Challenges in Plutonium Science

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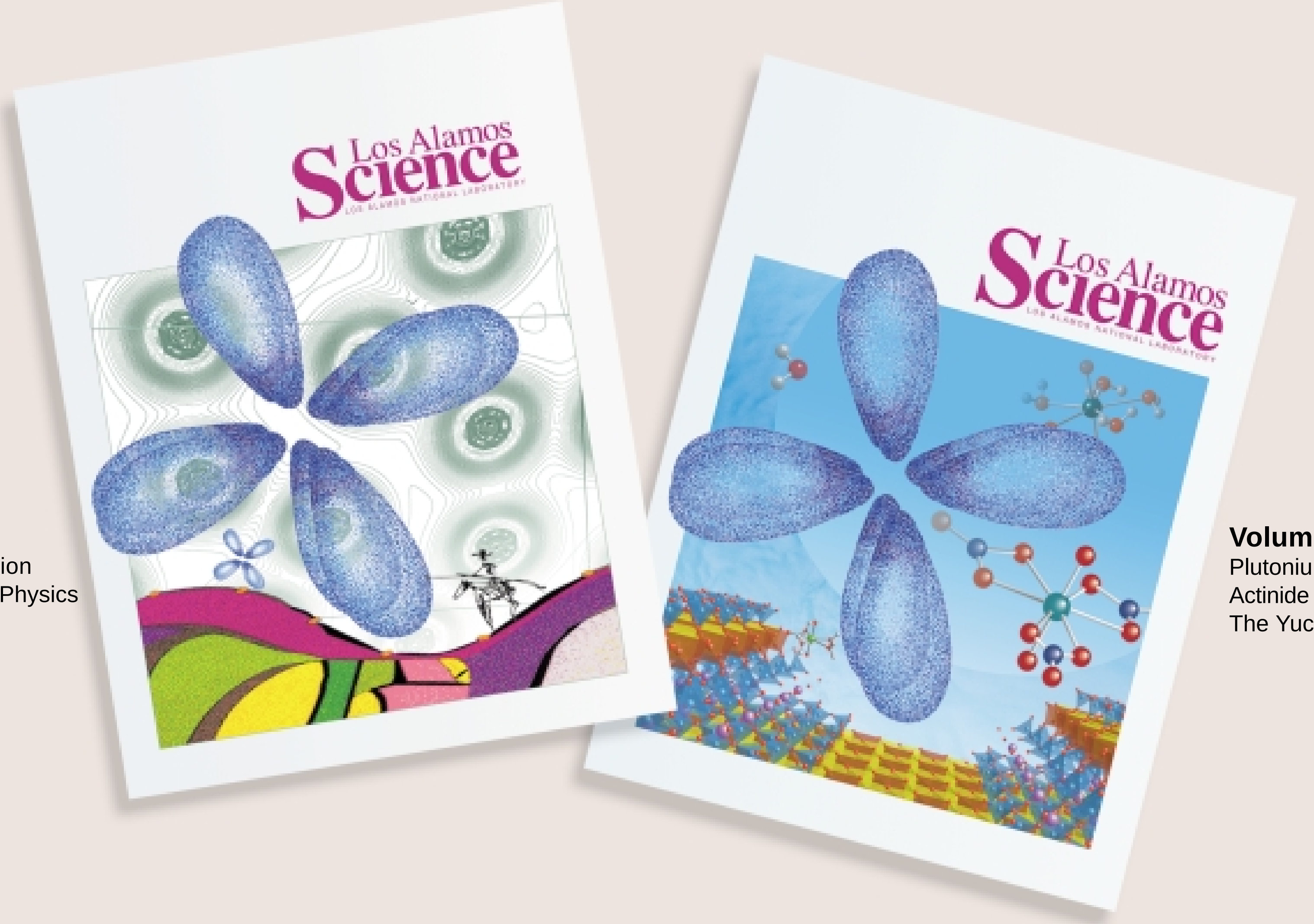

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Actinide Chemistry and the Environment
The Yucca Mountain Project

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Element 94 has baffled scientists since the days of the Manhattan Project. When the first ponderable quantities of reactor-made plutonium arrived at Los Alamos in 1944, the pure metal showed wildly differing densities, and the molten state was so reactive that it corroded almost every container it came in contact with. The engineering challenge was to prepare a stable metallic form and fashion it into a pair of hemispheres for the first implosion nuclear bomb. That task was accomplished in about 15 months, a remarkable feat considering how little was known about plutonium at the time.

