of an electron. The series of plots shows how the localized wave packet traces an elliptical orbit around the point where the nucleus resides (*white dots*). Note that the wave packet has begun to disperse after completing one revolution.

Throughout this century, physics has made use of two quite different descriptions of nature. The first is classical physics, which accounts for the motion of macroscopic objects, such as wheels and pulleys, planets and galaxies. It describes the continuous, usually predictable cause-and-effect relationships among colliding billiard balls or between the earth and orbiting satellites. The second description is quantum physics, which encompasses the microscopic world of atoms, molecules, nuclei and the fundamental particles. Here the behavior of particles is described by probabilistic laws that determine transitions between energy levels and govern tunneling through energy barriers. Because quantum mechanics is the fundamental theory of nature, it should also encompass classical physics. That is, applied to macroscopic phenomena, quantum mechanics should reach a limit at which it becomes equivalent to classical mechanics.

Yet until recently, the exact nature of this transition had not been fully elucidated. Now that goal is within reach. Atomic systems have been created that behave—for a short period—according to the laws of classical mechanics. Researchers fabricate such systems by exciting atoms so that they swell to about 10,000 times their original size. On such a scale the position of an electron can be localized fairly closely; at least its orbit no longer remains a hazy cloud that represents only a probable location. In fact, as the electron circles the nucleus, it traces an elliptical path, just as the planets orbit the sun.

The importance of understanding the classical limit of an atom takes on new meaning in the light of modern technology, which has blurred the distinctions between the macroscopic and microscopic worlds. The two domains had remained largely separate; a scientist would use classical mechanics to predict, say, the next lunar eclipse and then

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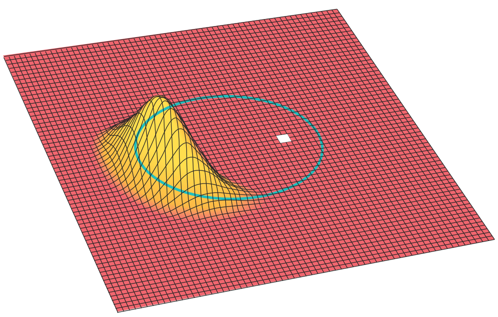
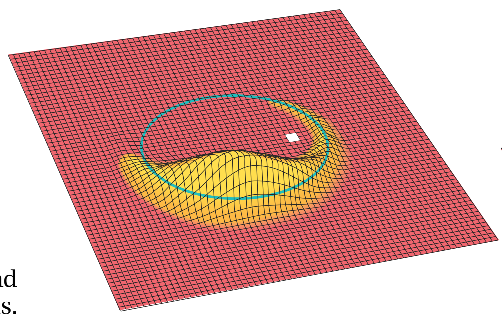
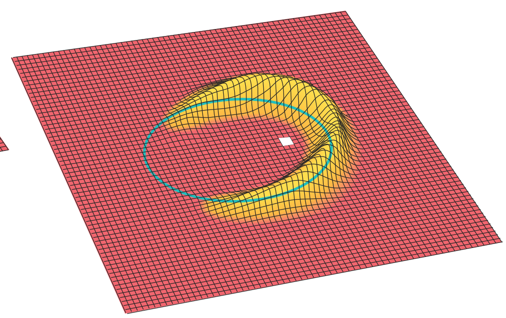
switch to quantum calculations to investigate radioactive decay. But engineers now routinely construct computer chips bearing transistors whose dimensions are smaller than one micron. Such devices are comparable in size to large molecules. At the same time, a new generation of microscopes can see and even manipulate single atoms. Finding the best way to exploit these technologies will be aided by the understanding we obtain from studies of the classical limit.

The profound differences between the quantum world and the classical domain emerged around the turn of the century. Experiments by such great scientists as Ernest Rutherford, the New Zealand–born physicist who worked at the University of Cambridge, established that the atom consists of a pointlike positive charge that holds negatively charged electrons. To early investigators, this arrangement mirrored the solar system. Indeed, the force that holds the electrons to the nucleus—called the Coulomb force—varies with the inverse square of the distance, as does the gravitational force.

