search & discovery

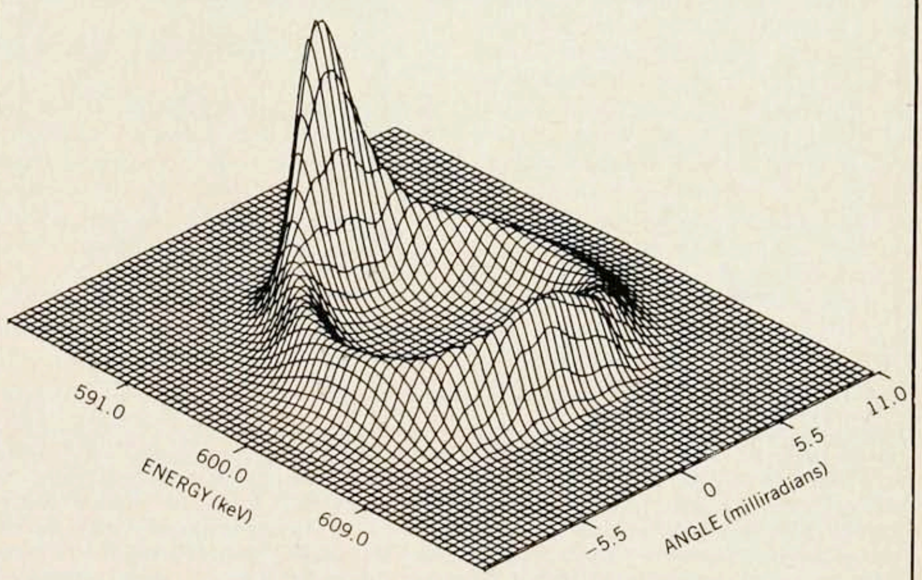
Fast ion beams reveal structures of molecular ions

Of the many ionic species that normally emerge from the ion source of an electrostatic accelerator, it has traditionally been only the monatomic ions that are considered to provide "useful" beams. The remaining beams of molecular ions (H_2^+, H_3^+, HeH^+, OH^+, etc.), which frequently constitute as much as 90% of the total ion-beam current, are seldom used in a nuclear-physics experiment.

But a group at Argonne National Laboratory, led by Donald S. Gemmell, has recently taken such molecular-ion beams, used them to bombard foil and gas targets and has looked at the dissociation fragments that emerge. With high resolution in energy (a few hundred eV) and angle (a few thousandths of a degree), Gemmell and his collaborators have observed a variety of new phenomena in the interactions of fast ions with matter. In addition the group has been able to measure the structures of some of the molecular-ion projectiles, determining bond lengths with accuracies of about 0.01 Å and bond angles to about 1°.

A crucial feature of these fast molecular-ion beam studies is that each projectile incident upon a solid target dissociates, forming a tight cluster of atomic ions that remain correlated in space and time as they progress through the target. Previous studies of the interactions of ions with matter have almost always employed monatomic ion beams that are usually treated as single-point projectiles.

Gemmell told us that "the cluster behavior exhibited by molecular-ion projectiles gives rise to new perspectives on ion–solid and ion–gas interactions and also poses a more stringent test for theories describing these interactions." As examples, he cites the use of molecular-ion beams in studying the interactions of ions with plasma oscillations in solids (the so-called "wake effect"), in stopping-power studies (distinguishing the contributions of close and distant electronic collisions), and in studying the charge-state distributions of fast heavy ions.



