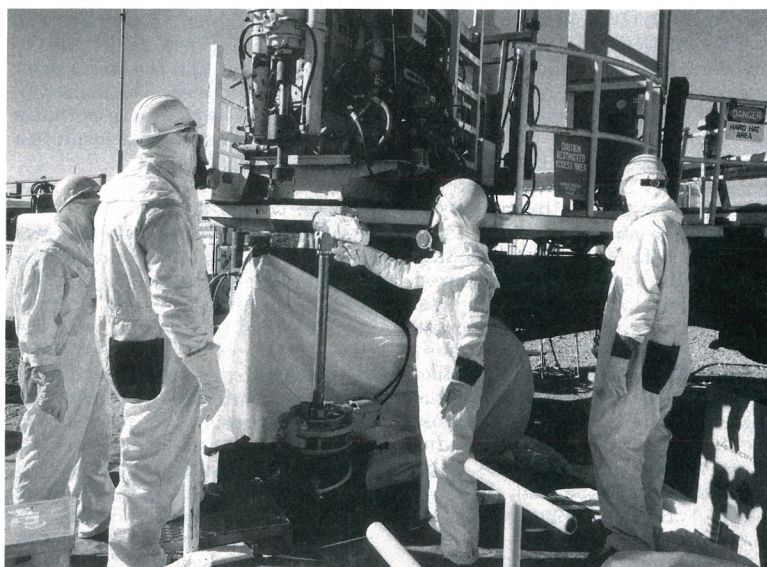


HANFORD SEEKS SHORT- AND LONG-TERM SOLUTIONS TO ITS LEGACY OF WASTE

The 560 square miles of desert occupied by the Hanford Site in southeastern Washington are strewn with the silent hulks of reactors and processing plants that once produced plutonium for nuclear weapons. Lurking beneath the surface is the legacy of those plants—nearly 50 years' accumulation of chemical and nuclear waste, including over 60% by volume of the nation's high-level radioactive waste from weapons production. Some of the wastes will remain sequestered from the general public long enough to allow research on the best long-range solution to the problem of their disposal. But others require immediate action. Among the most urgent are the 177 million-gallon storage tanks for high-level wastes, some of which contain potentially explosive mixtures of chemicals.

Managing these wastes has become the chief task of the Hanford facility, where \$783 million was budgeted in fiscal year 1991 for waste management and environmental restoration, compared with Hanford's total budget of \$1.4 billion. It may take more than \$50 billion over the next 30 years to clean up the radioactive and chemical wastes that have been stored in tanks, buried in trenches or dumped directly into the earth at Hanford. Similar expenses confront other facilities in the defense complex.

The magnitude of the problem is so enormous, with contaminated soil being measured in billions of cubic meters and high-level wastes in tens of millions of gallons, that new ideas will clearly be needed to find the most effective, safest and most efficient waste disposal methods. ("High-level waste" is a term that generally refers to the intensely radioactive material in spent fuel from nuclear reactors or in the waste from reprocessing this fuel.) Researchers must start by characterizing the waste, much of which was disposed of with little or no documentation, and then they must understand the behavior of the partic-



BOEING COMPUTER SERVICES

Extracting core samples in 1990 from underground tanks that store high-level radioactive wastes at Hanford. Evidence that some of the tanks produce potentially explosive mixtures has lent urgency to the task of understanding what is in the tanks and how to handle it.

ular substances in their particular environments. In some cases, the mixtures exist under conditions that have never been studied. In the tanks, for example, are highly alkaline and highly concentrated solutions at elevated temperatures. Some of these problems are now being addressed by the Pacific Northwest Laboratory, a multiprogram laboratory located at Hanford and operated by the Battelle Memorial Institute for the Department of Energy. (The Hanford Site is currently managed for DOE by Westinghouse.) DOE is funding a \$217-million Environmental and Molecular Sciences Laboratory at PNL to do long-range research on environmental remediation and waste management. We recently visited PNL to survey the specific problems faced at Hanford and to discuss some of the cleanup technologies be-

ing studied at PNL.