This simple planetary model did not prove satisfactory. According to classical electromagnetic theory, any electric charge moving in a closed orbit must radiate energy. Thus, the electron in an







elliptical orbit should quickly expend its energy and spiral into the nucleus. All matter would therefore be unstable. Furthermore, the radiation that an electron emitted as it plunged into the nucleus would have a continuous spectrum. But experiments indicated that electrons emit radiation in flashes, yielding a spectrum of discrete lines.

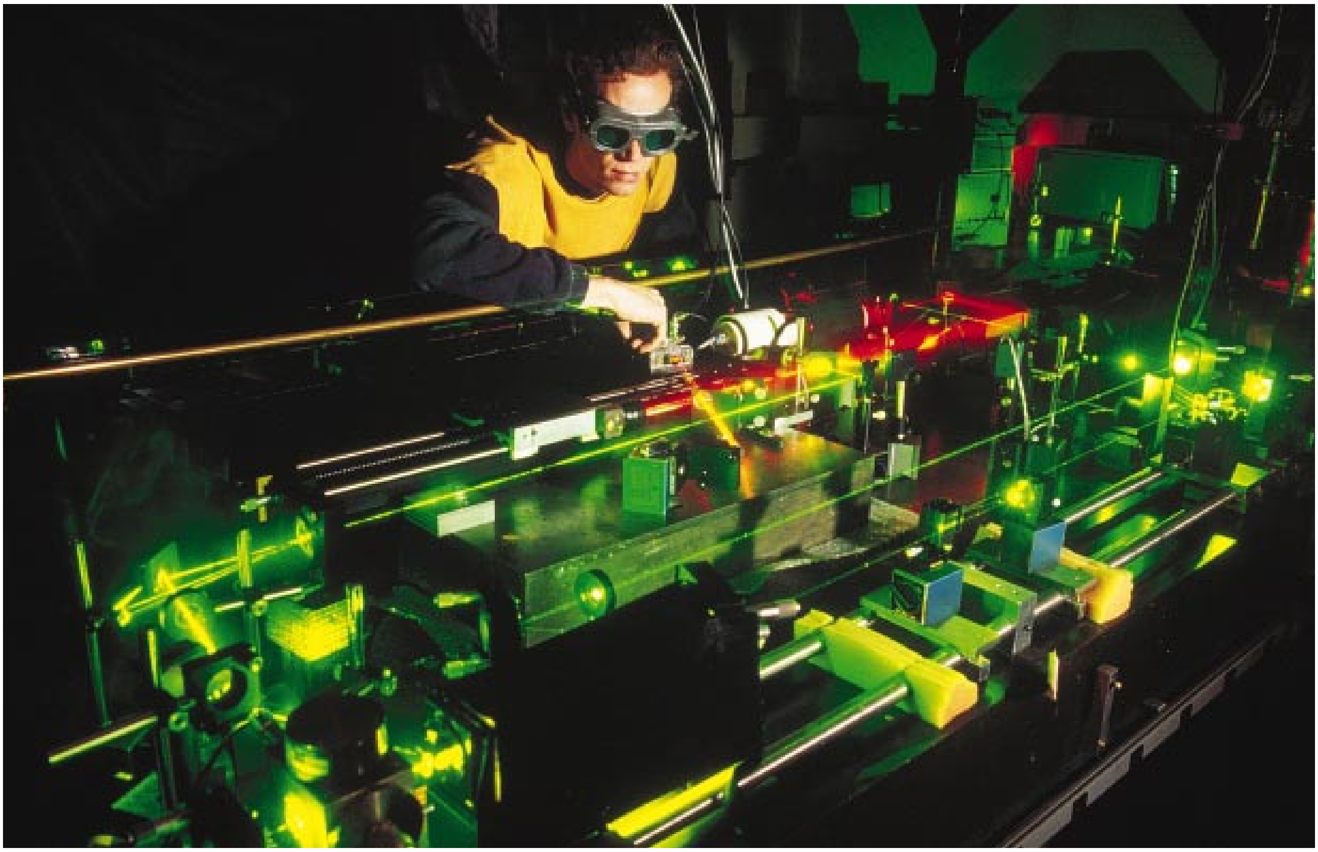
The Danish physicist Niels Bohr resolved some of these difficulties by augmenting the classical physics of the planetary model of the atom with a set of constraints. They were based on a theory about the nature of radiation first introduced by the German physicist Max Planck, who found that radiation is emitted in discrete units (the energy of which depends on the fundamental parameter now known as Planck's constant, h). Bohr retained the notion of classical orbits but assumed that only certain discrete values of energy and angular momentum were permitted. An integer, called a principal quantum number, characterized each energy state that an electron could occupy while associated with a nucleus. For example, the ground state was numbered one, the first excited state numbered two, and so on. Other quantum numbers describe a particle's angular momentum, which according to Bohr's theory would occur only in integer multiples of Planck's fundamental constant. Electrons could make transitions between orbits in the form of "quantum jumps." Each jump gave off a distinct frequency of light, which equaled the difference in energy between the two orbits divided by Planck's constant. The frequencies predicted in this way agreed completely with the observed discrete spectra of light emitted by hydrogen.