Experimental work since then has revealed the unusual ground-state structure of plutonium, its seven distinct crystallographic phases, its dimensional changes with temperature, pressure, and impurity content, its ability to combine with virtually every other element, its pyrophoricity, its multitude of oxidation states, its highly anomalous resistivity, its resemblance to heavy-fermion compounds and other correlated-electron materials, and on and on. This body of data has only added to plutonium's reputation for being the most perplexing element in the periodic table and, arguably, the most interesting. The solid-state, chemical, and metallurgical properties appear to be in a class of their own and, until recently, much too complex to be understood from first principles.

Ironically, the fissile properties of plutonium-239, which render it useful for nuclear bombs and nuclear reactors, are not nearly as difficult to understand. In fact, those nuclear properties had been predicted even before plutonium was made and isolated at Berkeley in 1941 and well before Enrico Fermi and Leo Szilard demonstrated the first self-sustaining fission chain reaction in December 1942.

The inspiration for this issue of *Los Alamos Science* is threefold. First, recent developments in understanding the chemistry and condensed-matter physics of plutonium at a fundamental level call for even greater involvement. Second, three missions of Los Alamos National Laboratory—stewardship of the nuclear weapons stockpile, reducing the threat of nuclear weapons proliferation, and cleanup of the nation's nuclear weapons complex—demand that we develop a deeper understanding of plutonium. Third, as Los Alamos has the only remaining national facility fully equipped for all aspects of plutonium research, it is our job to document the recent progress and to interest the next generation of scientists in keeping our nation at the frontier of this very difficult but dynamic field.

A few recent Los Alamos achievements will illustrate the new directions in research. Electronic-structure calculations from first principles have finally reproduced the ground-state properties of plutonium, including its atypical crystal structure. These calculations predict that the f-shell electrons of neighboring atoms overlap, but just barely, forming a narrow conduction band, a picture that has been confirmed directly at Los Alamos through the first photoemission measurements of electronic structure. Narrow bands are associated with strong electron-electron correlations, and the next step is to probe the nature of those correlations and their effects on the phase stability of plutonium and its alloys. Resonant ultrasound spectroscopy, a technique developed at Los Alamos, has yielded very accurate measurements of the elastic constants of both the α- and the δ-phase of new and aged plutonium. It should prove very useful in the study of aging effects as well as fundamental properties. X-ray absorption fine-structure (XAFS) spectroscopy is revealing the possible existence of new substructures in plutonium and its alloys and is proving to be extremely useful in characterizing chemical species in the environment. Surface studies are revealing new modes of corrosion in plutonium that must be understood for the safe disposal of plutonium over the long term.

If we can support the experimental work that needs to be done, our scientists are likely to predict, within the next decade, such critical matters as the aging of plutonium in weapons components and the rates at which colloidal forms might carry actinide wastes from particular locations underground. At the same time, the truly unique electronic features of plutonium are challenging established paradigms, and we can expect this element to influence the science of condensed-matter physics for a long time to come.

The larger context for plutonium research is covered in a sweeping overview by former Laboratory Director Sig Hecker. In tracing the history of plutonium on the planet, its use in nuclear weapons and nuclear energy, and the mission-oriented challenges facing our scientists, Hecker emphasizes the particular need to help the Russians safeguard their weapons-grade plutonium and to collaborate with them on health and environmental problems that have resulted from the excesses of the Cold War. The international forces that will determine the future of plutonium on the planet should also be kept in mind. The 200 tonnes of weapons-grade plutonium is dwarfed by the 1000 tonnes of plutonium present in spent fuel from nuclear reactors. Much of the world wants to use that plutonium in civilian power reactors. France and Japan are currently burning MOX fuel (mixed oxides of uranium and plutonium). Even in nuclear reactors now operating in the United States, about a third of the power comes from the fissioning of plutonium that has "grown in" within the uranium fuel through neutron irradiation.

U.S. policy notwithstanding, plutonium is likely to be used and coveted for the foreseeable future, and this Laboratory must continue its historic role—to prepare for all potential uses and abuses of this unique element.

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