Joint distribution in energy and angle for protons from the dissociation of 3-MeV HeH$^+$ ions incident on a 195-Å thick carbon foil. The diameter of this ring pattern is mainly determined by the bond length in HeH$^+$. The width of the "rim" reflects variations in the internuclear distance of the incident molecular ions due to their vibrational motion. The nonuniform distribution of intensity around the ring can be accounted for quantitatively in terms of forces due to the polarization "wakes" generated by the ions as they traverse the foil. (From ref. 1)

The Argonne experiments are at present being done by Gemmell, Patrick J. Cooney and Elliot P. Kanter using a 4-MV Dynamitron accelerator. The experimenters start with an ion source containing He and H_2 and select, say, a beam of 3-MeV HeH^+ by magnetic analysis. After tight collimation these ions enter a target foil (typically 100 Å of carbon), where each molecular projectile has its two binding electrons stripped off within a few angstroms after penetrating the front surface. That leaves an alpha particle and a proton about 0.8 Å apart. They repel each other apart in a "Coulomb explosion," dumping their initial potential energy (~36 eV) into center-of-mass kinetic energy. So, emerging from the foil, one has fragments with both the original beam velocity and a component from the Coulomb explosion. This component causes a shift in the laboratory energy and angle of each fragment; the shift depends on the orientation of the incoming molecule.

If the axis of the molecule is parallel to the beam, with proton leading and alpha trailing, the proton is kicked forward and the alpha backwards in the center-of-mass frame. Each has its laboratory energy shifted by about 8 keV. If the molecular axis is perpendicular to the beam, the laboratory energy of the fragments is unshifted, but they receive a maximal angular deflection (a few tenths of a degree).

Several meters downstream from the foil is a tightly collimated entrance aperture to an electrostatic analyzer. Here the fragments are energy analyzed with a $\Delta E/E$ of 6×10^{-4} (FWHM). Angular measurements are done with the aid of electrostatic deflector plates just downstream from the target. The experimenters measure joint distributions in energy and angle for the emerging fragments. These distributions are ring-like; the diameter of the ring is determined primarily by the bond length in the incoming projectiles, and the "thickness" of the ring reflects the distribution in internuclear separations due to vibrational motion of the projectiles.

The ring patterns are slightly distorted from the shape expected on the basis of a simple "Coulomb explosion." This distortion together with the non-uniform distribution of particle intensity around the ring can be quantitatively accounted for, Gemmell told us, in terms of interactions with plasma oscillations ("wakes")

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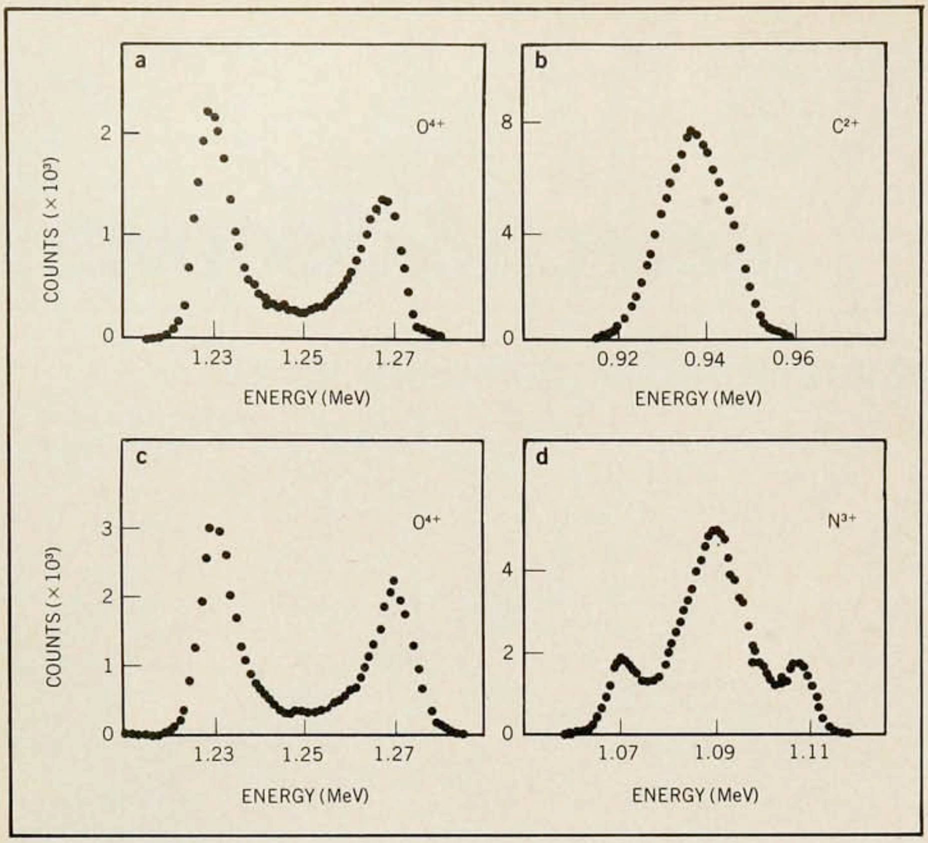

