

# light & MATTER



It's *Triplets* p.1



Some “Crazy Hat Day” participants at JILA on January 25, 2018. Credit: Catherine Klauss, JILA.

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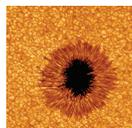
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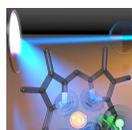
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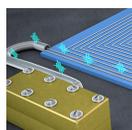
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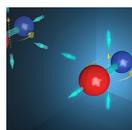
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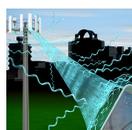
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# It's *Triplets!*

Newly minted JILA Ph.D. Catherine Klauss and her colleagues in the Jin and Cornell group decided to see what would happen to a Bose-Einstein condensate of Rubidium-85 ( $^{85}\text{Rb}$ ) atoms if they suddenly threw the whole experiment wildly out of equilibrium by quickly lowering the magnetic field through a Feshbach resonance<sup>1</sup>. Theoretically, this maneuver is predicted to make the atoms infinitely attracted to each other, and at the same time, infinitely repulsed by each other.

"This is a really crazy regime, and things happened really fast," explained Klauss. "At this resonance, the energy of the atom pairs equaled the energy of molecules, and the interactions were going on like crazy."

At first, Klauss and her colleagues thought they were losing most of the atoms in the experiment. However, they soon discovered the atoms were actually still there even though the researchers couldn't see them any longer. The atoms had been transformed into molecules, which had to be probed differently.

Once the researchers realized they'd made molecules, they decided to study them. First, they held the molecules at a specific magnetic field and watched them decay away by turning back into atoms. But, no matter how many times they repeated the experiment, there was always a two-component decay: a fast one and a slower one. The slower decay varied with the density of the initial

atom sample, which was expected for a two-atom molecule, or dimer ( $^{85}\text{Rb}_2$ ).

But the initial decay was happening much too fast to involve dimers. After consulting with JILA theorist José D'Incao, Klauss and her colleagues concluded they were making three-atom molecules, or trimers. And, the trimers were almost certainly the Efimov molecules ( $^{85}\text{Rb}_3$ ) that have been studied theoretically for nearly 50 years, including

work by D'Incao over the past decade. In this experiment, about 8% of the ultracold  $^{85}\text{Rb}$  atoms in the original BEC formed the exotic Efimov molecules.

"This is the first direct observation of Efimov molecules in an ultracold gas that we've already positively identified," Klauss said.

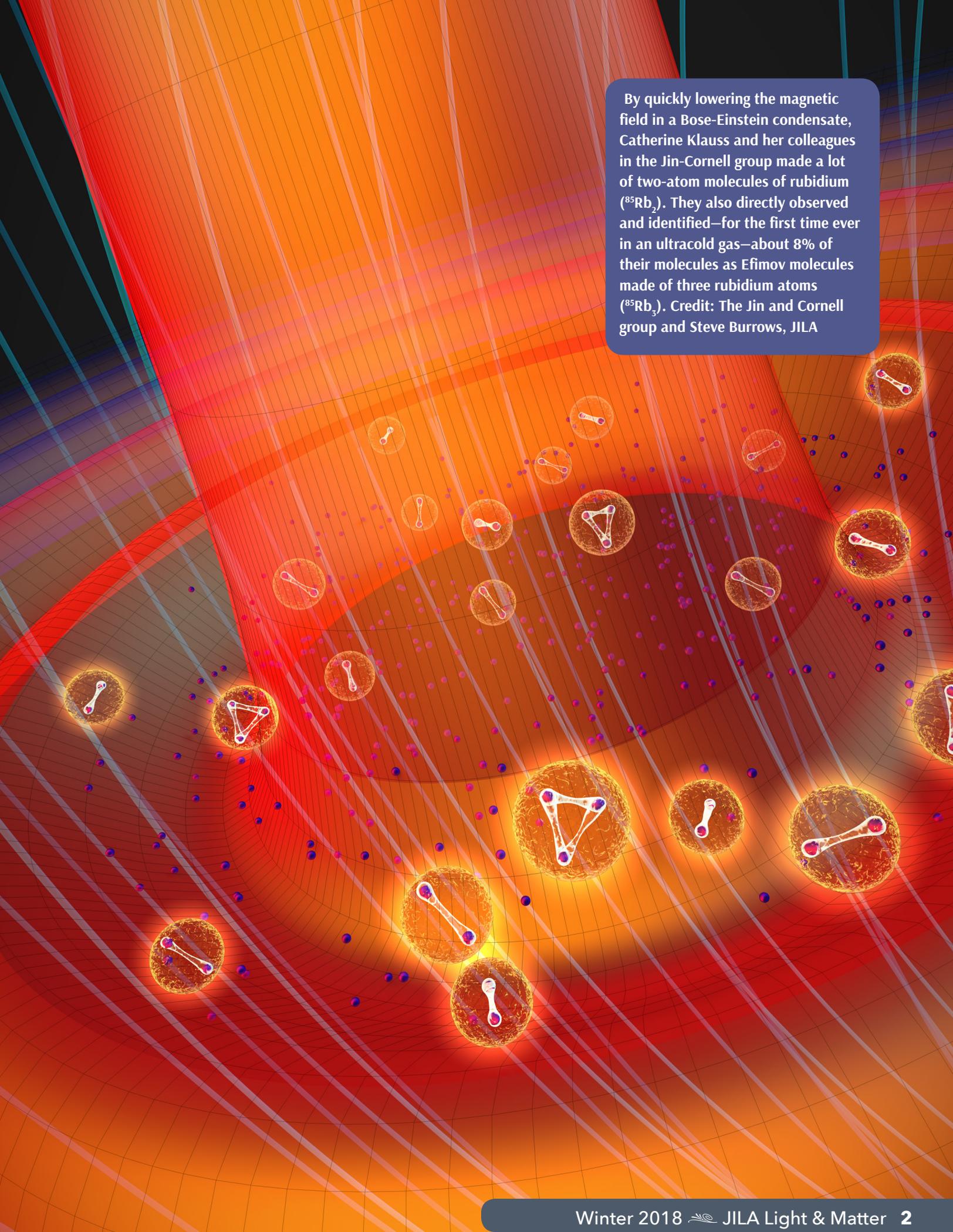
"You can tell these molecules apart from dimers because the Efimov trimers die faster. José's theory predicted that Efimov trimers would have a lifetime of about 100 microseconds ( $10^{-4}$  s), and that's exactly what we see in the lab!"

The researchers responsible for discovering and investigating the  $^{85}\text{Rb}$  triplets included Klauss, graduate student Xin Xie, University of Colorado Boulder undergraduate student Carlos Lopez-Abadia, senior research associate José D'Incao, Fellows Deborah Jin and Eric Cornell as well as Zoran Hadzibabic of the University of Cambridge. ✨

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Catherine E. Klauss, Xin Xie, Carlos Lopez-Abadia, José P. D'Incao, Zoran Hadzibabic, Deborah S. Jin, and Eric A. Cornell, *Physical Review Letters* **119**, 143401 (2017).

1. Near a Feshbach resonance, small changes in the magnetic field have dramatic effects on the interactions of atoms in an ultracold gas.



By quickly lowering the magnetic field in a Bose-Einstein condensate, Catherine Klauss and her colleagues in the Jin-Cornell group made a lot of two-atom molecules of rubidium ( $^{85}\text{Rb}_2$ ). They also directly observed and identified—for the first time ever in an ultracold gas—about 8% of their molecules as Efimov molecules made of three rubidium atoms ( $^{85}\text{Rb}_3$ ). Credit: The Jin and Cornell group and Steve Burrows, JILA

