to the NV spin via its magnetic moment. The resulting change of the spin's quantum state can be detected optically.

The interaction of a single spin and a mechanical resonator has already been studied from a somewhat different perspective: Using an ultrasensitive mechanical cantilever with a magnetic tip, researchers detected single electron spins embedded in a solid material (*10*), a technique known as magnetic resonance force microscopy. The experiment of Kolkowitz *et al*. turns this situation around and uses the spin to sense mechanical vibrations. During the measurement, the quantum state of the spin is controlled by microwave radiation. A clever sequence of coherent manipulation pulses is applied to the spin in orde r to enhance its sensitivity to the vibrations while suppressing noise from other sources. In this way, Kolkowitz *et al*. sense mechanical vibrations down to a few picometers in amplitude.

In a previous experiment on spin-resonator coupling, Arcizet *et al*. coupled an NV spin to the vibrations of a SiC nanowire (*11*). The NV was fixed to the tip of the nanowire while the magnet was placed near to it. They observed nanowire vibrations of a few tens of nanometers in amplitude through a change in the line shape of the NV spin resonance. Kolkowitz *et al*. used the coherent quantum dynamics of the NV spin to reach higher sensitivity to mechanical vibrations.

The resonator motion detected here is in the classical regime at room temperature, but with the magnetic coupling mechanism, it is in principle possible to reach the quantum regime (*6*). Several technological improvements are necessary to reach this goal, including fabrication of a smaller cantilever with a much higher mechanical quality factor and a magnet made from a different material, and working at cryogenic temperatures. Ultimately, the system will have to reach the so-called strong-coupling regime, where the excitation of a single quantum of vibration (a single phonon) in the resonator is sufficient to flip the spin, and a single spin flip is sufficient to excite vibrations of the resonator. In this regime, the system realizes a mechanical analog of current experiments in cavity quantum electrodynamics (*12*), where the internal state of a single atom is strongly coupled to a single quantum of the electromagnetic field (a photon) inside an optical cavity.

A strongly coupled spin-resonator system in the quantum regime bears some analogy with the infamous Schrödinger's cat: The microscopic spin could be prepared in a quantum mechanical superposition state of pointing up and down at the same time. It could then influence the macroscopic mechanical cantilever (playing the role of the cat) so that the cantilever ends up in a superposition state of vibrating in two opposite directions at once. Although some caution is in order—the cantilever contains a macroscopic number of atoms, but a superposition state involving only a few phonons is still in some sense microscopic—such an experiment would certainly be intriguing. From an applied perspective, the use of a single spin, the most elementary quantum system, to read out mechanical vibrations opens the path to force detectors operating at the ultimate limits of sensitivity. The experiment of Kolkowitz *et al*. is an exciting first step in this direction.

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GEOCHEMISTRY

Keeping Time with Earth's Heaviest Element

New $^{238}U/^{235}U$ ratios for uranium-bearing minerals provide a better chronometer for dating geological processes.

Claudine H. Stirling

Uranium is the heaviest naturally occurring element on Earth. It has three natural isotopes (^{238}U, ^{235}U, and ^{234}U), of which ^{238}U and ^{235}U are the parent nuclides of the ^{238}U- and ^{235}U-decay series chains, which ultimately decay to stable isotopes of lead (Pb), thereby forming the basis of the U-Pb chronometer. Conventional theories of stable isotope fractionation have dictated that uranium is too heavy to display resolvable mass-dependent isotope effects. The expectation was that Earth would display homogeneous $^{238}U/^{235}U$ isotopic compositions. The convention has been to adopt an invariant present-day $^{238}U/^{235}U$ ratio equal to 137.88 throughout the solar system, on the basis of early studies of uranium ore deposits. This critical assumption, which underpinned the veracity of the U-Pb chronometer for the past 30 years, was overturned by the discovery of surprisingly large $^{238}U/^{235}U$ variations in Earth's surface environments (*1*, *2*). On page 1610 of this issue, Hiess *et al*. (*3*) report the $^{238}U/^{235}U$ composition of a large suite of U-bearing accessory minerals to facilitate a more accurate U-Pb geochronometer. These new results also provide fundamental but unexpected insights into the mechanisms controlling $^{238}U/^{235}U$ fractionation.

