Trapped nobelium ions yield first direct measurements of transuranic masses

In the search for the island of stability expected to lie not far beyond the heaviest known elements, it will help to have good measurements of nuclear binding energies along the way.

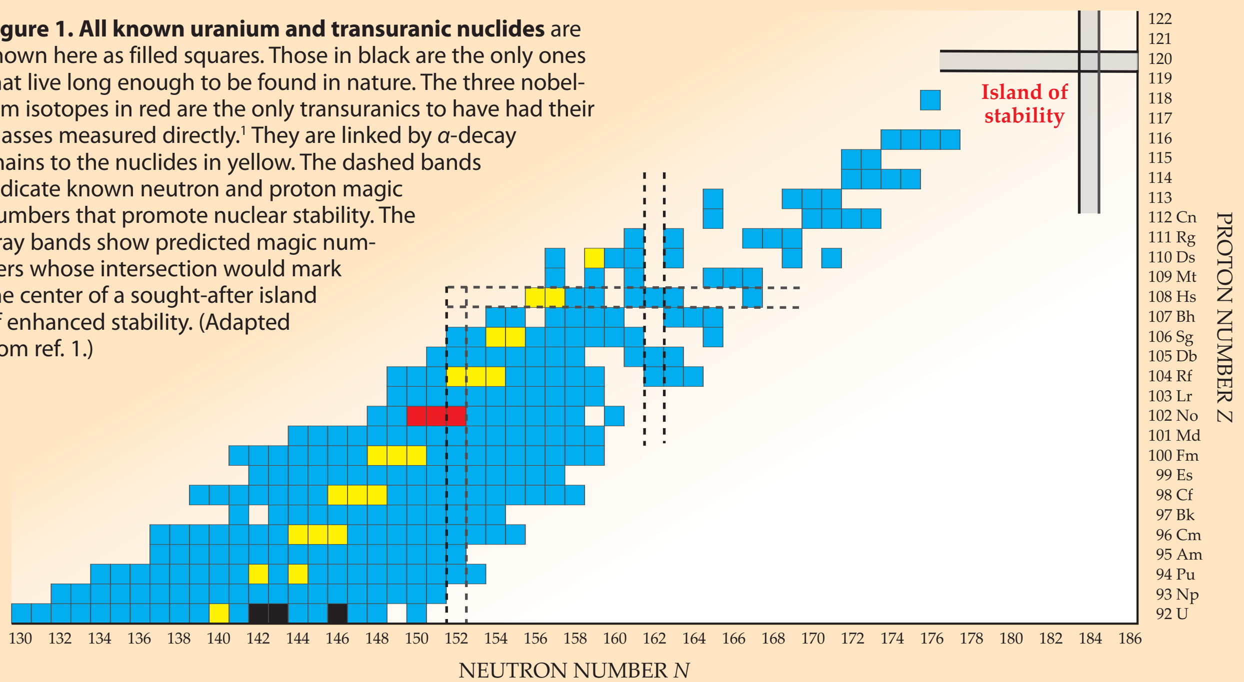
To first approximation, the mass of a nucleus is the sum of its constituent proton and neutron masses. But the roughly 1% correction for nuclear binding energy embodies important information about the configuration of the particular nucleus. Binding energies are especially interesting, and correspondingly hard to measure, in two cases: short-lived isotopes with unusually large or small neutron numbers N for their proton numbers Z, and the transuranic nuclei ($Z > 92$) approaching the so-called island of stability expected to be found somewhere near $Z = 120$ and $N = 184$. (See figure 1.)

Nowadays the best measurements of nuclear masses are done in Penning traps—small, cylindrically symmetric cavities in which charged particles can be kept away from the walls by static electric and magnetic fields and stored for long periods (see PHYSICS TODAY, May 1985, page 17). Beyond its custodial function, the trap's uniform axial magnetic field can reveal the mass of stored particles by the cyclotron frequency with which they orbit the axis. By such means, the masses of stable nuclei have been measured with precisions approaching a part in 10^{11}.

For short-lived nuclei, precision is limited by the time available for determining the cyclotron frequency. Still, Penning traps have yielded the masses of isotopes with half-lives as short as 10 milliseconds. But until now, there has been no measurement of the mass of any transuranic nucleus by Penning trap or any other "direct" means. Nuclear physicists have had to rely on measuring the kinetic energies of α particles emitted in α-decay chains extending down to cisuranic ($Z \leq 92$) isotopes of well-measured mass. In favorable cases, such indirect mass determinations can yield binding-energy uncertainties smaller than 10 keV, more than adequate for comparison with current theoretical predictions. (The nucleon mass is about 1 GeV, and binding energies per nucleon are of order 10 MeV.) But some chains involve α decays to excited intermediate states that confound the energy bookkeeping. And others, especially decay chains starting with the heaviest known transuranics (up to $Z = 118$), terminate in fission before reaching terra cognita.