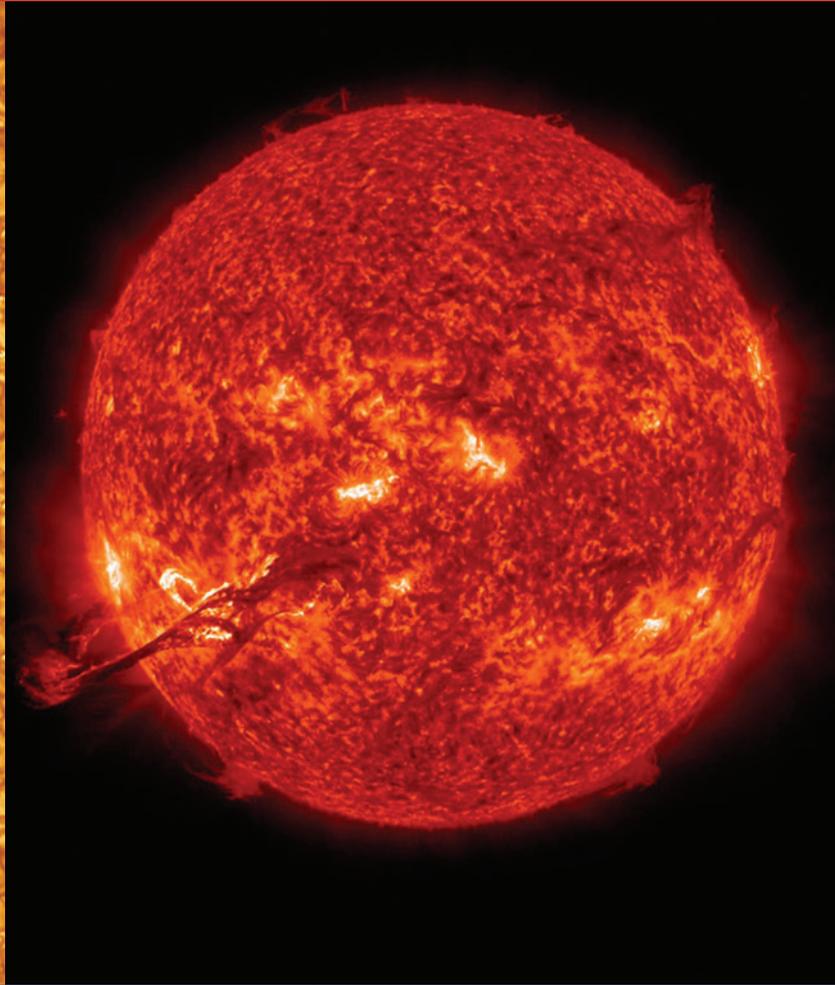
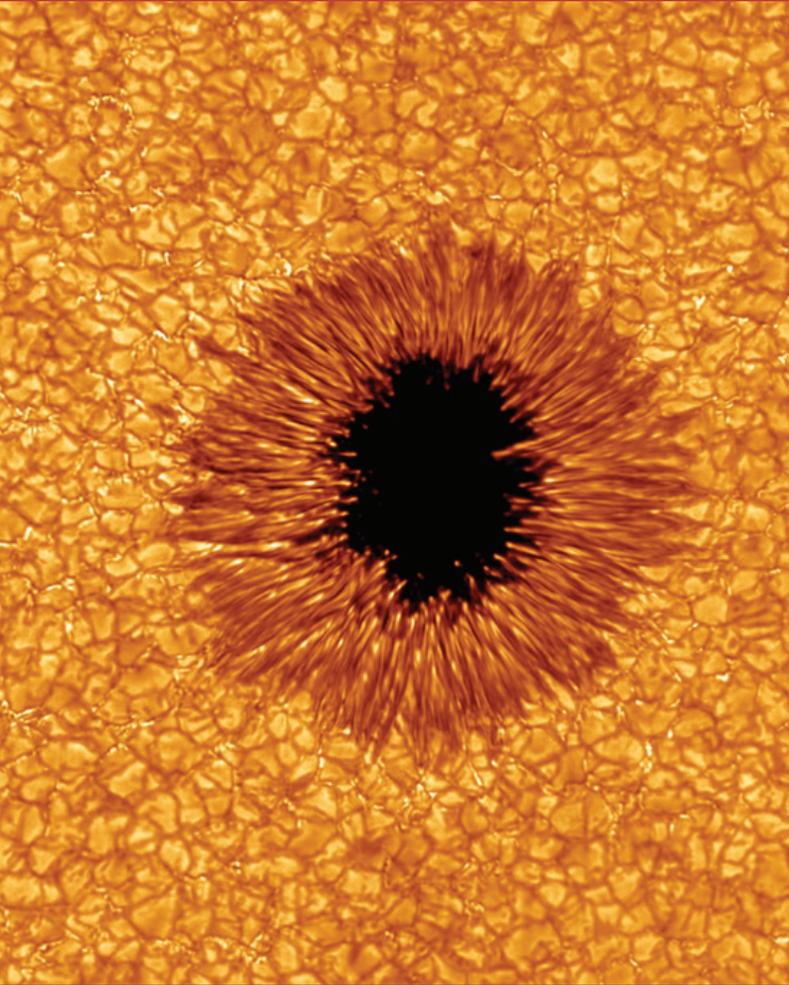


Figure (left): Sunspot on the surface of the Sun surrounded by megameter-sized granules. Credit: Big Bear Solar Observatory

Figure (right): Solar Dynamics Observatory photograph of the Sun's corona. Deep inside the Sun, hurricane-like swirling columns generate megameter-sized granules and 30 megameter-sized supergranules that appear under the corona on the Sun's surface. Credit: [sdo.gsfc.nasa.gov](http://sdo.gsfc.nasa.gov)

## SOMETHING NEW UNDER THE SUN

The Sun isn't working the way we thought it did. Many astrophysicists haven't actually understood one aspect of how the Sun worked—until former senior research associate Nick Featherstone and senior research associate Brad Hindman set the record straight.

Stars like the Sun have to get rid of the heat generated by thermonuclear reactions in their centers. The Sun's secret is vigorous convection, particularly in the outer third of the Sun closest to its surface. Like a pot of boiling water, hot fluid moves upward and cooler fluid moves downward, carrying heat outwards toward the surface of the star. For a long time, astrophysicists thought these motions came in two different sizes called granules (because these megameter-sized structures looked like grains of rice through early telescopes) and giant cells.

The trouble with the giant cell idea is that while granules appear on the Sun's surface, giant cells have never been seen in spite of the fact astronomers have been looking for them for a long time. And, if giant cells exist, they would be 200 megameters<sup>1</sup> wide, which is definitely large enough to be detectable. For instance, the largest cells seen in the Sun, called supergranules, are only about 30 megameters wide. Featherstone's and Hindman's new theory explains why supergranulation appears and why giant cells are absent.

"I've been thinking about this problem for about 10 years," Hindman said. "About two to three years ago, I decided we should accept the huge volume

of observational data that has never detected a single giant cell to mean that they aren't there."

Hindman and Featherstone used some fancy math and numerical simulations that included rotation and coriolis forces to see what might actually be generating supergranules on the surface of the Sun. Much to their surprise, they discovered multiple column-like structures not unlike spinning hurricanes on Earth.

**Thus it seems, the Sun has two distinct layers of convection: (1) a deep layer where little spinning columns form, and (2) a layer near the surface where flows are too fast to form columns, so they form granules and supergranules.**

"There is nothing like this on the surface of the Sun," Hindman explained. "Instead they exist below the surface in the top third of the Sun, but why?"

The answer is simple. Deep layers of the Sun have fluid motions with the same time scale as the Sun's period of rotation. These deep-layer structures are prone to spin, unlike what happens near the surface where lateral flows are much faster. And, the columns must be smaller than 30 megame-

ters in radius, or telescopes would have detected them long before now.

Thus, it seems, the Sun has two distinct layers of convection: (1) a deep layer where little spinning columns form, and (2) a layer near the surface where flows are too fast to form columns, so they form granules and supergranules.

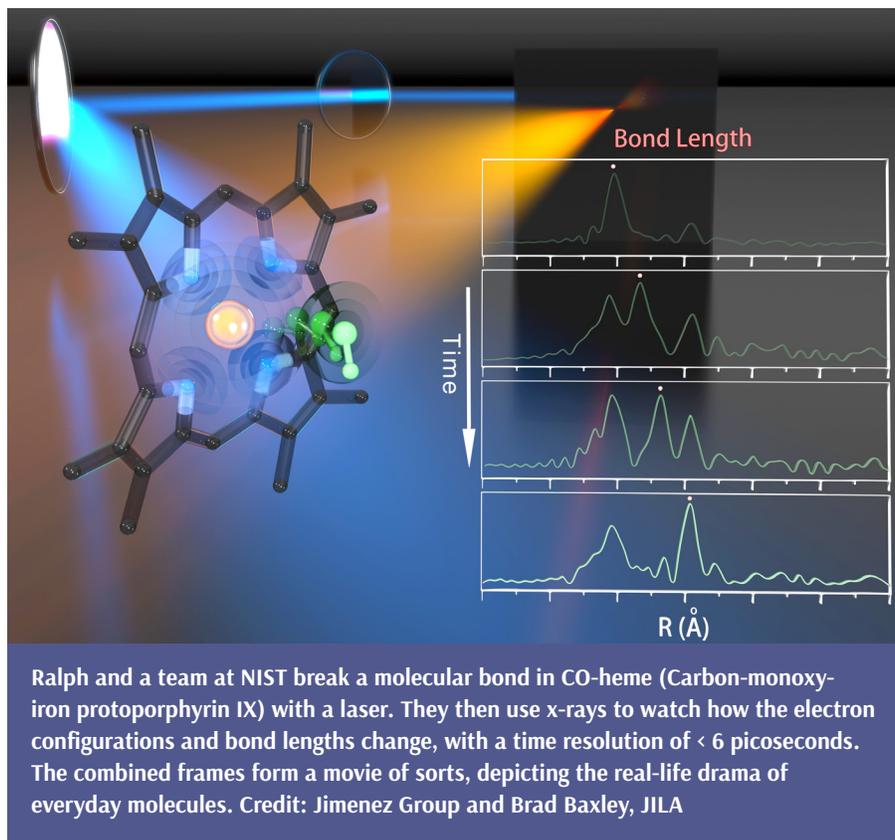
The conclusion is clear: The Sun has no giant cells, which is why no one has ever seen them.\*

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Nicholas A. Featherstone and Bradley W. Hindman, *The Astrophysical Journal Letters* **830**, L15 (2016).

1. A megameter is 1000 kilometers, and one mile is about 1.6 kilometers.

# Molecule Movies, Now Filming at NIST



The actors are molecules. The plot, broken molecular bonds. JILA Fellow Ralph Jimenez and a team of detector experts at the National Institute of Standards and Technology (NIST) are working together to make x-ray movies of a molecular drama.

The team at NIST built a microcalorimeter x-ray spectrometer capable of performing time-resolved spectroscopy; in other words: a camera to film molecules. They use this camera to learn how molecules break their bonds—do the electrons rearrange, do the other atoms quake?