Bohr also postulated a rule that identified the classical limit of his quantum theory. This rule is named the correspondence principle. It states that for large quantum numbers, quantum theory should merge into classical mechanics. This limit corresponds to physical situations in which the classical action is much larger than Planck's constant. Therefore, it has become customary to refer to the classical limit as the scale at which Planck's constant vanishes. Bohr's correspondence principle has remained as a basic guideline for the classical limit of quantum mechanics, but as we shall see, this principle, while necessary, is not sufficient to obtain classical behavior.

The Bohr-Rutherford model successfully explained the characteristics of hydrogen. But it produced difficulties and inconsistencies when applied to the behavior of more complicated atoms and to the properties of molecules. The German physicist Werner Heisenberg surmised that to make further progress, a quantum theory of atoms should be based only on directly observable quantities, such as the well-known spectral lines mentioned above. He believed certain concepts of classical physics, such as the electronic orbits that Rutherford and Bohr used, had to be completely discarded. He wrote to his Austrian colleague Wolfgang Pauli that these orbits do not have the slightest physical significance. Indeed, his matrix formulation of quantum mechanics did away with electron orbits entirely. Heisenberg accounted for the frequency and magnitude of the discrete spectral lines in terms of Planck's constant and other fundamental values in nature. Independently, the Austrian physicist Erwin Schrödinger derived an alternative but equivalent formulation. Following ideas of the French physicist Louis de Broglie, he represented physical systems with a wave equation. Solutions to this equation assigned probabilities to the possible outcomes of a system's evolution.

Whereas Heisenberg felt that classical orbits had no place in quantum theory and should be abandoned, Schrödinger was of a different mind. From the start he was concerned with the relation of the microscopic to the macroscopic world. Classical dynamics, he believed, should emerge from his wave equation. As a first step, Schrödinger investigated a very simple kind of system, called the harmonic oscillator. This system is not exactly that of an orbiting body; it corresponds to the up-and-down motion of a block hanging from the end of a spring. The harmonic oscillator shares a crucial feature of an orbit around a Coulomb or gravitational potential: periodicity. Such an orbiting body repeats its motion once each cycle—the period of the earth's orbit is just a year. The suspended block also has a cycle: it completes one up-down action over some unit of time.

Schrödinger managed to extract clas-



sical behavior from his theory for a harmonic oscillator. He did so by constructing a solution for his equation that was a sum of solutions that had discrete energy values. Graphically, these solutions resemble sinusoidal waves of different frequencies. Superposing such waves produced a "Gaussian wave packet," which looks like a bell-shaped curve. The remarkable property of this wave packet was that it remained localized around a center that executed classical, periodic behavior. Schrödinger, however, failed to derive similar classical motion for more complex cases, such as the movement of an electron in the hydrogen atom.