Now, however, a team at the GSI Helmholtz Center for Heavy Ion Research in Darmstadt, Germany, has reported the first ever direct mass measurement for transuranic nuclear species.[1] Led by Michael Block, the team has measured the masses of three nobelium ($Z = 102$) isotopes in a Penning trap. The half-lives of the three isotopes, ^{252}No, ^{253}No, and ^{254}No, range from two minutes down to two seconds. "That wasn't the principal problem," says Block. "The real challenge was the painfully slow rates at which such nuclei are produced in the lab by fusion reactions."

Figure 1. All known uranium and transuranic nuclides are shown here as filled squares. Those in black are the only ones that live long enough to be found in nature. The three nobelium isotopes in red are the only transuranics to have had their masses measured directly.[1] They are linked by α-decay chains to the nuclides in yellow. The dashed bands indicate known neutron and proton magic numbers that promote nuclear stability. The gray bands show predicted magic numbers whose intersection would mark the center of a sought-after island of enhanced stability. (Adapted from ref. 1.)



Making and catching nobelium

The lightest transuranics can be made in high-flux reactors by neutron capture and subsequent β decay. Beyond fermium ($Z = 100$) however, experimenters have to smash accelerated ion beams into targets and tease out the desired fusion product. To create the several No isotopes, Block and company directed intense 220-MeV beams of calcium-48 ions at lead target foils of isotopically almost pure ^{206}Pb, ^{207}Pb, or ^{208}Pb, depending on which No isotope they were after. The desired reaction yields a fusion of beam and target nuclei minus two liberated neutrons.

Just downstream of the target, a 10-meter-long array of static magnetic and electric fields served as a velocity filter that transmitted the meager yield of emerging No candidates while diverting the torrent of much faster surviving beam ions and debris. With a beam flux of 10^{13} Ca ions per second hitting the ^{206}Pb target, less than one ^{252}No ion per second passed through the velocity filter. For the other two No isotopes, the production rates were only slightly higher.

Emerging from the velocity filter with kinetic energies of about 40 MeV and large charges, the No ions were next injected into a helium gas cell to be thermalized to room temperature and reduced mostly to the +2 charge state. They then entered an ion guide in which RF quadrupole fields accumulated and bunched them for synchronous insertion into the first of two Penning traps in an ongoing sequence of half-second measurement cycles.

Like similar facilities worldwide, the GSI chain of heavy-ion accelerator, fusion target, velocity filter, and Penning traps has been used for some time to measure the masses of relatively rare cisuranic isotopes. In that context, it's become habitual to speak of plural "ions" entering the Penning traps per cycle. But in this first foray beyond uranium, the production rates are so low that a given cycle rarely captures more than one ion. Often it captures none.

Tandem Penning traps

A charged particle in a Penning trap is confined in the trap's axial direction by a weak electric quadrupole field. Radial confinement is accomplished by the strong axial magnetic field B, which keeps ions of mass m and charge q essentially in cyclotron orbits whose frequency

$$\nu_C = qB/2\pi m$$

is independent of an ion's energy—so long as it's nonrelativistic. In the GSI experiment, the trapped ion's ν_C was determined by resonant effects between ν_C and the adjustable frequency ν_{RF} of RF pulses imposed on the cavity.

The two sequential Penning traps in the GSI measurement chain sat together in the uniform 7-tesla field of a superconducting solenoid magnet. Any particle coming from the ion guide was first vetted by the so-called purification trap, which kept interlopers from the second, precision trap by passing on only particles whose ν_C resonated with an RF pulse whose frequency spread was just broad enough to encompass any doubly charged ion within 5 MeV of the desired isotope's mass.

Any ion that made it to the precision trap was then subjected to a narrowband RF excitation pulse before leaving the trap and having its velocity gain measured. The trapped ion experienced resonant acceleration only if the imposed ν_{RF} was very close to its ν_C. Figure 2 shows the result of one four-hour cyclotron-resonance measurement for ^{253}No. As the ν_{RF} passes through ν_C, the ion's flight time to a downstream detector manifests the resonant excitation.