Making waste

Over the years, nine plutonium-production reactors were built at Hanford, the first in 1943 and the last (the N Reactor) in 1963. By 1971 all of these had been shut down except the N Reactor, which continued to operate until 1988. (See the map on page 19.) The plutonium fuel from these reactors was reprocessed on site in chemical processing plants, the most recent of which was the Plutonium and Uranium Extraction plant. PUREX is currently idle, but DOE has considered restarting it to reprocess some remaining N-Reactor fuel that now sits in 40-year old underwater storage basins near the Columbia River, awaiting permanent disposal. About 3-5% of the fuel elements are leaking and have contaminated the

pools. One option for disposal involves reprocessing at the PUREX plant, but the facility would have to undergo considerable renovation before it could operate within environmental regulations.

In the process of extracting uranium and plutonium from the fission products, the reprocessing plants convert the waste to a liquid form occupying a much larger volume than the original fuel elements. According to a 1991 report by the Office of Technology Assessment,¹ the extraction of one kilogram of plutonium at the PUREX plant produces over 340 gallons of liquid high-level radioactive wastes, more than 55 000 gallons of low-to-intermediate-level radioactive wastes and over 2.5 million gallons of cooling waters, and the US has produced about 10^5 kg of plutonium for military use. The operators of Hanford historically used methods of treatment, storage and disposal of these various waste streams that would not meet today's standards. The high-level radioactive wastes, containing most of the fission products mixed with organic and aqueous solvents, were poured into waste storage tanks—149 single-shell tanks built between 1944 and 1966, and 28 double-shell tanks constructed after the single-shell tanks were found to leak. The low-to-intermediate-level radioactive wastes were put into cribs, which are like septic-tank drainage fields. And the cooling water was pumped to surface ponds.

Hanford now has to deal with its legacy of some 1700 waste sites. The single-shell tanks have leaked at least 750 000 gallons (and possibly much more) of their waste into the ground, according to a 1991 report by the Government Accounting Office.² About 157 million curies of radioactive material remains in the single-shell tanks, and about 111 million curies in the double-shell tanks.¹ (For comparison, about 50 million curies are thought to have been released into the environment by the Chernobyl accident.) In addition to the leaks in these tanks there is the threat of detonations from explosive combinations of chemicals in some of the tanks. The waste dumped into cribs has reached the aquifer underlying the Hanford Site, and the contamination in this aquifer flows toward the Columbia River, the source of drinking water and irrigation for a wide area. Already tritium from this waste has reached the river, although so far it is diluted enough to remain far below acceptable drinking standards.

Problems with reprocessing waste are not unique to Hanford: Plutoni-

um for weapons has also been reprocessed at the Savannah River plant in South Carolina. (The Idaho National Engineering Laboratory has reprocessed highly enriched uranium from the naval nuclear reactor program, and a demonstration plant at West Valley, New York, reprocessed a small amount of commercial fuel from 1966 to 1972.) Together Hanford and Savannah River have the lion's share of the defense high-level waste. While Hanford accounts for the largest percentage of the volume of the high-level wastes, Savannah River has a slightly higher share of the total radioactivity. Although the waste at Savannah River is also stored in tanks, the tanks are of different designs and the waste itself is chemically simpler. The main concern about the tanks there is that a failure of ventilation could result in a buildup of hydrogen gas.

Danger of explosions

The Hanford tanks contain a potent brew of chemicals: At the reprocessing plant the spent reactor fuel was dissolved in nitric acid, and after the uranium and plutonium were removed, the plant operators added sodium hydroxide to the processing waste to neutralize the acid and prevent corrosion of the storage tanks. In some cases plant operators wanted to reduce the radioactivity of the processing waste, so they injected ferrocyanide and sodium titanate to precipitate cesium-137 and strontium-90, respectively. (The cesium and strontium, whose activity is about 180 million curies, are now stored in capsules on the Hanford Site.) As a result of such additions the tanks have become active chemical crucibles, in which constant transformations occur through both chemical and radiological means, stimulated by the abundance of chemical species, the presence of radioactive emissions and the elevated temperatures provided by the decay heat.