The microcalorimeter spectrometer is not the only tool that can film molecules, but it is the smallest. Previously, only huge, multibillion-dollar synchrotrons were capable of producing x-rays and probing molecules. Beam time at these facilities has to be reserved in advance, sometimes months, or even years. These long wait times, combined with the expense of building and operating synchrotrons, can limit opportunities for new research.

In contrast, NIST's spectrometer fits on a tabletop and costs a fraction of an entire synchrotron system. But most importantly, NIST's spectrometer is easily accessible—it's like having the ability to film a Hollywood movie on your smartphone. Jimenez hopes that this new device will "broaden access and enable a wider range of molecules to be studied."

Despite its small size and modest cost, the NIST spectrometer is faster and more efficient than previous systems. The new spectrometer combines ultrafast x-ray pulses with an array of cryogenic microcalorimeter detectors. The microcalorimeter detectors are superconductors held at their transition temperature, where the resistance is maximally sensitive to temperature. When one of the superconductors absorbs an x-ray photon, its temperature is raised, and the energy of the x-ray photon can be deduced from the change in resistance. This enables the microcalorimeter array to count individual emitted x-rays and simultaneously measure their energies.

While a fascinating tool, you can't make a movie without actors. Jimenez is the casting director of the experiment, and writes the leading roles into the scripts. But not every molecule is charismatic enough for the big screen. "I tell them the reality," said Jimenez, "what science you can actually do with the system." Jimenez worked tirelessly to find a worthy molecule, considering "how much light it absorbs, signal to noise, structure change ... if we broke this bond, would it give a large enough signal? We can't just pop a molecule in the experiment and see it." So far, all of the molecules used have had a metal atom.

To make a molecular movie, the film crew first breaks a molecular bond with a laser. They then blast that same laser onto a material to create a plasma (a gas of free electrons). The free electrons collide into each other, causing them to emit x-rays. The result is an x-ray light bulb that glows over a fairly broad range.

When the x-ray light bulb shines over the newly broken molecule, it excites transitions in the core electrons of the metal atom. The timing between when the molecule breaks and when the x-rays probe can be delayed with picosecond (trillionth of a second) resolution. "And that's the kind of resolution you need to see molecular motions," said Jimenez.

Depending on the state of the core electrons, a specific energy of x-rays is absorbed by the metal atom in the molecule. This absorption spectrum can then be analyzed to determine the electronic state of the metal atom, and the positions of the other atoms in the molecule. This means that the team can see whether the electrons move around before a bond breaks, and watch the other bond lengths change. These measurements are repeated with picosecond time resolution, and the combined frames create a molecular movie.

With their setup, the NIST team has already made two new films. One film, documenting a bond

breaking in a ferrioxalate complex, settled a controversy in the literature about an important atmospheric chemistry reaction. Because molecules with heavy atoms, such as a ferrioxalate complex, have multiple oxidation states, they are useful for storing both energy and data. Filming how these molecules react to chemical changes can help develop more energy-efficient batteries, computer memories, and optical display technologies.

With a couple of molecular movies under his belt, Jimenez's casting is ready to move from high-school drama kids to Hollywood hotshots. "We plan to do in vitro measurements of purified proteins...we're going to try it this fall." But filming proteins no easy task. Proteins are much larger than the previously filmed molecules. And, as anyone who has had a medical x-ray knows, human bodies, and the proteins they are made of, are mostly transparent to x-rays. Many of our proteins, however, contain iron, an atom easily probed by NIST's detector. Jimenez is not one to back down from a challenging experiment: "I think everyone who was on the team said, 'these were the hardest experiments we've ever done.'"

The NIST detector experts (camera crew) are Joe Ullom, William Doriese, and Dan Swetz, as well as the tireless film crew, Luis Miaja-Avila and Galen O'Neil. Ralph Jimenez recruited casting assistance from Niels Damrauer and JILA postdoc D. M. Sagar. The team's recent work was published in *Physical Review X* and the *Journal of Physical Chemistry Letters*.✱

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Luis Miaja-Avila, Galen C. O'Neil, Young I. Joe, Bradley K. Alpert, Niels H. Damrauer, William B. Doriese, Steven M. Fatur, Joseph W. Fowler, Gene C. Hilton, Ralph Jimenez, Carl D. Reintsema, Daniel R. Schmidt, Kevin L. Silverman, Daniel S. Swetz, Hideyuki Tatsuono, and Joel N. Ullom, *Physical Review X* **6**, 031047, (2016).

Galen C. O'Neil, Luis Miaja-Avila, Young II Joe, Bradley K. Alpert, Mahalingam Balasubramanian, D. M. Sagar, William Doriese, Joseph W. Fowler, Wilfred K. Fullagar, Ning Chen, Gene C. Hilton, Ralph Jimenez, Bruce Ravel, Carl D. Reintsema, Dan R. Schmidt, Kevin L. Silverman, Daniel S. Swetz, Jens Uhlig, and Joel N. Ullom, *The Journal of Physical Chemistry Letters* **8**, 1099–1104 (2017).

# A NEW QUANTUM DRUM REFRAIN

Quantum computers require systems that can encode, manipulate, and transmit quantum bits, or qubits. A creative way to accomplish all this was recently demonstrated by Adam Reed and his colleagues in the Lehnert group. The researchers converted propagating qubits (encoded as superpositions<sup>1</sup> of zero and one microwave photons) into the motion of a tiny aluminum drum. The successful conversion is considered a key step in using a mechanical drum to (1) transfer quantum information between microwave and optical frequencies or (2) store quantum information inside a quantum computer.

“We wanted to ask whether these electromechanical devices could manipulate fragile signals that encode quantum information,” Reed explained. “We’re trying to determine how suitable this device is for quantum information applications.”

And the answer is: The Lehnert group is getting there. When the researchers measure how the state encoded in the drum’s motion compares with the original propagating qubits, they find that the fidelity is about 80% for any qubit state they convert, which is not too bad for a brand new kind of device.

One long-range goal of this project is to use the mechanical drums as converters of information that link between microwaves and visible light. These converters will play a key role in future quantum networks, which will have vastly increased bandwidth and security. These networks will rely on

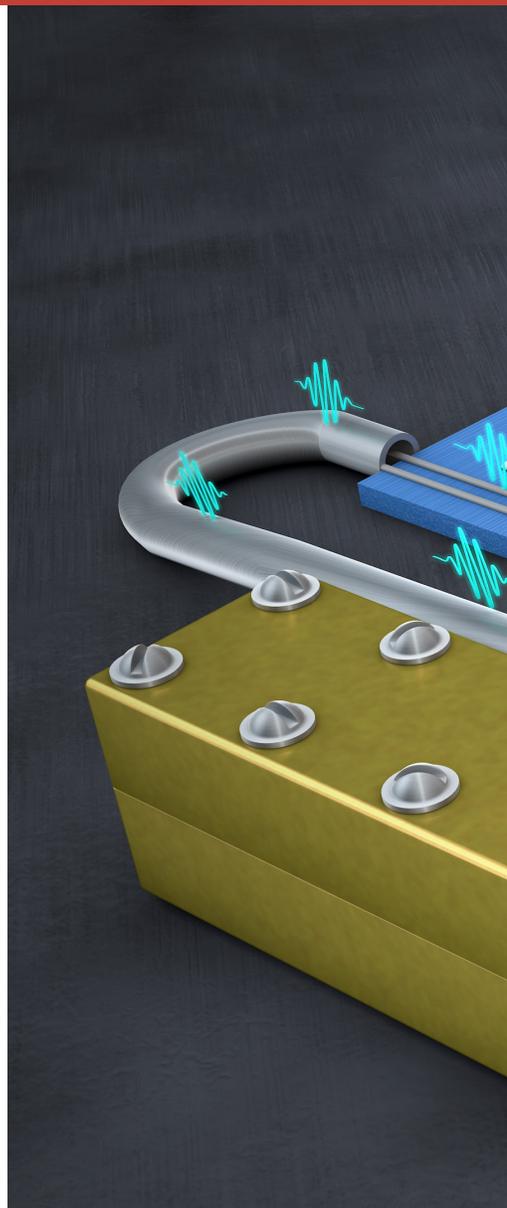
1. In the quantum world, any two (or more) quantum states can be added together (“superposed”), and the result will be a third quantum state.

