

A molecular merry-go-round

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For almost 80 years the marvels of the subatomic world have been revealed through collisions of charged particles confined in circular accelerators. Now we are beginning to build analogous machines that confine bunches of neutral molecules.

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John Fenn's foreword to Atomic and Molecular Beam Methods (volume 1, Oxford University Press, 1988) included the following testimonial about molecular beams: "Born in leaks, the original sin of vacuum technology, molecular beams are collimated wisps of molecules traversing the chambered void that is their theatre. . . . On stage for only milliseconds between their entrances and exits, they have captivated an ever growing audience by the variety and range of their repertoire." The tiny millisecond performance time results from the molecules moving quickly—hundreds of meters per second—and acting in a 1-m-long vacuum chamber. In a molecular synchrotron, however, slowed neutral molecules travel inside a circular 1-m vacuum chamber for more than a mile and thus stretch the duration of their performance by a factor of 10 000. The molecular synchrotron was developed by Gerard Meijer and coworkers, who began their work a decade ago at the FOM Institute for Plasma Physics Rijnhuizen in the Netherlands.

Circular reasoning

Dealing with circular motion has traditionally been the domain of cyclotrons, synchrotrons, and other charged-particle carousels. The simplest of those devices, the cyclotron, relies on incremental, multistage acceleration by moderate voltages. Gustaf Ising conceived the idea in 1924, albeit with a linear arrangement of electrodes; four years later Rolf Widerøe realized Ising's proposal. At about the same time, Ernest Lawrence recognized that charged particles held in orbit by a magnetic field will maintain the same orbital period even when accelerated by a superposed electric field applied across a fixed electrode pair. That's because both the velocity and the radius of the particles' cyclic motion increase after each traversal of the electric field. The sight of cyclic wine-glass imprints triggered a congenial contemplation of the idea by Lawrence and Otto Stern during a 1929 carousal in a Boston speakeasy. Stern urged Lawrence to "get to work on that" right away, and in 1932, Lawrence conducted the first cyclotron experiments at the University of California, Berkeley. That was the annus mirabilis of nuclear physics, whose beginning was marked by the splitting of lithium nuclei at the Cavendish Laboratory at Cambridge University with 700-keV protons energized by a single-stage electrostatic accelerator. A few weeks later, Lawrence used 1-MeV protons to confirm and extend the Cavendish result.

The cyclotron uses an oscillating electric field, typically an RF field, to accelerate charged particles. The constancy of the orbital period ensures that a particle always encounters the same phase when it gets a momentum kick. As a result, a cyclotron does not need to track where the accelerating par-

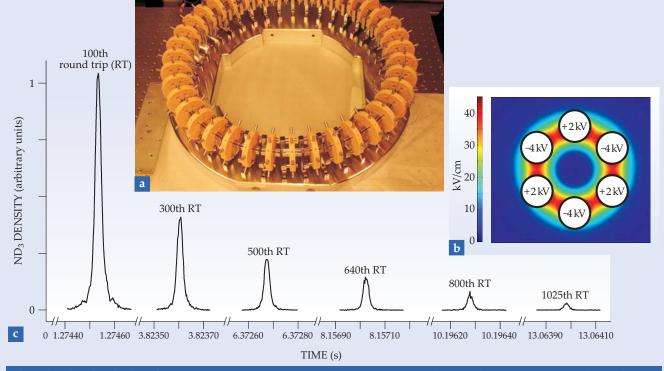
ticles are. Such is not the case when the particles reach relativistic velocities. In that regime the increase in velocity imparted by the electric field is not precisely compensated by the increase in radius; particles could thus arrive at the electrodes when the electric field there would decelerate them. In a synchrotron, controlled application of the oscillating electric field guarantees that particles always encounter an accelerating field at the electrodes. As a result, a particle's energy is limited only by the synchrotron radiation it necessarily leaks. (To learn about accelerators in industry, see page 46.)

Like the cyclotron, the synchrotron accelerates a bunch of particles with a range of velocities and positions, not just a single particle. That crucial feature ensues from the synchrotron's so-called phase stability, a concept developed independently by Vladimir Veksler and Edwin McMillan in 1944–45. McMillan explained the idea in a *Physical Review* paper. Suppose, he argued, that the particle crosses the electrodes just as the electric field passes through zero. Suppose further that the phase of the field is such that an earlier arrival of the particle would result in its acceleration. The orbit is obviously stationary in the sense that the particle neither accelerates nor decelerates. Moreover, the orbit for a bunch of particles is stable and the particles remain longitudinally confined. To see that, suppose a particle arrives at the electrode too early, before the field passes through zero. It is then accelerated. The increase in energy causes an increase in the orbital period, which makes the time of arrival later. A similar argument shows that a decrease of energy from the equilibrium value also tends to correct itself. The orbits that don't quite pass through zero field oscillate, with both position and energy varying about the equilibrium values.