Together with the other actinides, uranium was produced over the lifetime of the galaxy in massive exploding stars (supernovae) and was then incorporated into Earth during the formation of the solar system more than 4.5 billion years ago. In the past decade, high-precision measurements of $^{238}U/^{235}U$ were initially driven by the search for the former existence of extinct ^{247}Cm (*4*, *5*), the short-lived precursor nuclide of ^{235}U. ^{247}Cm is a crucial actinide for evaluating models of solar system formation and is detected as small positive anomalies in the $^{238}U/^{235}U$ ratio of extraterrestrial meteorites (*6*). Contrary to expectations, initial measurements of $^{238}U/^{235}U$ in terrestrial reference materials revealed surprisingly large isotopic variability of approximately 1 per mil (‰) in a wide range of low-temperature environments (*1*, *2*). These isotopic shifts

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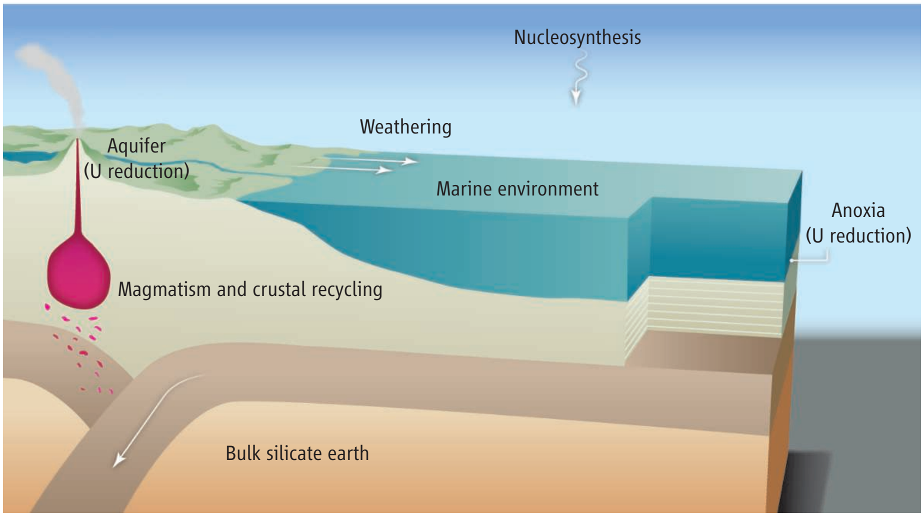

Uranium $^{238}U/^{235}U$ cycling in the environment. Most terrestrial and extraterrestrial materials formed in high-temperature environments define the $^{238}U/^{235}U$ composition of "unfractionated" bulk silicate Earth. Superimposed on bulk silicate Earth are natural variations in $^{238}U/^{235}U$ in some other environments: Percent-level excesses in ^{235}U and low $^{238}U/^{235}U$ values are found in some mineral inclusions of extraterrestrial meteorites; in the hydrological environment, uranium reduction leads to "light" $^{238}U/^{235}U$ signatures in the unreacted U(VI) water and "heavy" compositions in aquifer deposits and organic-rich seafloor sediments containing reduced U(IV); leaching of exposed minerals during chemical weathering results in marginally low $^{238}U/^{235}U$ values in the modern marine environment (including seawater precipitates). Some materials fractionated in these and other low-temperature processes are recycled at depth through crustal recycling. Additional $^{238}U/^{235}U$ fractionation likely operates at magmatic temperatures.

cannot be explained by conventional theories of stable isotope fractionation. Instead, they appear to be largely controlled by the mass-independent nuclear field shift effect (*7*, *8*), which arises from varying nuclear volumes and electron density distributions between different isotopes of the same element. Isotopic fractionation governed by the nuclear field shift favors heavy isotopes in the lower oxidation state where the electron density near the nucleus is lower, and is predicted to have a strong effect on the heavy masses.