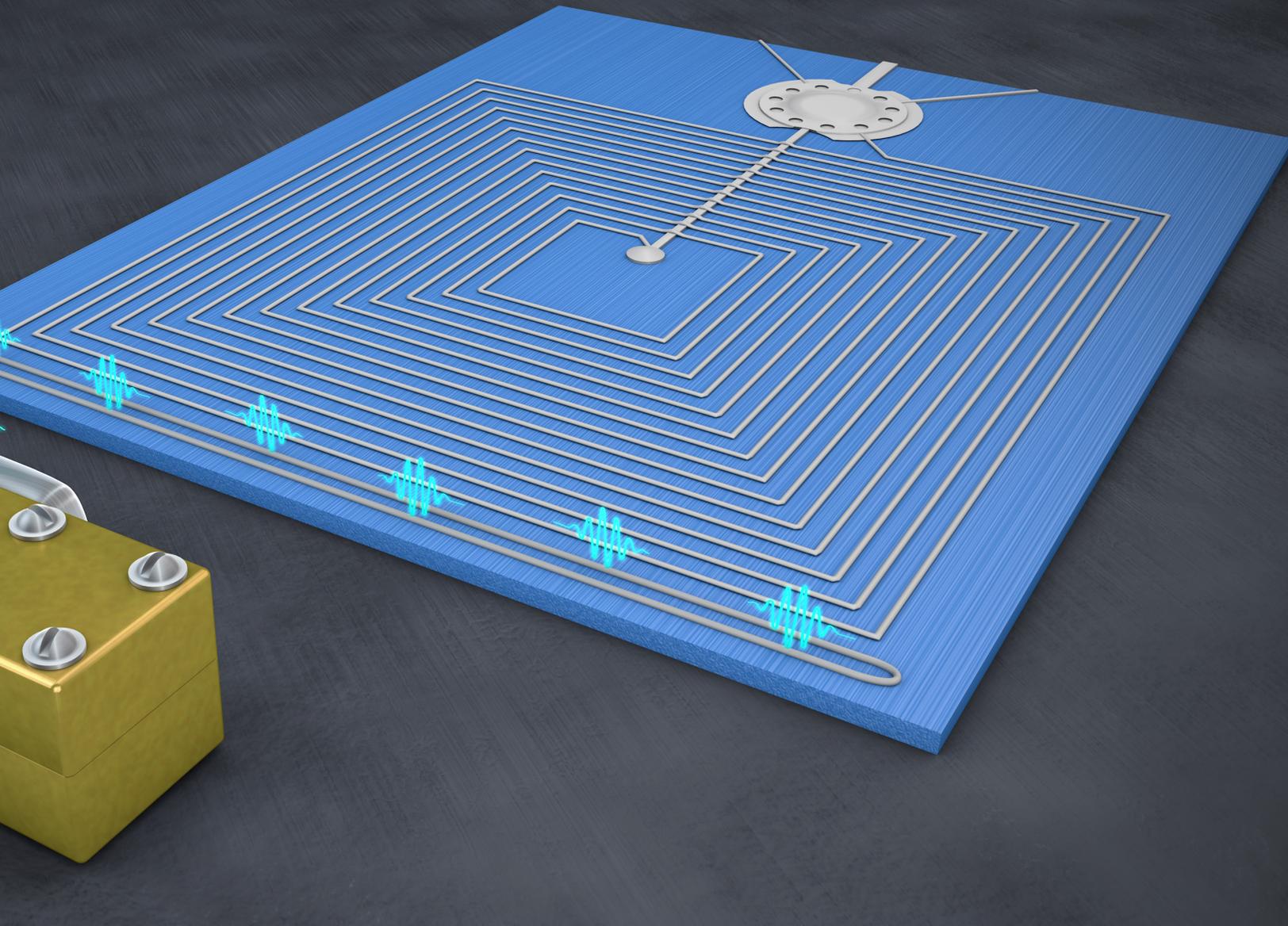
optical signals transmitted over fibers between nodes storing and processing information with microwave (electrical) signals. And, they will require perfect and flexible conversion between optical qubits and microwave (electrical) qubits. Reed and his colleagues’ work is helping to build the technology for these future quantum networks.

“It’s not so far out to believe you can take a quantum state generated by a superconducting qubit and convert it to motion, then build a system that completes the transfer to light,” Reed said.

Reed and his colleagues’ result is the very first step in showing that it’s possible to take a quantum signal, encode it in an electronic pulse, then convert that pulse to mechanical motion. The final step will be converting the mechanical motion to optical photons. Every step in this process has been sufficiently challenging to require years to complete. That’s why the new result is so exciting

“For the first time, we were able to take our electromechanical devices and use them to process



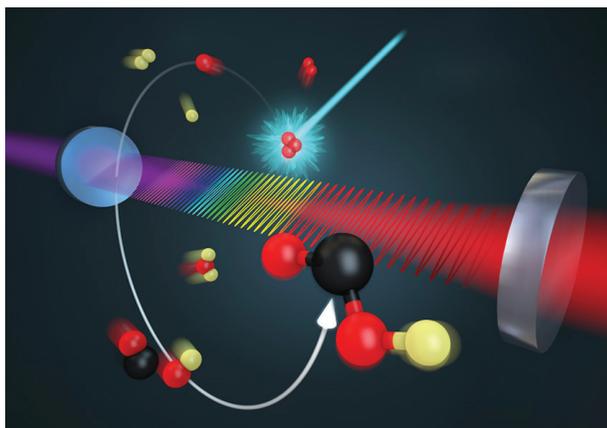


The Lehnert group recently demonstrated that this electromechanical device, which consists of a microwave circuit and a tiny aluminum drum (center), can process qubits. Credit: The Lehnert group and Steve Burrows, JILA

qubits, and we do that all in the microwave domain,” Reed said. “This accomplishment may lead to offshoots in many directions for building quantum networks that could potentially interface with light.”\*

A. P. Reed, K. H. Mayer, J. D. Teufel, L. D. Burkhardt, W. Pfaff, M. Reagor, L. Sletten, X. Ma, R.J. Schoelkopf, E. Knill, and K. W. Lehnert, *Nature Physics* **13**, 1163–1167 (2017).

## MOLECULAR SPECTROSCOPY AT JILA



Artist's conception of an infrared frequency comb "watching" the reaction of a molecule of carbon monoxide (CO, red and black) and hydroxyl radical (OH, red and yellow) as they form the elusive reaction intermediate DOCO (red/black/red/yellow) before DOCO falls apart. This chemical reaction was seen for the first time in 2016 under normal atmospheric conditions in Jun Ye's lab. Credit: The Ye group and Steve Burrows, JILA

Since its inception, JILA researchers have achieved some remarkable results with molecular spectroscopy, which is the study of the interaction of molecules with wavelengths of light ranging from the infrared through the x-ray regions of the electromagnetic spectrum. JILA scientists have measured electron affinities in atoms and molecules, probed the internal workings of living cells, elucidated chemical reactions in the atmosphere and combustion, investigated the quantum mechanics occurring inside molecules, and watched chemical reactions unfold in real time.

For instance, in 2016 the Jun Ye group used molecular spectroscopy to identify an analog of a chemical reaction intermediate, trans-HOCO, which had never before been observed despite being predicted for more than a century. This intermediate is critical to a chemical reaction that adds carbon dioxide to the atmosphere, one reason why it has attracted so much recent interest from chemists. The reaction of OH with CO (carbon monoxide) to

produce H (hydrogen) and CO<sub>2</sub> (carbon dioxide) also plays a key role in combustion and chemistry in the interstellar medium.

In 2017, the Ye group watched every step of the same reaction and discovered that an isomer of the molecule they had thought was the reaction intermediate, called cis-HOCO, was actually responsible for the formation of the reaction products. The two isomers, called cis-HOCO and trans-HOCO, vary only in their angle of rotation around a single C-O bond.

This amazing feat of chemical detection was due to a collaboration of physicists in the Ye group with postdoc Thinh Bui, who holds a doctorate in chemistry from CalTech. In 2016, Bui and his team had looked for, and identified, a heavy-hydrogen (D) version of what they thought was the reaction intermediate known as trans-DOCO. OD was used in place of OH in the experiment because it was easier to "see" with the group's frequency-comb laser. However, like HOCO, DOCO exists as two different isomers.

Bui and his colleagues observed the reaction OD + CO → D + CO<sub>2</sub> initially form approximately equal amounts of cis-DOCO and trans-DOCO. The two isomers then flip-flopped back and forth between the cis and trans isomers. But, there was an unexpected twist to what the researchers observed.

The cis-DOCO isomer was able to quantum mechanically tunnel through an energy barrier to form the reaction products D + CO<sub>2</sub>. After a while, all the cis-DOCO formed the D + CO<sub>2</sub> products, leaving just enough of the longer-lived trans-DOCO to have been erroneously identified as the reaction intermediate in the earlier experiment. With the latest work, Bui and his colleagues clarified that trans-DOCO itself does not actually directly form

reaction products; only cis-DOCO is able to do that. This stellar feat of molecular spectroscopy appeared in *Science Advances* in December 2017.

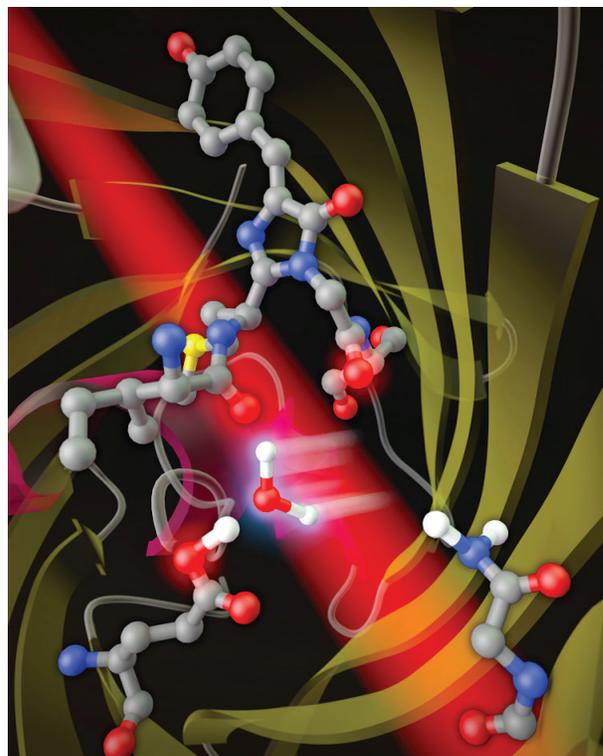
Molecular spectroscopy is also a critical method in the Ralph Jimenez lab. This group performs molecular spectroscopy with visible or infrared light rather than with a frequency-comb laser. Optical molecular spectroscopy makes it possible for the biophysicists in this group to investigate how light-emitting structures work inside red fluorescent proteins. The Jimenez group's work focuses on the light-emitting structure, called a chromophore, embedded in the fluorescent protein matrix. The group's goal is to understand how a protein absorbing laser light determines the color and efficiency of the light emitted from such a large molecule.