For each of the three No isotopes, Block and company carried out at least three such extended runs. To translate the measured cyclotron frequencies into precise masses, one has to know B with comparable precision or calibrate it with other ions of much better-known mass. Because the magnetic fields of superconductors inevitably suffer gradual decay by what's called flux creep, the GSI team recalibrated the field every few hours with singly charged cesium-133 ions.

Comparing results

Figure 3 compares the No atomic masses thus obtained (after correction for the two missing electrons) with

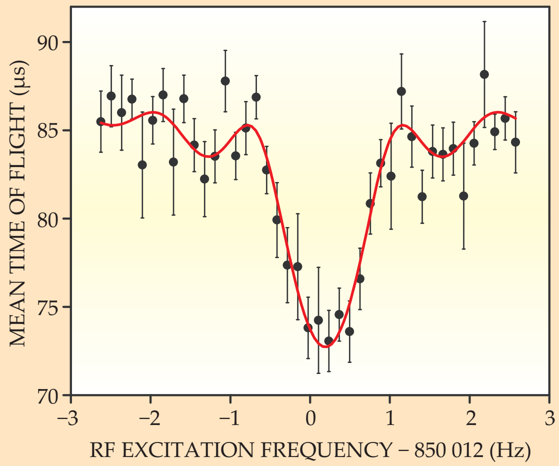


Figure 2. Cyclotron resonance for doubly charged nobelium-253 ions in the 7-tesla magnetic field of a Penning trap at the GSI Helmholtz Center for Heavy Ion Research. The trap reveals ion masses by their cyclotron frequencies. Over many trials, the departing ion's mean flight time to a downstream detector is plotted against the frequency of an imposed RF excitation pulse. Only when the radio frequency approaches the ion's cyclotron frequency does resonant excitation significantly accelerate the ion. The resultant resonant peak of the red curve, a fit of the theoretical line shape to the data, yields the best value of the ion's cyclotron frequency and thus of its mass. (Adapted from ref. 1.)

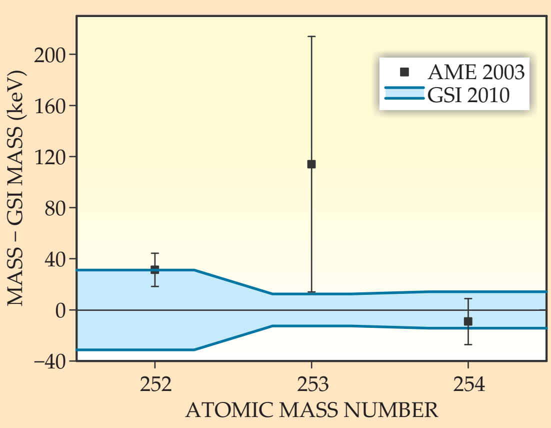


Figure 3. Comparing the Penning-trap mass measurements of three nobelium isotopes at the GSI Helmholtz Center for Heavy Ion Research with the masses determined from α-decay chains and listed in the 2003 Atomic Mass Evaluation compendium. The GSI measurements and their one-standard-deviation uncertainties are represented by the horizontal axis and its enveloping blue swath. (Adapted from ref. 1.)

those given in the international Atomic Mass Evaluation (AME) consortium's most recent (2003) compilation. All three GSI measurements are consistent with the AME masses, which were determined from α-decay chains. The most striking improvement is GSI's eightfold reduction of the 100-keV uncertainty on the mass of ^{253}No. The earlier uncertainty was so large because ^{253}No's α-decay chain involves excited states in fermium-249 and californium-245 whose energies and orderings were unclear. Such troublesome excited states are common in the α decays of "odd–even" nuclei, with neutron and proton numbers of opposite parity.

The other two isotopes, being even–even nuclei, don't have that problem. So the uncertainties on their old AME masses were no worse than those of the new Penning-trap measurements. In fact, for ^{252}No, which had the slowest production rate in the GSI experiment, the 30-keV uncertainty on the new mass is more than twice the AME uncertainty. "Nonetheless," says Block, "the direct weighing of ^{252}No against ^{133}Cs already adds reliability to a mass previously known only indirectly, from α-particle measurements that might involve significant unknown systematic errors."

The GSI experiment was a proof-of-principle demonstration. All three No isotopes had fusion-production rates at least 10 times slower than that of any nuclide previously weighed in a Penning trap. The team hopes soon to reduce uncertainties significantly by raising the overall efficiency, now only about 3%, with which the desired nuclide created in the target makes it all the way through the measurement sequence. An important step will be enlarging and cryogenically cooling the helium gas cell.