Natural variability in $^{238}U/^{235}U$ will undoubtedly lead to revision of some critically important cosmochronological and geochronological models. Coupled measurements of $^{238}U/^{235}U$ and $^{207}Pb/^{206}Pb$ have already begun in the field of cosmochronology, where models of solar system formation are often based on relative age differences of only a few million years (*9*). Hiess *et al.* have initiated efforts in the terrestrial realm by characterizing the $^{238}U/^{235}U$ composition of a diverse range of U-bearing minerals that are essential in U-Pb geochronology. These new data display extremely large $^{238}U/^{235}U$ isotopic shifts exceeding 5‰. The most extreme compositions are displayed by titanite and monazite, and more uniform $^{238}U/^{235}U$ signatures are observed in zircon. The implication is that previous U-Pb ages for these accessory minerals may be inaccurate by up to 10 million years. Hiess *et al.*'s study reemphasizes the need to independently characterize the $^{238}U/^{235}U$ composition of every sample to obtain accurate U-Pb chronologies. Because this new data set is traceable to the International System (SI) of units, it can facilitate a revised, albeit provisional, estimate of the decay constant for ^{235}U.

Efforts in the field of uranium "stable" isotope geochemistry have also focused on gaining an improved understanding of the mechanisms controlling $^{238}U/^{235}U$ fractionation in low-temperature environments where the largest isotopic shifts have occurred. In this context, the U(VI)-U(IV) redox transformation has received considerable attention (*1*, *2*, *10*–*13*). Specifically, the reduction of uranium in water results in a dramatic removal of dissolved U. In parallel, a corresponding isotopic shift toward depleted $^{238}U/^{235}U$ signatures occurs in the unreacted U(VI) water because of the nuclear field shift effect. Therefore, the ^{238}U-^{235}U system already demonstrates considerable promise as a tool to monitor redox conditions in U bioremediation studies (*11*) and as a paleo-redox tracer to quantify the extent of anoxia in the historical oceans (*14*).

Hiess *et al.* extend our understanding of the mechanisms controlling $^{238}U/^{235}U$ fractionation for several reasons. First, the reported data set unequivocally shows that $^{238}U/^{235}U$ variability is not limited to low-temperature settings but is also prevalent in higher-temperature magmatic environments, and strongly implies that material fractionated through low-temperature processes is incorporated into higher-temperature systems through crustal recycling (see the figure). Second, the magnitudes of $^{238}U/^{235}U$ isotopic shifts in high-temperature magmatic environments appear to rival those in lower-temperature systems. Third, their compilation of all previously published $^{238}U/^{235}U$ observations to provide a global, SI-traceable data set allows for a rigorous comparison among laboratories and reveals the true complexity of the ^{238}U-^{235}U system. The majority of samples show a relatively uniform $^{238}U/^{235}U$ composition that gives rise to an absolute $^{238}U/^{235}U$ value of 137.818 ± 50 for "bulk silicate Earth." However, there is a 0.4‰ enrichment in ^{235}U over ^{238}U in the present marine environment that appears to be driven by the long-term chemical weathering of exposed crustal rocks and preferential release of lattice-bound ^{235}U from the leaching of exposed minerals. This surprising result corroborates earlier findings (*1*) and gives rise to a correlation between the relative abundance of ^{235}U and ^{234}U in some low-temperature environments, even though the excess ^{234}U is not lattice-bound and need not be correlated with ^{235}U release at all. The study by Hiess *et al.* therefore supports the idea that $^{238}U/^{235}U$ variability is not exclusively controlled by the U(VI)-U(IV) redox transformation and will undoubtedly drive future directions in uranium "stable" isotope geochemistry.

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