Keeping neutral molecules in place

Unlike protons or electrons, polar molecules such as ammonia or the heteronuclear diatomics OH and NO are electrically neutral. But those molecules carry an electric dipole moment that makes them amenable to efficient manipulation by an inhomogeneous electric field that can accelerate, decelerate, or deflect a beam of molecules—or confine it.

Indeed, with a molecular synchrotron, the issue is not acceleration; rather, it's the confinement of packets of neutral molecules. The device must both keep the molecules in their orbit—that is, prevent them from crashing into the electrodes—and keep the particles bunched together. To achieve those goals, the molecular synchrotron includes an array of electrostatic focusing elements separated by small gaps. In analogy with the synchrotron's handling of charged particles, a synchronous change of the electric fields with the molecules' passage through the electrodes keeps the neutral molecular-beam packets bunched together in stationary orbits.



A long-lived molecular bunch. (a) The 0.5-meter-diameter molecular synchrotron at the Max Planck Society, Berlin. The small separation of the device's 40 focusing elements enables a large number of particles to be transported in the ring. (b) Electric field gradients are responsible for the forces acting on the polar molecules circling in the synchrotron. Shown here is the transverse electric field in the plane of the molecular synchrotron's hexapole focusers. Note that no force acts parallel to the hexapole axis at the locations of the hexapoles; the longitudinal force responsible for focusing a bunch of molecules arises only in the gaps between them. (c) As this plot of the density of deuterated ammonia (ND₃) molecules versus time shows, the ND₃ survives for more than 13 s, time enough to cross 41 000 hexapole elements in the course of a journey of more than a mile. (Adapted from P. C. Zieger et al.)

The circling packets of molecules can then repeatedly interact at well-defined times and positions with electromagnetic fields or with other atoms or molecules.

When particles encounter particles in the synchrotron, the device serves as a low-energy collider with which one can measure collision cross sections. In that case, the number of interactions per unit time scales with the square of the number of packets in the ring. More segments means more packets; in addition, a greater number of segments enables the stored packets to be of higher density. Therefore, it's advantageous to cut the ring into as many segments as practically possible. Panel a of the figure is a photograph of the Berlin molecular synchrotron. The machine includes 40 hexapole focusers (see panel b), each 37 mm long. Adjacent hexapoles, whose axes make an angle of nine degrees with respect to one another, are separated by gaps 2 mm wide.

The diameter of a synchrotron scales with the square of the velocity of the molecules it confines. So as a practical matter, the molecules in the synchrotron can't go too fast and need to be slowed prior to being loaded. The Berlin synchrotron has a diameter of 0.5 m, and the deuterated ammonia (ND₃) molecules that circle within are decelerated to a speed of about 125 m/s before being injected tangentially into the machine. Each ammonia packet is several millimeters long when it enters the synchrotron, and each consists of about a million molecules. Once the first molecular packet is inside the synchrotron, the hexapole fields are switched on. In the run corresponding to panel c of the figure, Meijer and colleagues injected a total of 13 packets at a rate of 10 packets per second. The first and last packets are four hexapoles apart; all other neighboring pairs are separated by three hexapoles. The density of the ND₃ molecules in the synchrotron decreases exponentially with time; that decrease is caused about equally by collisions with background gas and by excitation to untrappable states through the blackbody radiation present in the room-temperature chamber.

Pushing the limits

The Meijer group's experiments epitomize the level of control over molecular beams that can currently be achieved and set the stage for new ventures that will explore fundamental, poorly understood physics. Prominent among them will be studies that use co- or counterpropagating packets to explore cold molecular collisions that are more aptly described in terms of breaking waves than crashing billiard balls. The long-lived spatial confinement obtained in molecular synchrotrons should help experimenters push energy resolution to new highs, limited only by the energy-time uncertainty relation. It also enables experimenters to observe long enough to accurately measure the radiative lifetimes of metastable molecular states or the effects of the blackbody radiation on molecules; such experiments may in time see applications to spectroscopy and metrology. Quite a day's work for wisps in a chambered void.

Additional resources

- ▶ E. O. Lawrence, "The Evolution of the Cyclotron," Nobel Prize lecture, available online at http://nobelprize.org/nobel_prizes/physics/laureates/1939/lawrence-lecture.pdf.
- ▶ M. S. Livingston, ed., *The Development of High-Energy Accelerators*, Dover, New York (1967).
- ▶ R. V. Krems, W. C. Stwalley, B. Friedrich, eds., *Cold Molecules: Theory, Experiment, Applications*, CRC Press, Boca Raton, FL (2009).
- ▶ P. C. Zieger et al., "Multiple Packets of Neutral Molecules Revolving for over a Mile," *Phys. Rev. Lett.* **105**, 173001 (2010).