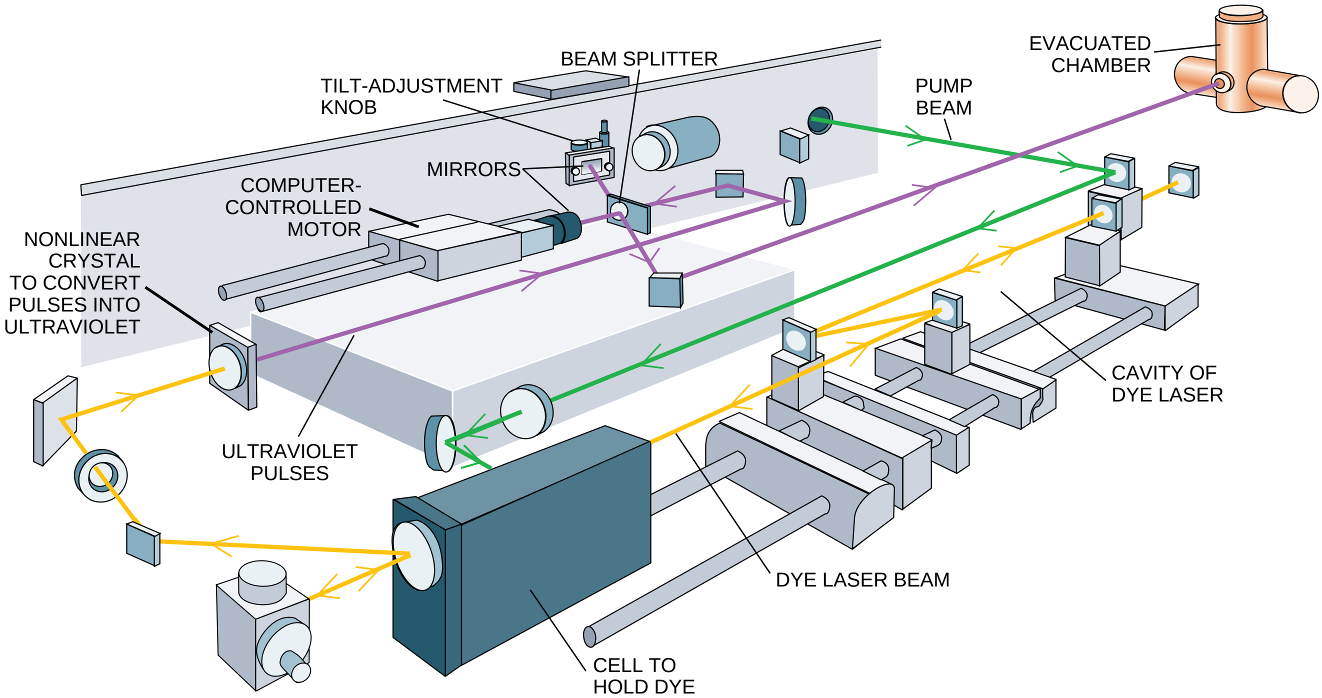
On the face of it, formulating a classical wave-packet description for an electron associated with an atom would not seem to be difficult. One would similarly choose appropriate atomic energy states, find their wave solutions and then superpose them. The problem lies in the way energy states are actually separated. A theorem developed by the French mathematician Jean-Baptiste Fourier indicates that only energy levels that are equally spaced with respect to one another can be combined to form a coherent state that moves periodically. But in an atom, the adjacent energy states are not equally spaced. For example, the energy separating the ground state from the first excited state is extremely large compared with the energy gaps at high quantum numbers: the first gap is one million times greater than that separating energy states whose quantum numbers are 100 and 101. A wave packet made up of a superposition of states near the ground state therefore disperses shortly after its creation. Obviously, a classical atom cannot be constructed from such states.

As Bohr noted, the key to achieving classical correspondence is to work with high-energy states, which have large quantum numbers. The energy separating these adjacent states is proportional to the inverse cube of the principal quantum number. That means, for large quantum numbers, the energy spacings between adjacent states are almost equal. In this limit, the spatial localization should persist for some time, permitting the center of the wave packet to evolve in a classical manner. Thus, the bigger the quantum numbers used, the easier it should be to produce a relatively stable, classical atom.

Until recently, no experimental device existed by which researchers could test the proposition by creating a superposition of excited atomic states in the laboratory. The development of lasers that can deliver short, powerful pulses of light proved to be the answer. By means of such devices, researchers formed the first localized wave packets in atoms during the late 1980s. Among the successful groups were ours at the University of Rochester, Ben van Linden van den Heuvell and his colleagues at the FOM Institute of Atomic and Molecular Physics in Amsterdam and Paul Ewart and his co-workers at the University of Oxford.