Energy spectra for (top left) O^{4+} and (top right) C^{2+} arising from 3.5-MeV CO_2^+ incident on a 133-Å carbon foil and for (bottom left) O^{4+} and (bottom right) N^{3+} arising from 3.5-MeV N_2O^+ incident on a 160-Å carbon foil. Particles detected are those emerging parallel to incident beam. The left-hand spectra indicate all the O atoms in both CO_2^+ and N_2O^+ are on the "outside" and that the two O atoms in CO_2^+ occupy equivalent positions. Right-bottom spectrum show two non-equivalent positions for the N atoms in N_2O^+, one on the "outside" and one roughly in the center. Right-top spectrum indicates C atom is roughly in the center of CO_2^+. (From reference 5)

generated in the target by the passage of the projectiles.[1] The wake potential behind a fast ion can be viewed as the sloshing to and fro of the electron sea through which the ion travels. In this respect it is analogous to the wake behind a boat. Imagine a second boat trailing in the wake of the first—it feels the wake. In an experiment with HeH^+ ions, the two boats might be the alpha as the leading boat and the proton as the trailing boat. The wake force tends to make the proton, in this case, fall in line behind the alpha.

The idea of a polarization wake induced by a fast charged particle moving through a solid was first considered by Niels Bohr in 1948. Over the past few years, the topic has been discussed by several groups. For example, Rufus Ritchie (Oak Ridge) and Werner Brandt (New York University) have proposed that the potential minima in the wake behind a fast ion may be capable of trapping and binding other charged particles. They suggest that the capture of target electrons in this way may account for the pronounced group of electrons observed to emerge with the beam velocity from foils bombarded by fast ions. Last year Roman Laubert (NYU), Ivan Sellin (Oak Ridge) and their collaborators studied the properties of a large number of "convoy electrons" ejected in the forward direction by heavy ions passing through thin solid targets. Their observed[2] yields and energy distributions disagreed with the properties predicted by the wake-riding theory.

At the University of Lyon, Bic Remillieux and Jean-Claude Poizat reported[3] that there is a small but definite probability (10^{-4} to 10^{-8}, depending on target thickness, beam velocity, and so on) that fast diatomic molecular ions are transmitted through thin foils. Gemmell and his group have also observed this phenomenon and have measured the energy losses and multiple scattering distributions for the transmitted molecules. Ritchie and Brandt have suggested that molecular transmission occurs when one ion in the diatomic projectile becomes trapped in the wake of its partner.

Structure measurements. In early Argonne experiments the group was able to measure the ring diameter to 1% accuracy and obtain bond lengths for many simple diatomic molecular-ion projectiles. The result agreed with theory to 0.01 Å. Following this, a collaborative effort between Argonne, the University of Lyon in France, and the Weizmann Institute in Israel focused on the structure of H_3^+. Using variations on these "Coulomb explosion" techniques, each lab confirmed that H_3^+ is an equilateral triangle in shape and measured the bond length to be close to 1 Å.[4]

The standard way to obtain molecular structure with great precision is to shine visible, infrared or microwave radiation on a gas and observe rotationally resolved fluorescence or absorption spectra. For neutral molecules, thousands of structures have been determined. For molecular ions, on the other hand, it is difficult to obtain sufficient spatial density for rotationally resolved spectra. However, there has been a considerable amount of vibrational structure measurements from which the qualitative geometrical structure can be deduced.

