Unlocking Physical, Chemical Secrets Of Plutonium

Researchers at Rutgers, The State University of New Jersey, have unlocked some of the physical and chemical secrets of **plutonium**, an element known for its use in atomic weapons and power plant fuel. While the complex nuclear characteristics of plutonium are well-known, it has properties as a metal or a chemical compound that have often left scientists scratching their heads.

Writing in this week's issue of Nature, a prestigious international scientific journal, Rutgers physicists report that the valence electrons – those which control how atoms bond with each other – fluctuate among different orbitals in solid plutonium metal on a very short time scale. In contrast, earlier theories specified fixed numbers of valence electrons in those orbitals. The Rutgers findings help explain some contrary characteristics of plutonium: Unlike many metals, plutonium is not magnetic and not a good conductor of electricity, and it shows greater changes in volume under small changes in temperature and pressure.

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While the authors' findings and study methods are mainly of interest to other researchers seeking clear explanations of complex materials, the knowledge may someday help scientists create safer and more versatile nuclear materials for energy, industry and medicine.

"Previous theories about plutonium's makeup placed a fixed number of valence electrons in the particular orbital we examined, known as the 5f orbital," said Kristjan Haule, an assistant professor of physics and astronomy at Rutgers. "Different theories assigned different numbers of electrons to that orbital – some four, others five and yet others, six. But whatever number the theory prescribed, it remained constant. Each theory could explain some of the element's characteristics, but none could account for all the experimental evidence."

The Rutgers approach abandoned the idea of a fixed or unique number of valence electrons in the 5f orbital. "We revisited the notion of valence in a solid," Haule said. "While it happens rarely in nature, we thought it should be possible for the number of valence electrons to fluctuate among orbitals in atoms that are part of a solid structure."

It turns out that plutonium is especially suited to exhibiting this behavior. The Rutgers physicists determined that almost 80 percent of the time, there are five electrons in the 5f orbital. Almost 20 percent of the time, there are six, and less than 1 percent of the time, there are four.

"A theory that permits fluctuating valence electrons consistently explains properties that scientists observe in laboratory experiments," Haule said, citing recent results using X-ray absorption and electron energy-loss spectroscopy. "In addition, the theory accurately predicts the properties of two neighboring elements, americium and curium, which have similar atomic structures but show greatly differing magnetic and electric properties."

The new approach involves a merger of two existing theories, known as local density approximation and dynamical mean field theory, or LDA+DMFT. Taken separately, they and others fell short in accounting for all of plutonium's observed physical characteristics.

The work done by Haule and his colleagues is in a branch of physics known as condensed matter physics, which deals with the physical properties of solid and liquid matter. In particular, their work focuses on strongly correlated materials, which have strongly interacting electrons and, therefore, can't be described using theories that treat electrons as largely independent entities. The radioactive metals, such as plutonium, and their periodic table neighbors, known as rare earth elements, are examples of strongly correlated materials, with highly localized electrons in their f orbitals. In these elements, most of the physical properties such as electrical resistivity and magnetic characteristics depend on the **f-orbital electrons**. The findings reported in Nature strengthen methods for predicting characteristics of all of these complex materials.-Rutgers, the State University of New Jersey

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