In a typical experiment, a brief, ultraviolet pulse of laser light lasting a mere 20 picoseconds (20 trillionths of a second) intersects a beam of potassium atoms in an evacuated chamber. Potassium is used because it readily absorbs the energy from our lasers, and, like hydrogen, it has one electron available for bonding. Each pulse excites an electron from a single ground state to many very high states. The result is a wave packet localized at a distance of about one micron from the nucleus.

Laser pulses of picosecond duration are essential because short bursts have a broad spectrum of frequencies. The spectral width of such a coherent pulse is proportional to the reciprocal of its duration, so that a pulse with a spectrum wide enough to overlap many levels must be extremely short. Traditional spectroscopy relies on long pulses, which contain a narrow band of frequencies and so excite only one or a few states. In our experiments, the average quantum number excited was 85, and about five states were superposed.



REACHING THE CLASSICAL LIMIT demands the excitation of atoms by brief pulses of laser light. A green laser beam emerges from behind the right side of the partition. It "pumps" a dye laser, which then produces yellow pulses (it appears faint green in the photograph on the opposite page). The nonlinear crystal converts the yellow light into ultraviolet (*invisible in photograph*). A beam splitter separates each ultraviolet pulse into two parts that move along different paths. A computer-controlled motor can alter the length of one path by shifting a mirror. Such adjustments allow one pulse to lag behind the other: a 0.3-millimeter increase produces a one-picosecond delay. The beams are recombined and directed at atoms in an evacuated chamber. The first pulse excites the atoms; the second pulse probes the result. The red and orange beams, used to maintain mirror alignments, and some components have been omitted from the diagram for clarity.

We probed the characteristics of our wave packet by measuring how it absorbs energy from a second laser pulse, fired shortly after the first one. At the perigee of its orbit, the wave packet absorbs the most energy. In fact, the amount of energy absorbed is sufficient to tear the electron away from the atom. Thus, to map out the electron's orbit, we simply measured the number of atoms that were ionized as we varied the delay between the two laser pulses. The ionization signals correspond to the expected oscillation of the wave packet as it periodically moves through the perigee of its orbit.

This method excites orbits of a fairly well defined energy and angular momentum. It does not select the orientation of the orbits. Instead the wave-packet state resides in the form of a statistical ensemble of classical orbits. Each member of the ensemble possesses the same radius and eccentricity but occupies all possible orientations in space. This superposition is well localized only in the radial dimension—that is, at a particular time, its distance from the nucleus is about as well determined as Heisenberg's uncertainty principle permits. Hence, investigators have christened this object a radial wave packet.

The motion of the radial wave packet contains many classical elements. The wave packet evolves from the nucleus toward the edge of a classical orbit and then returns. The period of this oscillation is just that of an electron following a classical elliptical orbit about the nucleus. Moreover, the wave packet moves most slowly at the apogee of its circuit and most quickly at the perigee, just as do comets and other orbiting bodies in their paths around the sun.

In forming a radial wave packet, we created a state that exhibits strong classical characteristics. Our goal, however, was to form a classical atom. In that regard, the radial wave packet has a shortcoming. Despite the classical orbital period of its oscillations, the packet follows a planetary trajectory only in a statistical sense. An electron in such a wave packet traces orbits that are oriented at all angles in space. In effect, the particles move about in a spherical shell wrapped around the nucleus [*see upper left illustration on next page*]. Obviously, this picture is not equivalent to that of a planetary system, in which the major axis of the ellipse describing the motion of a planet is (approximately) fixed in space. Furthermore, the wave packet spreads as it propagates radially, a behavior comparable in classical physics to a planet breaking up as it moves in its orbit.

Jean Claude Gay, Dominique Delande and Antoine Bommier of the École Normale Supérieure in Paris and one of us (Nauenberg) recently propounded a detailed theory that shows how to construct a wave packet that is oriented in a particular direction in space. We found that, for large quantum numbers, a stationary solution of Schrödinger's equation exists that amounts to an "elliptical stationary state." This state is unusual. A conventional atomic state has a discrete energy value and a range of angular momenta [see "Highly Excited Atoms," by Daniel Kleppner, Michael G. Littman and Myron L. Zimmerman; SCIENTIFIC AMERICAN, May 1981]. The elliptical stationary state, however, consists of a well-defined linear superposition of these ordinary atomic states that centers within a spread of angular momenta. The eccentricity of the corresponding elliptical orbit determines the spread. The square of the magnitude of the wave function gives the probability for finding the electron at a particular position. Graphically, this probability appears as a bump on the orbit, representing the maximum value of the wave function [*see upper right illustration on next page*].