Jimenez uses time-resolved optical spectroscopy because large molecules like proteins don't have nice, high-resolution line signatures of quantum states.

Despite this drawback, Jimenez likes working with proteins because it's possible to genetically engineer them to have precisely the structure you want to investigate.

"The laser spectroscopy of complex proteins rests on a foundation of a whole history of laser spectroscopy of simpler systems over all the years of work here and elsewhere," Jimenez said. "My work employs a combination of changing the structure and probing the electronic states, both at the same time. As a chemist, you like to do both in an experiment on protein dynamics."

In addition to his work on protein dynamics, Jimenez participated in a novel experiment in 2017 to advance the science of x-ray molecular spectroscopy. Jimenez worked with a team from the National Institute of Standards and Technology (NIST) on using a new x-ray spectrometer to "watch" high-power x-rays break molecular bonds. NIST's new spectrometer, which fits on two lab benches and costs just a tiny fraction of an entire

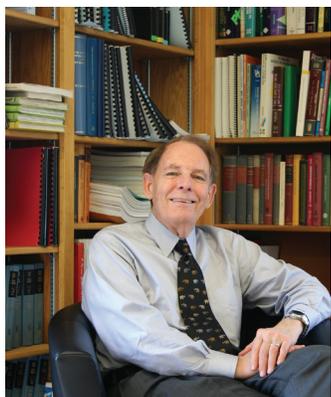


Light-emitting molecular arrangement in the chromophore of an mPlum fluorescent protein. The incorporation of a water molecule into this structure causes the emission of red light. Credit: The Jimenez group and Steve Burrows, JILA

synchrotron system, proved to be faster and more efficient than the much larger and more expensive synchrotron systems.

Using the new spectrometer, Jimenez and the NIST team were able to watch how electrons and bond lengths changed in response to a laser breaking a molecular bond in CO-heme (carbon-monoxyl-iron protoporphyrin IX). The spectrometer was able to make x-ray movies of the entire process of molecular bond breaking, in the process demonstrating that the new spectrometer is competitive with huge multibillion-dollar synchrotrons for producing x-rays to probe molecules.

The Kapteyn-Murnane group also investigates molecules with extreme ultraviolet (EUV) or soft x-ray light. Initially, the group was interested in capturing the sequence of events that occur as a molecule reacts to being irradiated with



W. Carl Lineberger in his office.  
Credit: Brad Baxley, JILA

x-rays. For example, after exciting molecules of nitrogen ( $N_2$ ) with ultrafast x-rays, the group used a laser pulse to peer inside the excited molecules and watch the molecule rapidly explode. Then, they tried the same experiment with an oxygen molecule ( $O_2$ ), which took much longer to just fall apart.

Next, the group developed its own tabletop method of high-harmonic generation (HHG) to create EUV and soft x-ray light for probing molecules. HHG uses a high-intensity laser pulse to pluck high-speed electrons out of vibrating atoms in a molecule. The atoms then crash back into the original molecule, causing the emission of EUV or soft x-ray light. Over the years, the group has learned to carefully select the wavelengths of light produced with this method, making HHG an extremely useful tool for studying the physics of molecular behavior.

Gaining a molecular-level understanding of how chemistry works is also a goal of the Mathias Weber group. The group is currently using molecular spectroscopy to explore how light excites vibrations in clusters and complexes.

"If clusters or complexes absorb an IR photon, that means the IR photons we've irradiated them with have caused a vibrational transition in our ionic clusters or complexes," Weber explained. "We detect the fact that they have absorbed light by watching them fall apart. They are not very strongly held together with van der Waals interactions rather than molecular bonds." The hot clusters evaporate solvent molecules to cool down, which allows the researchers to detect that the cluster has absorbed a photon.

The Weber group performs a similar experiment with charged molecules or clusters by irradiating them with visible or ultraviolet light, which makes the molecules or clusters fall apart. An examination of the decomposition products reveals information about the electronic levels of the molecules. The group also does photoelectron spectroscopy of molecules, a technique pioneered at JILA by Carl Lineberger.

"JILA has always been strong in molecular spectroscopy," explained Weber. "The complexity of the molecules we can look at and learn something about is growing, and the precision with which we can learn something about molecules is also growing.

"Now we want to know what is the quantum structure of a molecule. That's the underlying question you ask with molecular spectroscopy."

The recent achievements in molecular spectroscopy at JILA have been built upon a solid foundation in molecular spectroscopy laid early in the Institute's history. JILA's first molecular spectroscopist was Dick Zare, who began using lasers at JILA in 1966 to look at molecules. Zare published what is considered to be the first-ever report of molecular spectroscopy in the *Bulletin of the American Physical Society* in 1967.

Like other JILA scientists who subsequently participated in the development of molecular spectroscopy, Zare profited from Jan Hall's invaluable work on developing stable lasers. Although Zare's tenure at JILA was relatively brief, he left an indelible mark on JILA by developing the technique known as laser-induced fluorescence, or LIF, in collaboration with Visiting Fellow Herb Broida, a molecular physicist working at NIST Gaithersburg.

"Dick brought the first real chemistry to JILA, and by chemistry I mean molecular spectroscopy," said Carl Lineberger, who arrived at JILA as a postdoc after a three-year stint in the Army, just as Zare was

leaving in 1968. “These were chemistry-motivated types of experiments, but very much at the physics end of things.”

Fortunately for Lineberger, former Visiting Fellow Broida (who loved Colorado) came to work in the Lineberger lab for several summers, helping Lineberger establish a research focus on molecules. In the early years, it was lonely being a chemist in JILA, which had laboratory astrophysics as its main research focus.

Nevertheless, JILA’s small, but determined, chemistry group proved to be exceedingly important to JILA—and to Jan Hall’s development of the critical laser technologies that played a key role in JILA’s ongoing success in molecular spectroscopy.

In the late 1960s, for example, the best lasers around were helium lasers in the infrared, but there was no atom that could improve the function of those lasers. So, Hall asked Lineberger if there were a molecule he could put inside his helium laser that would work. Lineberger suggested methane, which Hall tried, and the new laser instantly worked much better. Because of Lineberger’s suggestion, the stability of Hall’s lasers increased on the order of a million-fold, opening the door to the development of molecular spectroscopy on a serious level.

“My suggestion was the first in a sequence of things that led to the kinds of things that are going on now,” Lineberger said. He credits Hall’s invention of the methane-stabilized laser with starting his own career as a molecular spectroscopist.

“Jan and others were extraordinarily helpful with technologies because they were the tool builders who had no idea what to do with them,” Lineberger said. “So when someone would come to them and

take one of their toys and do something interesting and unique, they were more than happy to see that happen.”

The other key ingredient in setting the stage for molecular spectroscopy was the arrival of Terry Brown and other electronics wizards at JILA.

“All those things led to the beginning of molecular science in JILA,” Lineberger said, noting that the combination of stable lasers and excellent electronics allowed him to establish a major research focus using photoelectron spectroscopy, or PES, to probe molecules.

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“I did photoelectron spectroscopy in many forms,” Lineberger said. “Everything eventually came back to lasers and negative ions. Ions were a sensible thing to be using because you can detect them so much better than neutrals.”

Using photoelectron spectroscopy, the Lineberger lab was able to measure the vibrations and electronic-state excitations of molecules—measurements that had been unattainable in other ways. The method also made it possible to determine the differences in structure between negative ions and their associated neutral molecules, allowing for the determination of fundamental properties associated with adding an extra electron to a specific molecule.

The Lineberger group used PES to investigate systems that rearrange very quickly upon photoelectron detachment, small metal cluster properties, singlet-triplet splittings in diradicals, and transitions from stable anion geometries to transition states in the neutral.

“Eventually, I looked at one way in which energy is shared among 20-30 or more atoms in a

clump when you excited a particular place in it," Lineberger explained. "We asked the question: How does the behavior change as you build up a clump of atoms, one at a time? What does it take to make a gold atom look like a gold brick? After doing a fair amount of this work, you begin to see rules that are similar to the rules that govern the periodic table."

Lineberger's PES lab had a long and successful history in JILA until the lab finally closed in 2017. Nearly all the entries in the electron affinity tables in the *CRC Handbook of Chemistry* consist of measurements in the Lineberger PES lab, including most of the atoms and more than a hundred molecules.

Just a few years after Lineberger's work with photoelectron spectroscopy started, a young chemist from Harvard University, Bill Reinhardt, was recruited as a JILA Visiting Fellow for two years. At the end of this period (1973-1974), Reinhardt was brought in as an associate professor of chemistry at the University of Colorado Boulder.

"Bill came, and we really began to build up a presence in chemistry," Lineberger recalled. "No one here had much idea of what chemistry or molecular spectroscopy were, so Bill and I started up a Friday afternoon seminar program called Up to Speed with Molecules, and that's how we managed to get things moving."

