Research Group for Superheavy Elements

Group Leader: Matthias Schädel

The research objectives of this group are to understand chemical and nuclear properties of superheavy elements (SHEs) placed at the uppermost end of the Periodic Table and on the heaviest frontier of the nuclear chart. To clarify chemical properties of SHEs, we investigate valence electronic structures of SHEs through experimental determinations of ionic radii, redox potentials, ionization potentials, and formation of chemical compounds of SHEs. To elucidate limits of stability of the heaviest nuclei, we investigate the shell structure of superheavy nuclei through experimental assignments of proton and neutron single-particle orbitals and through the evolution of nuclear deformation at the highest proton and neutron numbers. Research topics in this fiscal year are summarized below.

Chemical properties of element 104, rutherfordium (Rf), in HCl solution: extraction chromatography using TOPO

From a simple extension of the Periodic Table of the Elements, element 104, Rf, should be a transition element and belong into group 4. Although chemical properties of Rf are expected to be similar to those of lighter homologues Zr and Hf, some significant differences are found in HCl and HF solutions [1–3], which provides valuable information on the valence electronic structure of Rf. In the present work, the extraction behavior of Rf onto a styrenedivinylbenzene copolymer resin modified by trioctylphosphine oxide (TOPO) from 2.0–7.0 M HCl solutions was investigated together with Zr and Hf [4]. TOPO has a chemical structure similar to that of tributylphosphate (TBP) and has a higher basicity value as a donor than that of TBP. The effect of the basicity of organophosphorus compounds on the strength of the formation of a Rf complex was examined by comparing the extraction sequence of the group-4 elements into TOPO with that into TBP [2]. The extraction yields of Rf, Zr, and Hf increased with an increase of HCl concentration, and the sequence of their extraction was Zr > Hf ≥ Rf as shown in Fig. 1. The result indicates that the stability of the $RfCl_4$·2(TOPO) complex is lower than that of the corresponding species of Zr and Hf. A basicity effect in the formation of TOPO and TBP complexes was not observed in the extraction sequence among Rf, Zr, and Hf in HCl solution.

First detailed spectroscopy for short-lived heavy actinide nuclei: the α decay of ^{255}No

Excited states in ^{251}Fm populated via the α decay of ^{255}No were studied in detail through α-γ coincidence and α fine-structure measurements [5]. Spin-parities and neutron single-particle configurations of the excited states in ^{251}Fm as well as the ground state of ^{255}No were unambiguously identified on the basis of deduced internal conversion coefficients, lifetimes of γ transitions, rotational-band energies built on one-quasiparticle states, and hindrance factors of α transitions. In particular, the α fine-structure spectrum (shown in Fig. 2) enabled us to firmly establish neutron one-quasiparticle states in ^{251}Fm and their rotational-band structures. Figure 3 shows excitation energies of neutron one-quasiparticle states in N = 151 isotones. It was found that the energy of the 1/2$^+$[620] state in the N = 151 isotones increases with the atomic number, especially at $Z \geq 100$ (^{251}Fm). This indicates that the N = 152 deformed shell gap size becomes larger as the atomic number increases. We calculated ground-state deformations and energies of one-quasiparticle states in the N = 151 isotones using a macroscopic-microscopic model, and found that the evolution of nuclear deformation involving the hexadecapole (β_4) and hexacontatetrapole (β_6) deformations largely contributes to the single-particle structures in the $Z \geq 100$ and $N \geq 152$ nuclei.

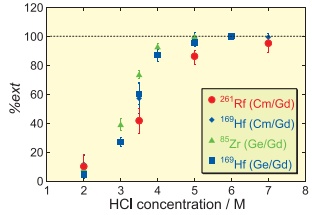

Fig. 1 Extraction of Rf, Zr, and Hf on 0.04 M TOPO as a function of HCl concentration. Cm/Gd and Ge/Gd represent targets used for the production of ^{261}Rf, ^{169}Hf, and ^{85}Zr.

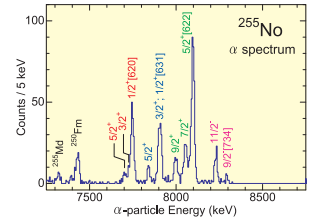

Fig. 2 α -energy spectrum of ^{255}No.

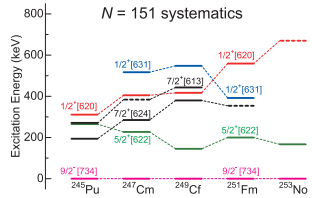

Fig. 3 Excitation energies of neutron one-quasiparticle states in N = 151 isotones.