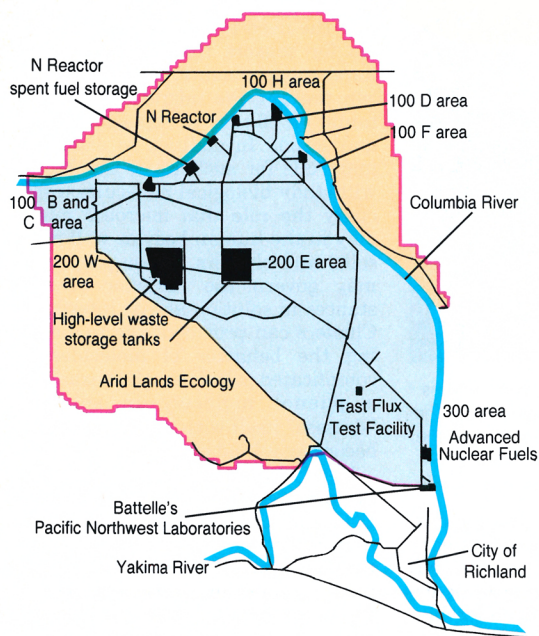
The high-level waste tanks have been troublesome all along, although DOE made the extent of the risk public only in the summer of 1990. The threat of explosions comes from two different sources. One is the hydrogen that is accumulating in some tanks. The hydrogen appears to be generated by the radiolytic decomposition of water and some organic compounds. In most tanks the hydrogen gas bubbles out of solution and is pumped out of the tank by the ventilation system. But hydrogen may still build up in some of these tanks.

In one of the double-shell tanks, labeled 101-SY, the constituents have

formed a very viscous slurry at the bottom of the tank, covered by a layer of liquid and topped with a 3-4-foot thick semisolid crust (This crust does not form a continuous cover). As the hydrogen forms in this tank it remains trapped in the slurry. Within the hydrogen bubbles there is also some nitrous oxide, which contributes to the explosive potential. These gas bubbles generate enough pressure within the slurry to raise its level, sometimes by as much as a foot. (The tank diameter is about 75 feet.) Then suddenly the hydrogen is released in one large "burp" as the gas-containing slurry and the liquid above it turn over. In a videotape taken by a camera inside tank 101-SY during a burping episode, the slurry looks like a thick brew of oatmeal at a slow, rolling simmer. The process repeats itself at intervals of a few months. In 101-SY, we were told, the hydrogen concentrations in bubbles under the tank crust are estimated to have reached of about 35% on occasion, compared with the flammability threshold of 5%. Nevertheless DOE maintains that the probability of an explosion is low, based primarily on the lack of an ignition source and the fact that the tanks have not exploded during the last 13 years.

If an explosion were to occur, how severe might it be? A study done by the Westinghouse Hanford Company³ found that an explosion might release as much as 2.2×10^6 kcal of energy, not enough to rupture the tank. However, the explosion might induce subsequent exothermic reactions among the other tank constituents, indirectly causing overpressurization and collapse of the tank and consequent release of some of its contents. In a report on DOE nuclear facilities issued last fall, the Advisory Committee on Nuclear Facility Safety, headed by John Ahearne (now executive director of Sigma Xi), expressed concern that a fire might begin in the gases at the top of the tank, with the potential to ignite the crust, which contains significant concentrations of the fission product cesium. Jess Cleveland, a member of the Ahearne committee who served on a subcommittee that studied the tanks, told us that such a fire might spawn a slow release of radiation lasting longer than that resulting from an explosion.

DOE is also concerned about 20 single-shell tanks to which ferrocyanide was added during one period to precipitate cesium-137. This compound, which is slightly reducing, is mixed in the waste with the nitrates and nitrites, which are oxidants. At sufficiently elevated temperatures,



The Hanford Site

borders the Columbia River in southeastern Washington. The nine plutonium production reactors were built in the so-called 100 area, along the river to the north. High-level waste tanks are buried in the 200 area, where the spent reactor fuel was processed. The 300 area was used mostly for fabrication of reactor fuel. DOE's Pacific Northwest Laboratory is located just south of the site. The section labeled "Arid Lands Ecology" is part of the Hanford site.

these ingredients might react exothermically. Based on experiments with much smaller and purer mixtures of these materials than exist in the tanks, DOE and Westinghouse believe that the temperatures in the tanks (about 57 °C) are well below those (about 285 °C) at which an explosion might occur. Arjun Makhijani of the Institute for Energy and Environmental Research in Takoma Park, Maryland, worries, among other things, about local hot spots where temperatures might be significantly elevated. (The temperatures are only measured at one position in each tank.) The Ahearne committee criticized DOE's heavy reliance on the temperature thresholds predicted in laboratory experiments and urged that the experiments be conducted at larger scales and on samples more representative of the real composition in the tanks. Large-scale tests with actual tank samples are formidable because of the high levels of radioactivity involved.

