Quantum Electrodynamics: Excited State of Positronium

Positronium consists of an electron and its antiparticle, the positron, which are bound together and orbit one another, thus resembling the orbiting by the proton and electron of a hydrogen atom. Because of this resemblance, scientists have long thought that positronium should also exhibit a spectrum of quantum states similar to that of hydrogen. Although the ground state of positronium was identified nearly 25 years ago, it was only within the last few months that investigators have experimentally verified the existence of its first excited state by observing the emission of ultraviolet light when the excited state decayed to the ground state.

More recently, the same group of researchers, Karl F. Canter, Allen P. Mills, Jr., and Stephan Berko of Brandeis University, Waltham, Massachusetts, devised a way to probe the details of the excited state. Further improvements in their technique may one day enable scientists to make tests of quantum electrodynamics (the basic theory that describes electromagnetic interactions between charged particles or between charged particles and photons) that are as precise as those that can be made with the use of conventional atomic systems, such as hydrogen, but without the complicating effects of the atomic nucleus.

Martin Deutsch of the Massachusetts Institute of Technology, Cambridge, discovered positronium in 1951, and completed a number of investigations of the properties of its ground state within a short time. Since then the ground state has been extensively characterized, but studies of excited states comparable to those possible in hydrogen have not been possible because experimenters have been unsuccessful in attempts to observe excited states.

The keys to the success of the Brandeis researchers in identifying the excited state were threefold: (i) they used positrons with low kinetic energies that had previously been shown at Brandeis to efficiently form positronium with electrons from solid surfaces; (ii) they carried out the experiment in a vacuum; and (iii) the positronium formation occurred well away (about 1.5 meters) from the positron source.

Traditionally, positronium experiments have been carried out in a gas. A radioactive isotope, such as sodium-22, acts as a source of energetic positrons with kinetic energies of hundreds of thousands of electron volts. These energetic positrons collide with gas molecules which serve the dual purpose of slowing down the positrons and of acting as a source of electrons with which the slow positrons can combine to form positronium.

Since the electron and the positron are antiparticles, they rapidly annihilate each other when they come sufficiently close together. In the annihilation event, gamma rays are given off. Although annihilation generally occurs directly, the negatively charged electron and positively charged positron are occasionally first bound together by their electrostatic attraction. In the ground state, the electron and positron are separated by about 1 angstrom, and the positronium "atom" exists for a characteristic lifetime that depends on whether the electron and positron spins are parallel $(1.4 \times 10^{-7} \text{ second})$ or antiparallel (1.25 \times 10⁻¹⁰ second). The latter lifetime corresponds to about 400,000 orbital revolutions, so that positronium is in a sense a stable system.

In accordance with quantum mechanical selection rules, two oppositely directed gamma rays are created in the case of annihilation when the spins are antiparallel, and three coplanar gamma rays when the spins are parallel. The properties of positronium, then, are obtained from studies of these gamma rays under various conditions.

Excited states might be detected by changes in the intensity of gamma rays when positronium is optically excited by a strong ultraviolet light source. Alternatively, observation of ultraviolet photons emitted when positronium decays from an excited state to the ground state would also signal the presence of excited states. However, the gas necessary for positronium formation by the traditional methods prevents these measurements. Collisions between gas molecules and the presumed excited positronium can cause either rapid decay to the ground state without emission of ultraviolet light or ionization of the electron-positron pair. In addition, the close proximity of the positron source to the gas means that there is a high level of background gamma radiation against which the desired effects must be seen.

For several years, researchers have been using positrons as a probe to study imperfections and, more recently, surfaces of both metallic and nonmetallic solids. In such an experiment designed to study the interaction of slow positrons with solid targets, the Brandeis scientists serendipitously discovered last year that they could make positronium without using a gas.

In their experiment, fast positrons were

slowed down by an assembly of magnesium oxide coated gold foils placed near the cobalt-58 source. About one in 30,000 fast positrons entering the foil assembly was reemitted as a slow positron. The slow positrons were guided along an evacuated $(2 \times 10^{-8} \text{ torr})$ 1.5-meter path with a bend in the center by a magnetic field. The velocity of the slow positrons reaching the target was controlled by a voltage placed on the target. Positronium production with efficiencies of up to 85 percent could be achieved, depending on the temperature of the target and the energy of the slow positrons.

With a germanium target held at room temperature, about 4000 positronium "atoms" were produced per second. With this much positronium formed in the absence of a gas, the investigators thought it might be possible to detect ultraviolet radiation of the wavelength (2430 angstroms) expected when positronium in the first excited state decays to the ground state. With the use of a photomultiplier tube to detect the ultraviolet light and interference filters to select the wavelength, a significant signal (50 percent above background) was obtained at the expected wavelength.

The investigators were careful to exclude the possibility that the observed signal was due to spurious sources, such as ultraviolet radiation (scintillations) caused by gamma rays or fast positrons in the target chamber. For example, they counted an ultraviolet signal only when it preceded the detection of a gamma ray by a time about equal to the calculated lifetime of positronium in the ground state, an event corresponding to the decay from an excited state followed by the annihilation of positronium in the ground state. About one of these coincidences was observed every 1000 seconds.

To a first approximation, the energy levels of positronium are obtainable from the Bohr model of the atom or from nonrelativistic quantum mechanics as $E_n =$ $-E_{\infty}/n^2$, where E_n is the energy of a quantum state with principal quantum number *n* (an integer), and E_{∞} is the ionization energy (6.8 electron volts). Increasing values of n corresponds to larger orbits or larger distances between the two particles until eventually the atom is ionized. Several quantum states may have the same value of n, provided that other quantities, such as spin or orbital angular momentum, are different, and are said to be degenerate (have the same energy). In actuality, however, there are small corrections to the energies of the nominally degenerate states that cause their energies to differ slightly. These corrections are calculable in quantum electrodynamics as coefficients in a series expansion in powers of a small quantity called the fine structure constant, although several effects may contribute to a particular order in the expansion.