Classical arguments explain the presence of the bump. The quantum-mechanical state is analogous to an ensemble of electrons traveling on classical orbits. Because their velocity is at a minimum at apogee, the electrons will tend to bunch up there. The bunching yields the bump on a graphical representation of the elliptical state. It represents the region in which the electron is most likely to be found. Making the elliptical stationary state in the laboratory is substantially more complicated than forging a radial wave packet. A short pulse of laser light that excites an atom is not enough. The set of states needed to form the elliptical state turns out to require a superposition of many angular momentum states rather than many energy states. The laser beam cannot directly excite such a superposition. An additional field must be applied simultaneously with the laser pulses. Several solutions have been proposed. Two of us (Stroud and Yeazell) have excited such a state by employing a strong radio-frequency field in conjunction with a short optical pulse.

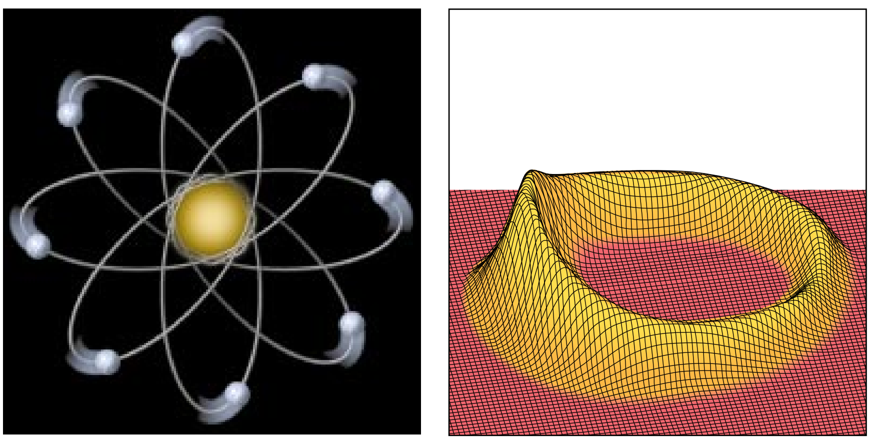
Although this elliptical state incorporates a definite angular orientation, it is stationary. It does not evolve in time. The final step in producing a classical state of the atom consists of making the wave packet move along the elliptical path [*see illustrations on pages 44 and 45*]. Although we have created such a wave packet as a solution of the Schrödinger equation on the computer, to date no one has succeeded in producing this state in the laboratory.

The theoretical wave packet we constructed is the most nearly classical state we know how to make. It shows striking classical properties but also maintains an underlying quantum-mechanical nature. As the wave packet moves around the elliptical path, it manifests one of its most obvious quantum properties. On each successive orbit, the wave packet spreads, a behavior akin to a classical group of electrons in which each particle moves at a different speed. Such a group would continue to spread indefinitely. But for the wave packet, a phenomenon quite distinct from classical behavior appears: quantum interference. This effect happens once the wave packet's head catches up to its tail and begins to interfere with it. Then, surprisingly, at a later, well-defined time, the wave packet reconstitutes itself, a behavior that does not have any classical analogue whatsoever. In between these full revivals, the state of the electron cannot be described as a single, spatially localized wave packet.