With traditional techniques, the structures of only eight polyatomic ions have been determined, according to Gemmell, and all eight are triatomic systems. Nevertheless, knowledge of molecular-ion structures is important to many theoretical chemists and astrophysicists. This knowledge gap has inspired the Argonne group to develop their techniques for measuring the structure for more complex molecular-ion projectiles.

To begin with, Gemmell, Kanter and W. J. Pietsch (Cologne) examined fragments from 3.5-MeV beams of CO_2^+ and N_2O^+. These ions are two of the eight whose structures are already known and have the linear structures O—C—O and N—N—O, respectively. The group was able to show[5] that by simply counting the number of peaks in the energy spectra, the structures could be inferred.

This ability to distinguish immediately the variations in structure of a given molecule is a very promising feature of the new technique. Gemmell believes it will be competitive with existing techniques for studying known molecular ions. For many ions, even the qualitative structure is not known. Gemmell's group has started work on the molecular ions CH_n^+, $C_2H_n^+$ and OH_n^+, where $n = 0$ to 4. The group expects to be able to distinguish between the various possible structural forms of each of these ions.

To improve the precision of their structure determinations on more complicated molecular ions, Gemmell and his collaborators have recently made extensive modifications to their apparatus to permit measurements on coincidences between fragments from a given projectile and tested the apparatus on the known structure, OH_2^+.

A leader in the structural measurements of molecular ions is Claude Woods at the University of Wisconsin, who has a large discharge tube—6 inches in diameter and 12 feet long. By looking for microwave absorption, he has determined over the last several years the structures of the molecular ions HCO^+, N_2H^+ and CO^+. Woods and his collaborators can obtain far greater accuracy in their determinations because of the inherent high resolution of microwave spectroscopy. The groups at Argonne and Wisconsin are consulting with each other to determine which molecular ions are particularly worthy of attention. Once the Argonne group determines the structure to 0.01 Å in bond length, that narrows the frequency range in which Woods's group would need to scan. The Wisconsin equipment is capable of measuring frequency to eight significant figures, yielding bond lengths to better than 0.001 Å. The precision of the bond-length determination is at present limited by the theoretical treatment of molecular vibrations. —GBL

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Gravity waves slow binary pulsar

Although most physicists have long been convinced that gravity waves would ultimately be detected, there has yet been no unambiguous experimental confirmation of their existence. Recent work by Joseph Taylor and Lee Fowler of the University of Massachusetts at Amherst and Peter McCulloch of the University of Tasmania, however, provides further evidence for the reality of these elusive waves. In the 8 February issue of *Nature* (277, page 437), Taylor, Fowler and McCulloch report that the orbital period of the only known binary pulsar is diminishing at a rate of about 3 parts in 10^{12}, which is consistent with quadrupole radiation of gravity waves by the system.

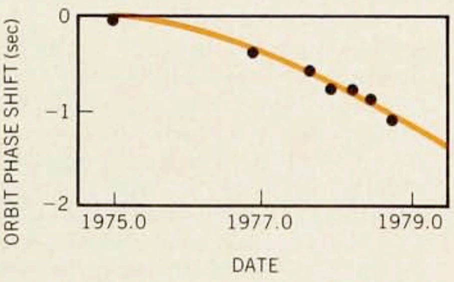
The data that have led to this conclusion come from four years of observation of the radio pulsar PSR 1913 + 16 with the 305-m radio telescope at Arecibo. The period of the pulsar is about a twentieth of a second and is known to a few parts in 10^{10}. To obtain the precise arrival times of the pulses, Taylor's group fits the long-term average pulse-profile to the sums of about 5000 pulses whose period has already been corrected for the Earth's motion. The position of some standard feature of the fitted profile then serves to define the arrival times of the pulses during a roughly ten-minute interval. Over the past several years the group has improved their equipment and analysis so that the uncertainty of their measured arrival times is now only about 50 microseconds.