Four years later, Reinhardt hired Steve Leone, who came from the University of Southern California in 1978.

"When I came to Boulder, the new thing I brought was the ability to use very sensitive infrared detectors to observe vibrational emission in the infrared and to use those observations to study the chemical dynamics of product state distribution, molecular state transfer, photofragmentation dynamics, and so on," Leone said.

Soon after arriving, Leone began working with Barney Ellison, Veronica Bierbaum and Chuck DePew in the University of Colorado Boulder Department of Chemistry. Thanks to another new machine built by the JILA Instrument Shop to study ion emission and product states of ion reactions, Leone and his colleagues from the chemistry department opened up a whole new research field.

In the late 1980s, Leone began using Fourier Transfer Infrared (FTIR) Spectroscopy in emission to look at products of different reactions, energy transfer, or fragmentation in chemical reactions. Once again, the JILA shops helped out by crafting big reaction chambers that would collect very weak IR emission and funnel it into a Fourier transform spectrometer. Leone and his team modified the spectrometer to operate in emission, rather than the traditional absorption mode. FTIR molecular spectroscopy turned out to be a huge field that produced volumes of very detailed results.

Another Leone lab project was bouncing molecules off surfaces and looking at state-changing collisions, or reactivity. This work would eventually become a major research area for Fellow David Nesbitt, who was Leone's first graduate student. Nesbitt was recently recognized during JILA's 500<sup>th</sup> Thesis Celebration on October 19, 2017, for writing JILA's 100<sup>th</sup> thesis in 1982.

Fortuitously, in 1983 Nesbitt spent a year as an NRC postdoc working with Jan Hall, who gave him one of the very first color-center lasers ever developed.

"I had a great deal to learn about lasers," Nesbitt recalled. "Jan taught me not just about servo loops and lasers, but also about electronics. Electronics was a game changer for me, and Jan had a fondness for me because I understood molecules."

After a year working with Hall, Nesbitt went off to Berkeley, where he learned to do first-class spectroscopy on highly chemically reactive radicals such as methylene. Nesbitt's work was the first

high-resolution study of a highly reactive molecule, and when he wasn't working in the lab, he educated his collaborators on the importance of quality electronics for molecular spectroscopy.

With Nesbitt's addition to the JILA faculty in 1984, JILA acquired a premier chemistry group with a strong grounding in molecular spectroscopy. Lineberger, Reinhardt, and Nesbitt were JILA's core group of molecular spectroscopists for many years.

Nesbitt studied molecules touching each other by examining the spectra of molecular complexes. To do this, he developed supersonic expansions of molecules through a slit. The molecules were so cold they started to stick to each other. Nesbitt passed multiple pathways of super high-resolution infrared light back and forth through the supersonic expansion. In the process, he launched an entirely new subfield of infrared molecular spectroscopy with studies of cold supersonic expansions of dozens and dozens of molecules—work that continues today.

"These molecules form clusters, and then they drop through my laser beam," Nesbitt explained. "Then I see photons being absorbed by the molecules, and I look at teeny little losses of IR photons from my laser beam, and I could see very small absorbances." This new technique yielded unprecedented sensitivity.

"Within about nine months, we had our first spectrum of hydrogen fluoride (HF) with an atom of argon sticking on it like a balloon would stick on your shirt. Then we would excite the high-frequency HF stretches, and it was like a buzz saw going on in these complexes. But the buzz saw was moving too quickly to get energy into this slow, lazy argon atom that wanted to vibrate maybe 20, 30, or 40 times slower."

It was so hard to get the energy to leave the molecules that they lasted 300–400 microseconds. The spectroscopy was absolutely definitive, allowing

the Nesbitt group to investigate van der Waals complexes before graduating to hydrogen-bonded complexes and stronger interactions. As a result, Nesbitt was instrumental in developing this entire field of molecular spectroscopy.



David Nesbitt. Credit: Greg Kuebler, JILA

Nesbitt's use of slit-jet infrared molecular spectroscopy taught him a great deal about the quantum mechanics governing molecules. He learned about tunneling, a process in which molecules can go where they are not allowed classically to go. Nesbitt's group was able to observe tunneling because, in addition to observing the molecular tumbling that naturally occurs in spectroscopy in the infrared, it was possible to watch molecules and atoms tunnel around.

"This technique was absolutely a gem of discovery," Nesbitt said. "The spectra do not lie. Spectroscopy is basically Nature's way of solving quantum mechanics, and Nature just does it." Nesbitt noted that it often took his group weeks and months to understand what Nature was saying in their experiments.

At the same time, Nesbitt was also using molecular spectroscopy to investigate how molecules relax. His group first asked the question: How long does it take a vibration to wax into a molecule if you set off a vibration in a single chemical bond? It turned out that the real question was: What quantum states can talk to your initial excited vibration? Nesbitt and his group looked for answers in their spectra and discovered they had a hundred-thousand-fold more sensitivity for observing what turned out to be a dense

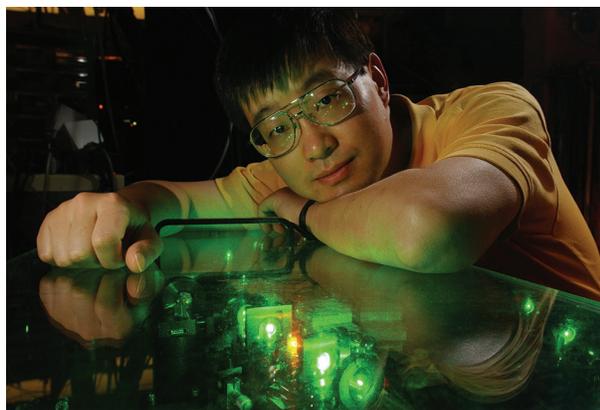
collection of quantum states. In studies of acetylene, for example, the group was able to answer the question: Does the triple bond carbon act like a brick wall to vibration? Or does the vibration pass through the bond? The answer counter-intuitively is that the carbon-carbon triple bond does a little of both.

Eventually the Nesbitt group used its slit-jet experiment to switch to Intramolecular Vibrational Redistribution, or IVR, spectroscopy. In so doing, the group implemented the idea of striking a discharge just before the molecules under study expanded and cooled.

"It turns out supersonic expansions are gorgeous media for striking discharges," Nesbitt explained. "We make our molecules go through hell in a microsecond." However, a microsecond is long enough for electrons to crack off bonds, forming radicals. But the radicals are frozen in a 25 K gas, moving parallel and not colliding, while being probed by a laser. In the past 10 years, the group has done a huge amount of spectroscopy with a variety of radicals as well as looking for cis- and trans-HOCO, the reaction intermediates probed by molecular spectroscopy in the Ye group.

"Jun's work is the first IR detection of the cis isomer of this radical," Nesbitt said, who speculated that cis-HOCO is hard to see because it immediately falls apart to  $H^+$  and  $CO_2$  when it is vibrationally excited. Another problem for Nesbitt is that supersonic methods frustrate the transformation from the trans to the cis isomers. Still, he is determined to get to the bottom of this reaction because it is critical in models of combustion and important for understanding atmospheric chemistry.

Today, molecular spectroscopy is no longer the exclusive purview of JILA chemists Nesbitt, Lineberger, and Weber. Biophysicist Ralph Jimenez uses molecular spectroscopy to study really, really large protein molecules: fluorescent proteins. Physicists Margaret Murnane and Henry



Jun Ye in the lab in 2007. Credit: Greg Kuebler, JILA

Kapteyn use molecular spectroscopy to better understand the physics of molecular behavior. And, Jun Ye also uses molecular spectroscopy to study the fundamental physical properties of cold and ultracold molecules.

"I got interested in cold molecules because I wanted to see if we could extend control to more complex molecules," Ye explained. "At first, we were more on the physics of 'toy' models, but we thought eventually our work might be relevant to real chemical reactions."

Molecular spectroscopy with a frequency-comb laser allowed the Ye group to observe the entire chemical reaction of  $OD + CO \rightarrow D + CO_2$ . The group is continuing its exploration of chemical reactions with ultracold K<sup>85</sup>Rb molecules. The group's goal is to control the states of the ultracold molecules and watch chemistry in real time as a cold chemical reaction goes from reactants to products.

"This is where the integration of physicists and chemists is coming together, Ye said. "This merging of physics and chemistry is very powerful" It should be. This kind of research has been enabled by state-of-the-art molecular spectroscopy techniques under development at JILA for more than five decades. ✨

# IN THE NEWS

## IN THE NEWS?

### YE ELECTED TO CHINESE ACADEMY OF SCIENCES

The Chinese Academy of Sciences announced on November 29, 2017 the election of JILA Fellow Jun Ye as a Foreign Member, China's highest honor for foreign scientists:

*The Chinese Academy of Sciences (CAS) promotes scientific and technological advances across the world. CAS includes a network of more than 100 research and development organizations across the world; three universities; and a traditional merit-based academy analogous to the US National Academy of Sciences to recognize and convene scientific leaders from across the world.*

Membership in the CAS is comparable to election to the US National Academy of Sciences, representing a significant honor for international achievement and impact in science. Members are selected to one or more Divisions, including Physics and Mathematics (the Division to which Ye was elected), Chemistry, Life Sciences and Medical Sciences, Earth Sciences, Information Technology Sciences, and Technological Sciences.