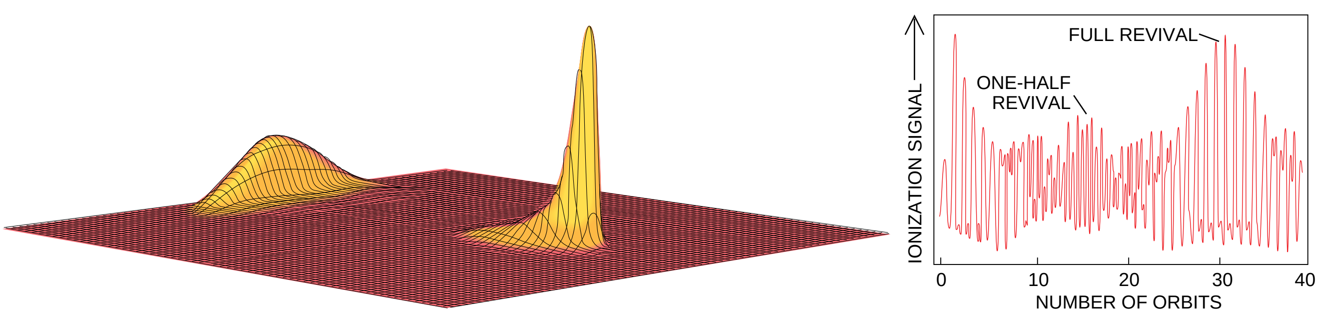
Indeed, windows in time exist in which the wave packet is localized in more complex structures. They constitute miniature replicas of the original wave packet that move classically as they maintain uniformly spaced positions on the orbit. These moments have been characterized as fractional, or partial, revivals. At a stage called the one-half revival, the wave packet has split into two smaller ones. Likewise, at the one-third revival, it has broken up into three packets, and so on. A classical particle by definition cannot spontaneously fracture and revive in this way, but a quantum particle can—and does.

A classical analogy can explain many features of the quantum revivals. In particular, they can be likened to the bunching of runners on a racetrack. The runners represent the ensemble of electrons we use to imitate the quantum state. The racetrack contains a set of discrete classical orbits that satisfy Bohr's quantum conditions. At the beginning of the race, the runners line up at the start—that is, they are well localized. Each one runs in one of the quantized Bohr orbits. During the initial laps, the runners remain closely bunched. But after a few circuits, the runners have begun to spread around the track. It is not the quantum constraints or discreteness that causes this initial spreading. It is simply that the wave packet consists of a collection of waves of varying frequencies—a group of runners moving at different speeds.

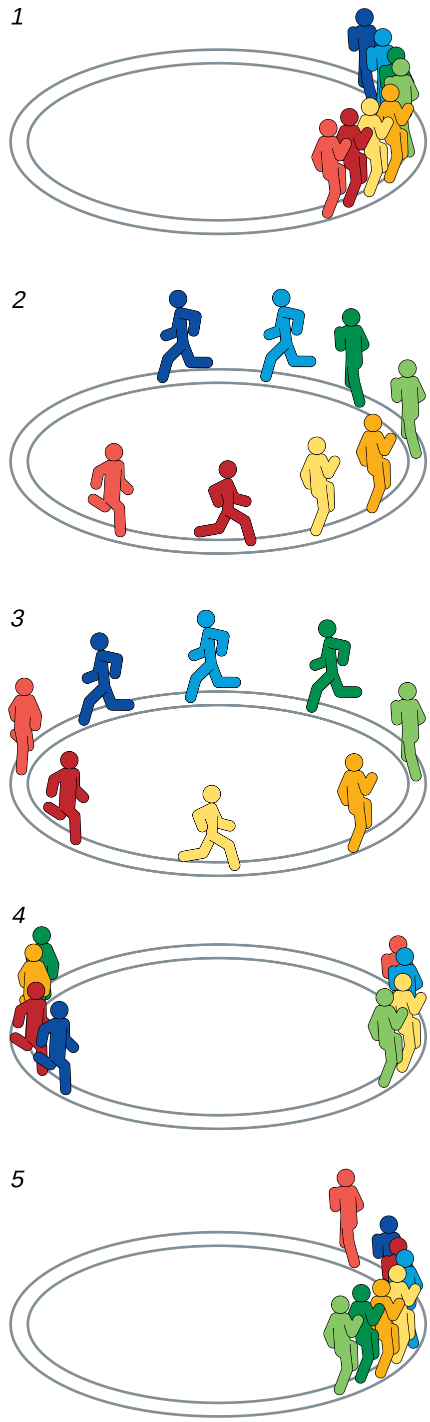
The quantum features begin to appear when the racers start to clump—that is, when the fastest runner catches up to the slowest runner. Further into the race, the quicker runners continue to pass the slower runners. Occasional-



ENSEMBLE OF CLASSICAL ORBITS (*left*) is one way to describe a radial wave packet. The packet consists of a superposition of several energy levels; in effect, an electron moves simultaneously in many orbits that surround the nucleus. A more planetlike behavior would have the orbits lie in one plane. Such a state, called the elliptical stationary state, has been created (*right*). The bump on the left side represents the most likely location of the electron.