The tank problems at Hanford invite comparison with a catastrophic 1956 accident at a Soviet reprocessing plant in Kyshtym in the Urals: The explosion of an underground high-level waste storage tank there spread about 20 million curies of radiation over an area of about 15 000 square kilometers.

In October 1990, DOE convened a panel of in-house and outside experts, headed by Mujid Kazimi of MIT, to advise the agency on plans to charac-

terize and mitigate the threat posed by these giant vessels. Kazimi told us that Hanford has made some progress in the past year. Workers at Hanford have taken a full core sample from tank 101-SY to determine the chemical mix, and they are doing experiments with more realistic synthetic tank samples. (See the photo on page 17.) In the single shell tanks they are measuring the location of gamma emissions to find concentrations of fission products and thereby the ferrocyanide, which should be found in the same places. They are reassured so far to have found the gamma activity spread throughout the tanks and not preferentially at the bottom, as they had feared.

Even once the potential of explosion is in some way reduced, DOE must dispose of these wastes in a more satisfactory fashion. DOE has not yet decided what to do with the single-shell tanks: Over the years much of the liquid waste has been pumped out into double-shell tanks or reduced in volume by evaporators, so that the remnants in most single-shell tanks are in solid form. To remove the wastes for treatment and disposal one might have to add water to the tanks to dissolve the contents, but doing so would risk further leaks. Yet leaving the tanks where they are poses its own hazards.

As for the waste from double-shell tanks, DOE plans to pump it through a processing plant that would separate it into a large-volume stream of

low-level wastes and a much smaller stream of high-level waste. The former would be mixed with a cement-like material to form a type of "grout" that would be buried on site. The latter would be converted to a glass-like substance by the waste vitrification plant scheduled to be under construction at Hanford by April. But these plans for disposal can proceed only after the buildup of hydrogen gas is understood and remedied. In a July 1991 review of the planned Hanford Waste Vitrification Plant, DOE concluded that "the detailed design of the HWVP is considered premature."⁴

Cleaning up

There is a range of time scales for dealing with the waste. The most urgent items on the agenda are to deal with the threat of a violent reaction in the tanks and to stem the migration of contaminants into those areas where they pose the greatest risks to humans. But for waste that does not pose such imminent dangers it may be best to wait as long as possible for research to determine the optimal solution.

About half of the research at PNL is devoted to environmental restoration and waste management issues. PNL has several projects to address the short-term needs in these areas. We spoke with Nick Lombardo in the Applied Physics Center, whose group has developed a computer code for three-dimensional hydrothermal analysis. They used the code to simulate the behavior of tanks such as 101-SY, and they confirmed that the gases remain trapped in the sludge until they burp, rather than venting continually into the space above the crust. His group is also simulating the effectiveness of proposals to homogenize the tank contents in preparation for their removal from the tanks.

PNL has also developed a process for *in situ* vitrification of wastes below the soil. (See the photo on page 20.) The technique is to embed a series of electrodes in the ground to be treated and slowly heat it over a period of weeks. The soil and its associated contaminants are turned into a glassy rock that greatly retards the movement of wastes. The process must still be carefully studied to ensure that high temperatures destroy noxious gases such as carbon tetrachloride and do not just volatilize them and drive them out of the soil. (A hood is placed over the area to trap emerging gases.) So far the method has been tested on soil volumes as deep as 17 feet, but it needs to treat soils twice as deep. Clyde Frank,



Mobile unit for in situ vitrification of underground waste. PNL is developing this technology, in which electrodes are implanted in the ground, heating it to high enough temperatures that the soil, and the waste it contains, is fused into a glass-like form.

associate director of technology development in DOE's office of environmental restoration and waste management, told us that the big challenge is to develop a technology that can melt the soil from the bottom up rather than the top down.

One appeal of *in situ* vitrification is that facility operators would not have to dig the contaminants out of the ground for treatment and disposal, a procedure known in the business as "suck, muck and truck." Furthermore it provides an option for treating so-called "mixed waste," a combination of radioactive and chemical waste, for which no disposal standards have yet been formulated. No one knows what to do with such waste for now except to store it. Critics worry, however, that vitrification may lock the waste into the soil prematurely and prevent the application of a better remedy when it comes along.