As is the case for other pulsars, the arrival times are known precisely enough to be sensitive to even subtle effects from the motion of the Earth, including the annual change in the gravitational redshift as the Earth moves closer to and farther from the Sun. By obtaining data at two radio frequencies, the group can also correct for the dispersive delay due to the interstellar medium. The result of these corrections gives, in effect, the pulse period that would be seen by a stationary observer near the two orbiting stars.

The pulsar is in orbit around another star, which has not yet been directly observed. This orbital motion, of course, affects the measured period of the pulsar, and the data are now numerous and precise enough to permit determination of the important orbit parameters. The orbit is a highly eccentric one, with a period of almost eight hours; its plane appears to be inclined at about 30° to the line of sight. The various parameters of the binary system can become disentangled because they affect the observed pulsar period with different functional forms and to different orders of v/c.

Two parameters are of particular interest for tests of general relativity: the rate of periastron advance, that is, the rate with which the orbit precesses, and the rate with which the orbital period decreases. The former is analogous to—but much larger than—the well-known perihelion shift of Mercury, which has provided one of the classic tests of general relativistic dynamics. The latter is the term providing evidence for gravitational radiation. The magnitude of the effect found by Taylor and his collaborators is consistent with the conventional general-relativity calculation for quadrupole radiation from two orbiting point masses, Taylor told us.

The only parameters of the orbiting stars that are not determined with great precision by the data are the masses of the stars. However, the formulas for several of the parameters used to describe the binary system do include the masses, and the measured values of these give masses near 1.4 $M_\odot$ for both objects.



Residual orbital period of PSR 1913 + 16 with respect to the initial observation. The uniform decrease in phase is due to an initial overestimate of the period. The second derivative of the theoretical curve represents the predicted effect of gravitational radiation from the system.

The interpretation of the data depends somewhat on the model one assumes for the two stars involved. The most probable situation, Taylor told us, is that these are two compact objects in orbit around each other, behaving almost like ideal point masses. The mass of each object is in the right range for neutron stars, although other possibilities, such as white dwarfs are not ruled out. In either case, as Malvin Ruderman (Columbia) told us, it seems unlikely that tidal or similar effects are important: Even if the companion is a white dwarf, it must be so compact that it would behave as a rigid body. Mechanisms other than gravitational radiation, such as tidal or magnetic effects, would presumably also affect the pulsar period itself; unlike other fast pulsars, however, PSR 1913 + 16 is slowing down only very slowly (less than one part in 10^{17}). A number of noted astrophysicists have said that the observed effect could, in principle, also be due to a Doppler shift if the binary star were in a slow orbit around a distant third companion. However, Taylor remarked that Nature would have to be perverse to have given the third object just the right mass and distance to produce the observed decrease in orbital period. He added that data gathered over the next few years should enable his group to distinguish between such orbital motion and gravity-wave damping.

The figure shows the phase of the orbital motion with respect to the initial (1975) observation. Each point represents an average of several weeks' or months' data. The theoretical curve is the prediction from general relativity assuming both stars have masses of 1.41 $M_\odot$. The important aspect is the apparent curvature; the mean slope of the points and of the theoretical curve indicate only the error in the value of the orbital period that was deduced from the initial observations. To confirm the fit, Taylor's group also computed the range of possible masses of the stars by fitting all orbit parameters except the periastron advance, but including the orbital-period decrease. This fit gave masses for the pulsar and its companion around 2 $M_\odot$ and 1.5 $M_\odot$, respectively, but Taylor does not believe that the difference is significant.

Irwin Shapiro of MIT, who has worked on many tests of general relativity, said that while he was impressed with the results he would be somewhat concerned about the statistical significance of the curvature, particularly in light of the gap in the data after 1975. Taylor agreed that the 1975 point appeared isolated, but he said that the remaining data show the same curvature, although with less statistical significance. He also felt confident about the reliability of the 1975 data, although they depend on less precise observations, because they do represent