ONE-HALF REVIVAL of a wave packet (*left*)—that is, the formation of two smaller packets after the original has dispersed—takes place after about 15 orbits. It is indicated by ionization signals that appear twice as frequently (*right*). After about 30 orbits, the ionization signal returns to its original value, showing that the wave packet has fully revived.



RUNNERS ON A TRACK can portray the wave-packet revivals. At the start (*1*), the runners are bunched together, representing a well-localized wave packet. During the course of the race, the faster runners pull ahead (*2*); soon they begin to lap the slower competitors (*3*). Eventually two clumps of runners form (*4*), corresponding to a one-half revival. After many more circuits, they clump back into a single group (*5*). A problem with this model is that the full revival actually takes place on the side of the track opposite from the location of the clumped runners.

ly several runners may form a clump. Because of the particular distribution of speeds allowed by the quantum constraints, there is a moment when the runners form two clumps on opposite sides of the track. This clumping corresponds to the one-half fractional revival. Quantum constraints sort the runners into groups, so that one pack contains all the odd-numbered runners and the other all the even-numbered runners.

As the race continues, the runners spread out and eventually clump again, but into three groups. Finally, after many circuits, each runner has run a full lap farther than the next slower runner, so a full revival occurs. The number of such fractional revivals depends on the number of runners in the race. It requires at least two runners to form a clump. Similarly, in the atom the number of fractional revivals depends on the number of levels in the superposition. Neither the fractional nor full revivals would appear in this classical model without the imposition of the quantum constraints that place the runners into discrete orbits.

Investigations into this realm of physics have shown that despite Heisenberg's attempt to banish them, classical orbits remain a part of modern quantum mechanics. But their role is far more subtle than even Bohr realized. Wave packets that travel on classical trajectories are not produced by simply letting the quantum numbers of the system become large. Rather the formation of a special coherent superposition of states that have large quantum numbers is necessary for a wave packet to demonstrate two hallmark classical features: spatial localization and motion along an orbital path. These classical actions persist for only a limited period. For longer times, the underlying quantum dynamics manifests itself in previously unexpected wave phenomena that have no classical analogy.

Such results may best be understood in terms of theories that incorporate classical dynamics into quantum mechanics. Such semiclassical techniques are invaluable because conventional quantum-mechanical calculations are difficult and time-consuming, even on the largest supercomputers. Moreover, by themselves the resulting numerical solutions often cannot be understood or interpreted physically.

Although semiclassical methods have been used for a long time, especially in descriptions of a quantum system's energy, they have only recently been extended successfully to the time domain. They can now predict quantum behavior, even under nonlinear, or chaotic, circumstances. For example, Eric J. Heller of Harvard University and Steven Tomsovic of the University of Washington studied the motions of a wave packet trapped inside a "box." They showed that semiclassical methods describe the packet's chaotic motions as well as quantum calculations do. Such schemes also promise to illuminate other topics associated with quantum chaos that have received much attention lately. Among them are the microwave ionization of atoms and the behavior of atoms in strong electromagnetic fields.

Of course, short intense laser pulses can excite systems other than atoms. When a molecule is excited this way, its atoms can form wave packets. Presumably an appropriate tailoring of the laser pulse could control the internal dynamics of the molecule [see "The Birth of Molecules," by Ahmed H. Zewail; SCIENTIFIC AMERICAN, December 1990].

These techniques have also been used to form wave packets of electrons, or even positively charged holes, in semiconductor quantum wells. The coherent oscillations of the wave packets can then produce novel devices that cannot be made with more conventional means of excitation. Such devices would be bonuses that come packaged with the fundamental information we seek at the classical limit of quantum mechanics.

FURTHER READING

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