PNL also has an active group studying bioremediation. The idea behind this technique is to enlist the aid of the army of microbes that naturally inhabits soils and sediments. When activated by some energy source, these microbes can gobble up and destroy organic contaminants, or they may change the valence states of some metals or radionuclides in a way that enhances or inhibits their migration through the soil and groundwater. One of the toughest problems at Hanford is carbon tetrachloride, a common contaminant at the site. Microbes cannot gain energy directly by breaking down CCl_4 , but they sometimes end up decomposing this compound as a step in another process in which there is an energy gain. Jim Frederickson told us that his group at PNL has found a microbe that will decompose CCl_4 in the process of converting nitrates (also common con-

taminants) to harmless nitrogen gas. But this microbe requires an energy source such as acetic acid. PNL researchers are now trying to determine whether the right microbial species survive in areas where the waste has been dumped, and they are exploring ways to activate these microbes *in situ*.

Unfortunately, the microbes "eat" the pollutants so slowly that cleansing the ground might take decades. Some of the research, including simulations of molecular dynamics, is therefore directed toward understanding the rate-limiting steps with a view toward speeding them up. But Frederickson feels that one way to improve the microbial rates by several orders of magnitude would be to genetically engineer "custom" microbes. Long-range research at PNL's Molecular Sciences Research Center is headed in this direction, although it may be ten years before this research can yield practical results. Clearly some cleanup will have to begin well before then.

Long-range research

PNL created MSRC two years ago to spearhead research on some of the longer-range issues in environmental restoration. It is headed by Michael Knotek, former director of the National Synchrotron Light Source at Brookhaven National Laboratory. Knotek conveyed to us a keen sense of the urgency and magnitude of the waste problem confronting Hanford. He recognizes the enormous sums of money necessary to deal with it. To save the nation from going broke on the cleanup, he asserts, research must not only help delineate cost-effective methods but also provide a basis for answering the question, "How clean is clean enough?" Coming from the

materials science community, Knotek sees the need to develop new materials for sensors that can withstand harsh environments, for barriers that can prevent the migration of pollutants, for filters to separate waste, and so on. In addition to exploring materials for such roles, the MSRC will study biological molecules (especially the role that microbes might play in the human body as well as in soils) and clusters, whose properties may govern the behavior of substances in solution and at interfaces. Clusters can provide a key to modeling the behavior of molecules in complicated solutions. (For example, a phenol molecule in solution "dresses" itself with water molecules, becoming a kind of cluster.)

The Environmental and Molecular Sciences Laboratory is being built at PNL under the auspices of the Molecular Sciences Research Center, with funding from DOE. It will be a user facility, and Frank told us that 80% of the experiments must be directly relevant to environmental waste problems. Topics to be studied include materials, structural biology, groundwater transport, chemistry in solution and computational modeling. The laboratory will feature a GHz-range nmr spectrometer, a mass spectrometer capable of determining with high resolution the masses of very large molecules (such as biological molecules), nonlinear optical spectrometers, a high-resolution x-ray photoelectron spectrometer and a high-performance computing center. Laser spectroscopy capabilities will help in studies of clusters. PNL has also joined the Concurrent Computing Consortium, which includes Argonne, Caltech, DARPA, NASA, and NSF, in order to gain access to parallel processing for modeling and simulations.

Environmental remediation and waste management have become watchwords at many of the weapons-production facilities. These facilities all face similar problems in dealing with their accumulated wastes, but the shape of the problem confronted at each facility varies with the particular nature of the waste at that site. DOE is trying to coordinate the efforts at all the national labs so that each can contribute its own particular strength to the massive job. As an example of the cooperation DOE is trying to foster, Frank told us about some work on robotics. DOE has worked with the labs, each of which had some type of robotics program, to define a common architecture. Right now a team from several labs is testing a robot that may eventually be used at Hanford to help retrieve

waste from the single-shell tanks. The tests, which are being conducted in a low-risk environment—silos at the Feed Materials Production Plant in Fernald, Ohio—are to assess the capability of the robot to map the topology of a surface. Frank also described a demonstration project DOE is sponsoring at Savannah River to clean up underground solvents.

Clearly the scale of the nuclear waste problem is staggering. It challenges both researchers and adminis-

trators to find the optimal routes through its murky maze.