The pairing bonus

To get the nuclear mass from the mass of a neutral atom, one has to consider not only the electron's mass (0.51 MeV) but also its binding energy. In the absence of experimental data, the total electromagnetic binding energy of nobelium's 102 electrons has to be estimated theoretically. But the error on that estimate is presumed to be negligible in the context of the Penning-trap measurements.

The nuclear binding energies, per nucleon, of all three No isotopes are near 7 MeV. With the masses of three adjacent isotopes in hand, one gets their neutron pairing energy Δ_n, an important quantity for discriminating between nuclear models. Defined as the difference between the mass of the middle nuclide and the mean of the two that flank it, Δ_n measures the binding-energy bonus gained by the pairing of outer-shell neutrons. For ^{252}No–^{254}No, the GSI experiment yields $\Delta_n = +565 \pm 21$ keV. The sign is positive, as one would expect when the middle isotope has the unpaired neutron.

For the superheavy nuclei approaching $Z = 118$, half-lives get ever shorter and production rates ever slower, eventually precluding mass measurement in Penning traps. But the direct measurement of masses for No and neighbors like rutherfordium ($Z = 104$) and dubnium ($Z = 105$) will provide terminal anchor points for α-decay chains originating as far up as $Z = 116$. And the demonstration that ions as rare as the GSI nobelium isotopes can be thermalized and trapped should make such exotic species accessible to laser and nuclear spectroscopy.

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Reference

1. M. Block et al., *Nature* **463**, 785 (2010).

Optical refrigeration sets solid-state cooling record

Certain high-purity solid materials can be cooled by an all-solid-state laser-based system.

Radiation, along with conduction and convection, is a form of heat transfer. But in some circumstances, radiation can also induce cooling: Laser light can cool dilute gases of atoms whose thermal energy takes the form of relative translational motion. It can also cool some specially prepared solids whose thermal energy is contained in lattice vibrations.

The basic scheme for optical refrigeration of a solid is shown in figure 1a. A laser excites a transition from an upper level of one state to a lower level of another, and a higher-energy photon is emitted, with phonons making up the energy difference. If the pump light is generated by a semiconductor diode laser, the cooling system as a whole has no moving parts or fluids—a particular advantage for spaceborne applications.

So far, every demonstration of optical refrigeration has used a transparent material doped with a rare-earth element—usually ytterbium, but some studies have used thulium and erbium. The surrounding atoms split the dopant ions' two lowest-energy states into several sublevels, as shown in figure 1b.

Now researchers led by Mansoor Sheik-Bahae of the University of New Mexico in Albuquerque and Mauro Tonelli of the University of Pisa in Italy have set an optical refrigeration temperature record.[1] They've cooled a solid sample of Yb-doped yttrium lithium fluoride (Yb:YLF) from room temperature to 155 K—about 10 degrees colder than can be achieved by standard thermoelectric Peltier coolers, previously the best-performing all-solid-state cooling system.

A balancing act

Each optical absorption–emission cycle extracts only a tiny amount of energy from the sample—at most a few percent of the absorbed photon energy. So for cooling to succeed, several processes that heat the sample need to be kept to

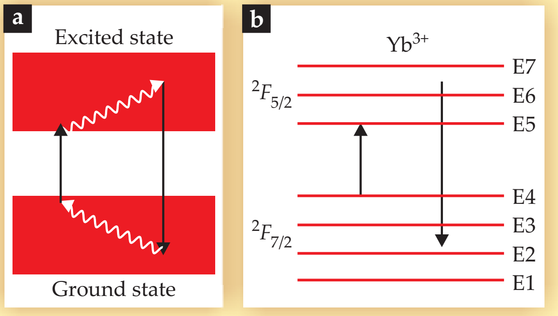

Figure 1. Optical refrigeration shown schematically. **(a)** In the general case, the solid sample absorbs a laser photon (black upward arrow) and emits a photon of higher energy (black downward arrow), with phonons (white arrows) making up the energy difference. **(b)** So far, optical refrigeration has been demonstrated only in solids doped with rare-earth elements such as ytterbium, whose two lowest-energy states are split by the surrounding host atoms into seven sublevels. The intrastate splitting is exaggerated for effect: In Yb-doped yttrium lithium fluoride, the energy difference between levels E4 and E5 is more than 70 times the splitting between E5 and E6.