References

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Research Group for Actinide Materials Science

Group Leader: Zachary Fisk

Many important properties of actinide elements and their compounds are determined by their 5*f* electrons. Because of the large number of degrees of freedom of 5*f* electrons and their sensitivity to chemical/physical environment, a variety of phenomena are realized. In a new compound an actinide experiences a different ligand field, with strong physical effects. Pressure modifies the interatomic distance continuously and this in turn has immediate influence on the 5*f* electronic behavior. Finally the purification of a known compound also provides us a new insight particularly for phenomena occurring at low temperatures, where impurity disorder can deeply disturb and hence hide the intrinsic behavior. Our group uses various techniques for the crystal growth and characterization of actinide compounds including highly radioactive transuranium elements. Hydrostatic pressure will be used to modify the electronic state by tuning the interatomic distances.

Ultra clean sample of URu_2Si_2 and the hidden order

URu_2Si_2 is a uranium-based intermetallic compound crystallizing in a tetragonal crystal structure (Fig. 1). It has two phase transitions. One is the superconducting transition at 1.4 K. Another one occurring at 17.5 K accompanies a thermodynamic anomaly. However the order parameter of the latter one is still unknown despite the huge experimental effort since its discovery in 1985. Using a high-quality single crystal with a very low residual resistivity from impurity scattering, new features were found by utilising novel experimental techniques.

Photoemission spectroscopy is a powerful tool to observe electronic band structure. By using a recently developed high energy-resolution laser photoemission, it was found that a narrow quasiparticle band appears below the hidden-order transition temperature [1]. On the other hand, a cantilever magnetometer measurement succeeded to detect a symmetry-breaking in the hidden order state. The four-fold anisotropy expected for the tetragonal basal plane is changed to the two-fold (orthorhombic) symmetry below the hidden-order transition temperature [2]. These observations provide new insights to the nature of the hidden-order state and accelerate further experimental and theoretical investigations. See 'Research Highlights' for details.

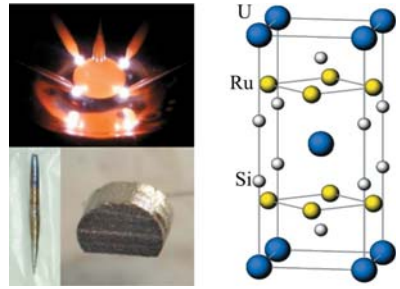

Fig. 1 (left) Czochralski-pulling method and a single crystal of high-quality URu_2Si_2. (right) Tetragonal crystal structure of URu_2Si_2.

Defect structure analysis of fluorite-based solid solutions

A variety of fluorite-based solid solutions are formed between $M^{4+}O_2$ (M = Ce, Th) and $Ln^{3+}{}_2O_3$ (Ln = lanthanide). The lattice parameter data of these $M_{1-y}Ln_yO_{2-y/2}$ solutions exhibit marked deviation from the Vegard's law (i.e., the linear variation with the Ln concentration y), most plausibly originating from the formation of oxygen vacancy (V_o) in the anion sublattice. However, this V_o effect has been neither well recognized nor well modeled. We carefully analysed the lattice parameter data, one of the most fundamental macroscopic quantity, of the solutions, and clarified their 'generalized non-Vegardian' behavior with non-random oxygen coordination around the cations. The present model is thus found to be useful not only as a new quantitative lattice-parameter model but also as a new direct-link to their controversial non-random local defect structure. For example, this can provide a new consistent description of their intriguing ionic-conductivity maxima behavior in low y range [3,4].

Hydrostatic pressure technique for actinide study

High pressure technique is a very useful tool to modify electronic states of actnide compound. Since the properties of actinide compounds are sensitive to the pressure inhomogeniety or uniaxial stress occurring at low temperature, it is necessary to achieve hydrostatic conditions. We have built a diamond anvil cell and selected appropriate pressure medium for the low-temperature and high-pressure experiments up to 10 GPa with nearly hydrostatic conditions [5].

The pressure technique was applied to the uranium-based antiferromagnet U_2Zn_{17} [6]. In general, application of pressure on localized cerium antiferromagnet suppresses the transition temperature at high pressure and drives the system to non-magnetic heavy electron state because of competing two effects, antiferromagnetic exchange and the Kondo interactions. In U_2Zn_{17}, however, the antiferromagnetic transition temperature is almost unchanged up to 5 GPa and gradually increasing above 5 GPa up to 9 GPa. Higher pressure far above 10 GPa is necessary to suppress the transition temperature, in contrast to the typical critical pressure of 2 GPa for the cerium compound with the similar bulk modulus. The weak pressure effect may be ascribed to the itinerant character of 5*f* electrons in this compound, as discussed theoretically in uranium monochalcogenide.

References

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