—BARBARA GOSS LEVI

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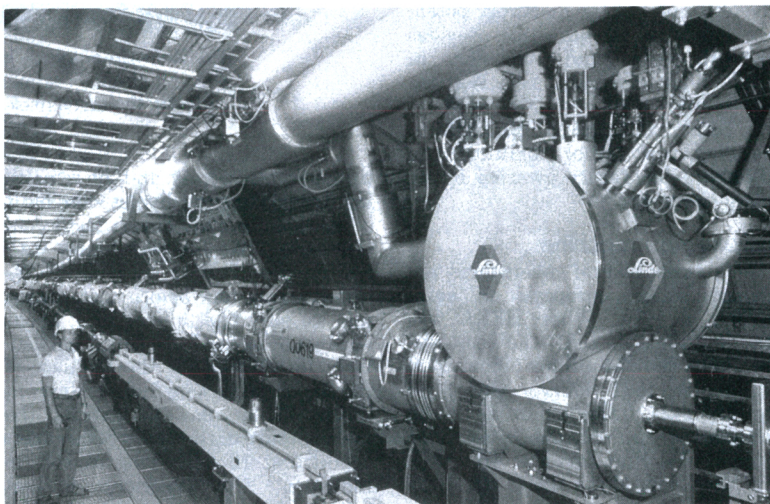
HERA, THE FIRST COLLIDER, WILL

If all goes well, the Hadron-Electron Ring Accelerator that makes its circular way for 6.3 kilometers under the streets and parks of Hamburg will be ready to do physics within the next two or three months. HERA is a unique high-energy storage-ring collider. All the other high-energy colliders, built or pending, make particles collide with others of the same species, or their antiparticles. And the particle energies in the two colliding beams are generally the same.

HERA, by contrast, is thoroughly asymmetric. Its two countercirculating rings look as different as they are. The 820-GeV proton ring requires powerful superconducting bending and focusing magnets with all their attendant cryogenics. Lying rather unobtrusively just below the massive proton magnets in the HERA tunnel is the 30-GeV electron ring. It requires only modest bending magnets. But with regard to synchrotron radiation loss, 6 kilometers is a very tight circle for 30-GeV electrons. The synchrotron radiation loss per trip around an electron storage ring is inversely proportional to the ring's circumference. Therefore it was decided that the radio-frequency accelerating cavities of the electron ring, unlike its magnets, should be superconducting.

Why would anyone take all this trouble to make head-on collisions between 30-GeV electrons and 820-GeV protons? After all, the high-energy physicists already have the 50×50 -GeV LEP electron-positron collider at CERN and the 900×900 -GeV proton-antiproton Tevatron collider at Fermilab. But what nobody has had before HERA was a means of colliding leptons with hadrons at center-of-mass energies above 30 GeV.

Deep inelastic scattering of leptons (electrons, muons and neutrinos) off protons and neutrons is the particle physicist's principal means of probing



The HERA tunnel houses two very different beam lines. The bottom row of conventional magnets bends the 30-GeV electron beam. Above it are the superconducting magnets that bend the 800-GeV proton beam, cooled by the helium transfer line on top.

the quark and gluon structure of hadrons. "Deep" refers to momentum transfers large enough to ensure that the lepton is scattered by an individual quark rather than by the hadron as a whole. The larger the momentum transfer, the smaller is the distance scale probed by the scattering lepton. The kinematic upper limit on the Lorentz-invariant magnitude of the momentum transfer reachable in a particular high-energy experiment is the center-of-mass energy of the colliding lepton and nucleon.

Without HERA, the most energetic lepton-nucleon collisions one can get come from 500-GeV muon beams hitting fixed targets at Fermilab. Electron beams for fixed-target experiments are limited to 50 GeV. Replacing a fixed hydrogen target by a high-energy beam of protons is an

enormous step forward. The HERA collider's 314-GeV center-of-mass energy will provide ten times the highest momentum transfers previously obtainable in lepton-hadron scattering, permitting experimenters to probe the nucleon down to 10^{-17} cm. On that distance scale they might even find that quarks themselves, which the standard theory of the hadronic interactions takes to be rigorously point-like, might have internal structure of their own. On the other hand, the Tevatron collider has already provided significant, if somewhat indirect, evidence¹ that quarks are indeed point-like down to 3×10^{-18} cm.

With momentum transfers on the order of 100 GeV, electron-nucleon scattering at HERA also offers unprecedented opportunities to test the standard unified theory of the weak and electromagnetic forces, which