The ultraviolet emission recorded by the Brandeis scientists corresponded to the decay from an excited state at n = 2 to the ground state at n = 1. To further characterize the n = 2 excited state, the investigators attempted to examine its fine structure, that is, the energy difference between two different quantum states with n = 2. To make this fine structure measurement, a radio frequency resonant cavity was constructed at the target chamber. Then, when microwave radiation of the appropriate frequency to cause a transition from an n = 2 state with zero orbital angular momentum (called the 2S state) to another with a nonzero momentum (called the 2Pstate) was introduced into the cavity, they found about a 10 percent increase in the ultraviolet emission that satisfied the criterion for coinciding with gamma rays. The increased ultraviolet emission is in accordance with quantum mechanical selection rules that prohibit direct transitions between two states both with zero orbital angular momentum, such as the ground state (1S) and some excited states (2S). The microwave frequency needed to cause the increased coincidence rate was 8628.4 ± 2.8 megahertz which is within 10 megahertz of the theoretical value calculated from quantum electrodynamics by Thomas Fulton (now at Johns Hopkins University) and Paul C. Martin of Harvard University in 1954.

Predictions stemming from quantum electrodynamics have been extensively and accurately tested in a wide variety of physical phenomena at the atomic level, with remarkable agreement between theory and experiment in every case. In the face of such spectacular success, there has arisen a widespread feeling that, at least at low energies, quantum electrodynamics is for all intents and purposes a satisfactory theory. Nonetheless, scientists say there are important reasons for looking to experiments on positronium to provide further confirmation of quantum electrodynamics and that make it likely that the Brandeis experiments will stimulate further attempts at precision measurements of the excited states of positronium.

For example, some of the tests of quantum electrodynamics involve precision measurements of small corrections to the energy levels of hydrogen and other simple atoms, such as the hyperfine splitting of the 1S ground state (due to the interaction of the magnetic moments of the nucleus and the electron in the case of hydrogen) and the Lamb shift (the difference in energy between the nominally degenerate 2S and 2P excited states). Agreement between theoretical and experimental values of these minute energy differences exists to several significant figures.

Complicating such tests, however, is the presence of the atomic nucleus. The nucleus itself has a structure, and effects arising from the nuclear structure can make small contributions to these energy differences. In positronium, moreover, there are additional effects which do not occur in atomic systems with nuclei. Thus, the study of positronium affords an opportunity to fully test quantum electrodynamics without the presence of complicating nuclear effects.

The fine structure measured at Brandeis agrees with that calculated through the third significant figure, but contributions from as yet uncalculated terms in the theory, as estimated by Fulton and Martin, are expected to be comparable to the present experimental uncertainties. According to Berko, a number of improvements and additional measurements would need to be carried out in order to increase the precision of the fine structure determination. Such improvements are likely to be made and precision measurements carried out in the future. As it stands, the Brandeis result is as precise as the theory with which it can be compared.—ARTHUR L. ROBINSON

Air Pollution: Where Do Hydrocarbons Come From?

Man's knowledge about air pollution and its causes is limited. Despite the laws that have been enacted to curtail emissions of potential pollutants from automobiles, for example, the role of automobile emissions in the genesis of the most severe forms of pollution, including photochemical smog, is incompletely understood. The lack of knowledge about this subject was recently illustrated by the controversy surrounding a report which concluded that, in certain areas and under certain conditions. hydrocarbons released from trees and other vegetation may be more important than those released from automobiles in the initiation of smog.

This possibility, which is based on very preliminary results obtained from limited data, has been sharply criticized by many investigators who have argued—largely on the basis of intuition—that the hypothesis cannot be correct. But the sad fact of the controversy is that there is not enough background information either to support or to refute the hypothesis.

The contested study was performed by

Raymond A. Saunders, Fred E. Saalfeld, and J. R. Griffith of the Naval Research Laboratory in Washington, D.C. The three investigators do not normally work in the field of air pollution research, a fact which has further fueled the controversy surrounding their conclusions. Their findings were a serendipitous result of their attempts to measure the concentrations of dissolved gases and hydrocarbons in ocean water. They developed a sophisticated gas chromatograph-mass spectrometer system for detecting and identifying the compounds and were refining the technique using water samples from local sources. On the spur of the moment, they decided to collect water samples from a rainstorm on 13 August 1973 that ended a period of intense smog in Washington.

They found that the rainwater contained unusually high concentrations of the cyclic ether 3-methylfuran; its concentration was, in fact, more than 100 times as great as that of any other hydrocarbon in the water. 3-Methylfuran is a relatively reactive substance that accounts for no more than about 0.3 percent of the hydrocarbons in automobile emissions. Saunders thus suggests that the most likely source of the 3methylfuran he observed is the atmospheric oxidation of terpenes that are released by trees and other vegetation, particularly during late summer. This oxidation, in turn, could contribute to the formation of smog-producing ozone by interfering with the natural cycle of oxygen and oxides of nitrogen.

In the absence of hydrocarbons, both oxygen and nitrogen dioxide in the atmosphere are in a rough equilibrium with ozone and nitric oxide. Nitrogen dioxide (NO_2) in the presence of sunlight is dissociated into nitric oxide (NO) and an atom of oxygen; the atom of oxygen then combines with a molecule of oxygen to form ozone. The ozone can then react with nitric oxide to re-form nitrogen dioxide, even in the dark. When several different hydrocarbons are present, however, they are converted by an incompletely understood mechanism to highly reactive peroxy compounds. These peroxy compounds in-