The CAS currently has 800 Chinese members and 92 foreign members, including the newest members. Foreign Members are elected from across the world based on their scientific achievements. A few examples of current foreign members of the CAS in addition to Ye include:

- Richard Zare, Stanford, father of ultrafast laser chemistry and former JILA Fellow.
- Klaus von Klitzing, Germany, Nobel Physics Laureate for integer quantum Hall effect.
- Steve Chu, Stanford and Bell Labs, Nobel Physics Laureate for laser cooling and former US Energy Secretary and former JILA Visiting Fellow.

## A selection of news, awards, and what is happening around JILA

- Charles Kao, multiple universities and industries in US, UK and Hong Kong, Nobel Physics Laureate for pioneering optical fibers.

Ye is also one of JILA's several members of the US National Academy of Sciences (NAS). Similar to the CAS, the US NAS has about 2,000 U.S. Members and about 400 Foreign Members.

Ye's election to the CAS recognizes his world leadership in ultracold atoms and molecules, in optical lattice atomic clocks, in ultrastable lasers and femtosecond laser frequency combs, and in precision measurements.

### YE WINS NIST JACOB RABINOW APPLIED RESEARCH AWARD

JILA and NIST Fellow Jun Ye was named the 2017 winner of the NIST Jacob Rabinow Applied Research Award, one of the top honors for the more than 1,600 scientific and technical NIST employees.

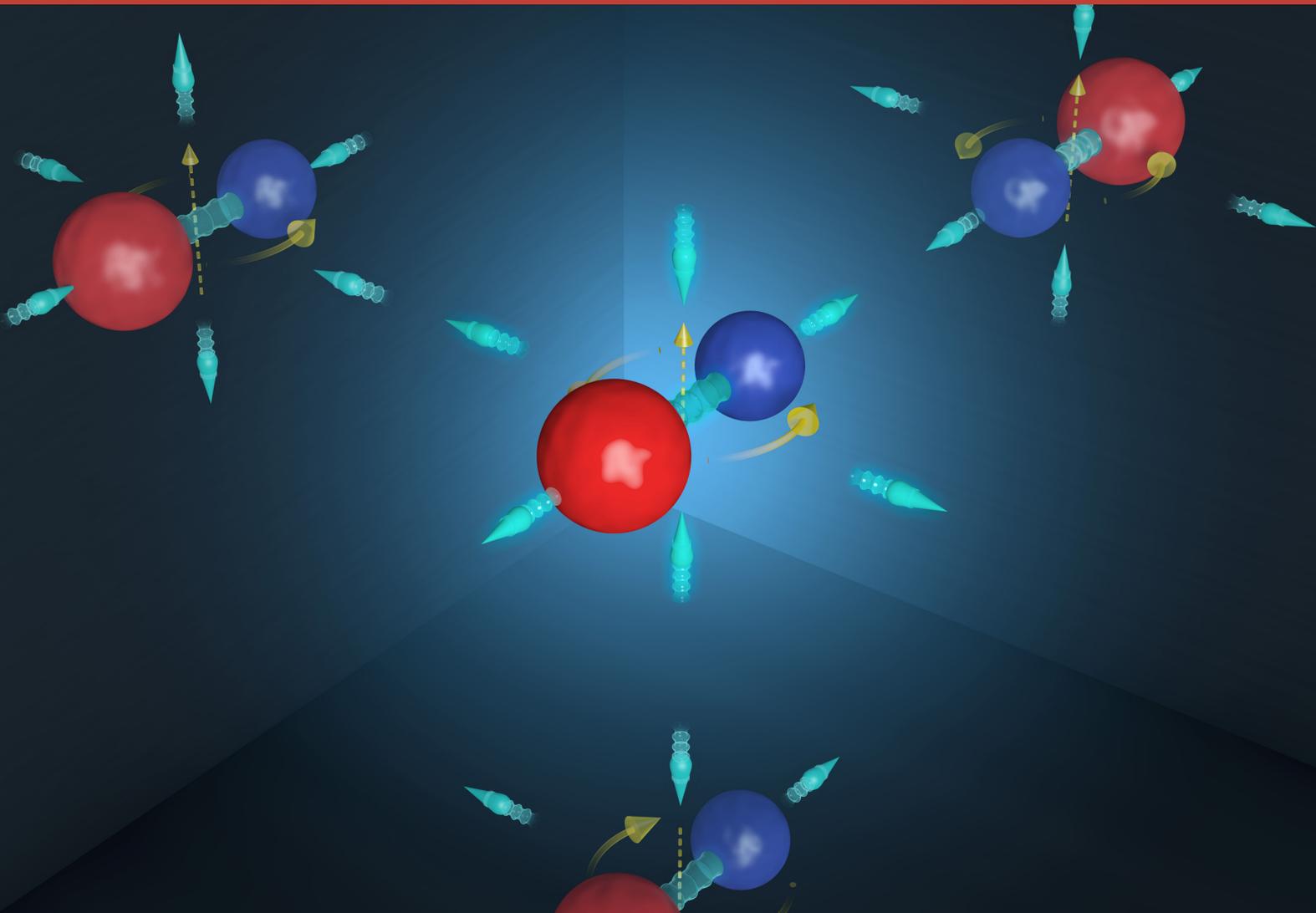
Jacob Rabinow was a prolific innovator working at the National Bureau of Standards (NBS), the predecessor of NIST, receiving more than 200 patents for his inventions of mechanical, electrical, and optical devices and systems.

The NIST Rabinow Award for applied science emphasizes the breadth, depth, and impact of Ye's work. Ye's leading-edge scientific research has resulted in innovations including ultrastable lasers, world-leading atomic clocks, and novel frequency comb applications. Such innovations and are used for a wide range of applications, from medical diagnostics to new ultraprecision chemical measurements.

Ye has received many international and NIST awards for his pioneering fundamental research. Ye is one of the very few people to have won both NIST's highest award for fundamental research (Samuel Wesley Stratton







Is the electron completely round, or is it ever so slightly egg-shaped because it has electric dipole moment? The Cornell group is looking for the answer to this fundamental question with a sophisticated trapped-ion apparatus that includes a rotating electric field. Credit: The Cornell group and Steve Burrows, JILA

## And, The Answer Is... Still Round

Why are we here? This is an age-old philosophical question. However, physicists like Will Cairncross, Dan Gresh, and their advisors Eric Cornell and Jun Ye actually want to figure out why people like us exist at all. If there had been the same amount of matter and antimatter created in the Big Bang, the future of stars, galaxies, our Solar System, and life would have disappeared in a flash of light as matter and antimatter recombined. But we know that's not what happened. After matter-antimatter recombination, sufficient matter remained to form galaxies, stars, planets, and physicists who wonder why on Earth things turned out the way they did.

The JILA team thinks precision measurement of the shape of the electron may help them figure out the answer. They think that in the beginning of the Universe, there must have been a slight excess of

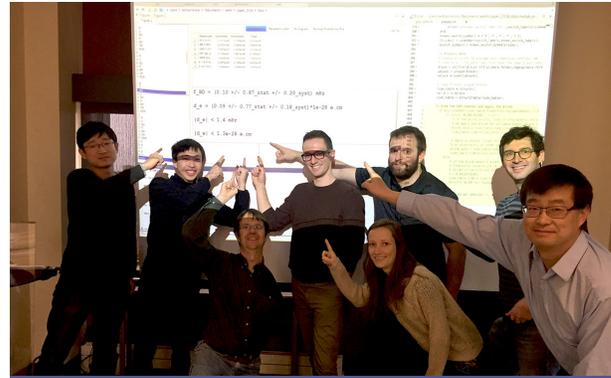
matter vs antimatter caused by tiny asymmetries in fundamental particles such as the electron. If, for example, the electron is ever-so-slightly egg-shaped (rather than round), then it may help explain why the scientists doing the experiment—and the rest of the material Universe—exist.

The challenge faced by the Cornell and Ye group is that an asymmetry in the electron’s shape—called an electron electric dipole moment, or eEDM—would be vanishingly small. If an atomic nucleus were the size of our Solar System, then the eEDM (if there were one) would measure only a few millimeters. The actual eEDM (if there is one) might end up being measured in nonillionths or even decillionths of a centimeter ( $10^{-30}$ – $10^{-33}$  cm). These are breathtakingly short distances that require sophisticated physics experiments to precisely measure.

Determining the length of something this tiny not only requires creative experimental design, but also a profound understanding and application of precision measurement techniques. Fortunately, precision measurement of ultrasmall things is a specialty of the Cornell and Ye groups. The team recently used its sophisticated trapped-ion, rotating electric-field apparatus in its latest series of precision eEDM measurements.

The Cornell-Ye collaboration determined an upper limit for the eEDM of  $1.3 \times 10^{-28}$  e cm, which is consistent with the upper limit of  $9.3 \times 10^{-29}$  e cm found in a different experiment by the ACME collaboration between Harvard and Yale in 2014. In other words, even at such tiny distances as  $10^{-28}$ – $10^{-29}$  cm, the electron is *still round*.

One very cool thing about this experiment is that the JILA team is positive its measurements were free of any bias on the part of the experimenters. The researchers did a year’s worth of experiments in different experimental configurations, all the while measuring the eEDM blind. Instead of having their computer tell them the results of individual measurements, they programmed the computer



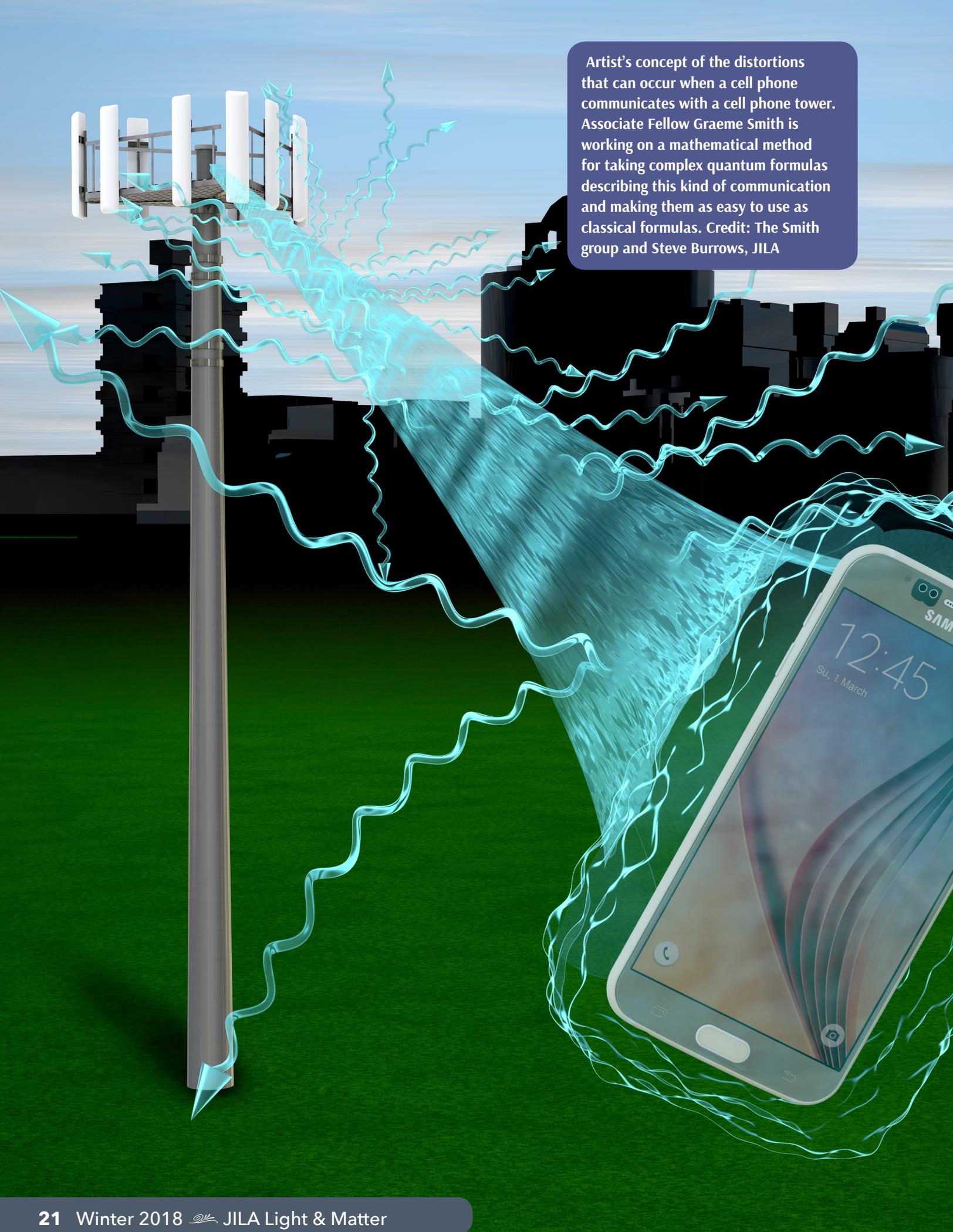
After more than a year of recording “blind” precision measurements of the electron electric dipole moment, a computer revealed a value of the eEDM consistent with a round-shaped electron. The Cornell and Ye group researchers in the photo are seeing their results for the first time. They are (l–r) Yan Zhou, Kia-Boon Ng, Eric Cornell, Will Cairncross, Tanya Roussy, Dan Gresh, Yuval Shagam, and Jun Ye. Credit: Julia Bachinski, JILA

to add a random number to the results, so what the researchers saw after each measurement was a meaningless number. They did their experiments like this to keep their personal expectations, or bias, out of the results.

Then after a year, they asked the computer for the answer. The answer was consistent with zero, meaning that at the level they could resolve, the electron was still round. However, Cairncross, Gresh, and their team aren’t about to give up. They’re building a new experiment expected to resolve length measurements of  $10^{-30}$  cm, tantalizingly close to where the researchers expect to see evidence of an eEDM, if it exists.

The researchers responsible for this multiyear project to seek out and measure the eEDM include graduate students Will Cairncross, Dan Gresh, and Tanya Roussy, research associate Yan Zhou, former research associate Matt Grau, JILA Ph.D. and former research associate Kevin Cossel as well as Fellows Jun Ye and Eric Cornell.✿

William B. Cairncross, Daniel N. Gresh, Matt Grau, Kevin C. Cossel, Tanya S. Roussy, Yiqi Ni, Yan Zhou, Jun Ye, and Eric A. Cornell, *Physical Review Letters* **119**, 153001 (2017).



Artist's concept of the distortions that can occur when a cell phone communicates with a cell phone tower. Associate Fellow Graeme Smith is working on a mathematical method for taking complex quantum formulas describing this kind of communication and making them as easy to use as classical formulas. Credit: The Smith group and Steve Burrows, JILA

# E.T. Phone Home

**W**hen Steven Spielberg’s adorable extra-terrestrial, E.T., wanted to phone home, he should have contacted an information theorist like JILA’s Graeme Smith. Smith could have at least explained how E.T. could have used a cell phone to send a low-noise message to a cell phone tower, and from there . . .



“Think of a cell phone,” Smith said. “You have some message you’re trying to send to a cell tower, and the electromagnetic signals that come out of your phone are going to get distorted as they travel through the air to the tower<sup>1</sup>. The signals get bounced around or even lost.” That’s why all cell phones and towers must use error-correcting codes.

However, being an information theorist, Smith is looking at quantifying how much error-free data it’s possible to send from a cell phone to the tower.

“When my cell phone tries to send a message to the cell tower, the electromagnetic signal gets distorted along the way,” Smith said. “You can actually write down a nice clean formula for how much error-free data you can send.”

The formula for classical communication channels (if there’s no quantum mechanics involved) was invented in the 1940s. The problem is that the nice, clean formula doesn’t work when quantum mechanics comes into play. Entanglement makes the situation a lot more complicated. In fact, the nice formula becomes insoluble.

1. Cell phones were invented in 1973.

So Smith is working on something called uniform additivity. Uniform additivity is a mathematical way of taking complex quantum formulas and trying to make them as easy to use as a classical formula. It’s a fascinating process.

One of Smith’s favorite questions is, “How do I send quantum bits from here to there, if I know there is noise in between?”

He starts with a list of the basic answers that may already exist to important questions in information theory. These are the most useful solutions in the quantum domain. And, if an answer isn’t on the list, it’s unlikely anyone can solve that problem. In other words, information theorists have some answers. Now they need to find the correct questions.

One of Smith’s favorite questions is, “How do I send quantum bits from here to there, if I know there is noise in between?” And, by better understanding the simple properties of noise, he hopes to learn how to transmit quantum states noiselessly in the presence of some noise. In recent work, he discovered an intriguing circumstance in which the classical and quantum descriptions of this process were identical.\*

Andrew Cross, Ke Li, and Graeme Smith, *Physical Review Letters* **118**, 040501 (2017).

## THE CLOCK THAT CHANGED THE WORLD New quantum gas clock has record-shattering precision

Imagine A Future . . . The International Moon Station team is busy on the Moon's surface using sensitive detectors of gravity and magnetic and electric fields looking for underground water-rich materials, iron-containing ores, and other raw materials required for building a year-round Moon station. The station's mission: launching colonists and supplies to Mars for colonization.



Meanwhile, back on Earth, Americans are under simultaneous assault by three Category 5 hurricanes, one in the Gulf of Mexico and two others threatening the Caribbean islands. Hundreds of people are stranded in the rising waters, but thanks to precision cell-phone location services and robust cell-tower connections in high wind, their rescuers are able to accurately pinpoint their locations and send help immediately.

Other new technologies in this revolutionary new era of quantum devices include precision instruments searching for dark matter and dark energy, ultraprecise electronics, unbreakably secure military communications, and ordinary laboratory-sized gravitational-wave detectors and x-ray lasers. There are also advanced scientific experiments making use of the most accurate absolute measurements in history—accurate to  $10^{-23}$ . These technologies were unheard of in 2017, when the world's first quantum-gas optical-lattice clock demonstrated its capabilities by measuring a  $3.5 \times 10^{-19}$  fractional frequency shift in 2.2 hours of averaging time.

The strontium-87 ( $^{87}\text{Sr}$ ) lattice optical atomic clock was the first in history to cool its atoms to quantum degeneracy. Quantum degeneracy occurs when all the atoms in the clock occupy their lowest-possible energy level<sup>1</sup>. Lowering the temperature of the clock's 10,000  $^{87}\text{Sr}$  atoms to quantum degeneracy allowed Sara Campbell's team to capitalize on the

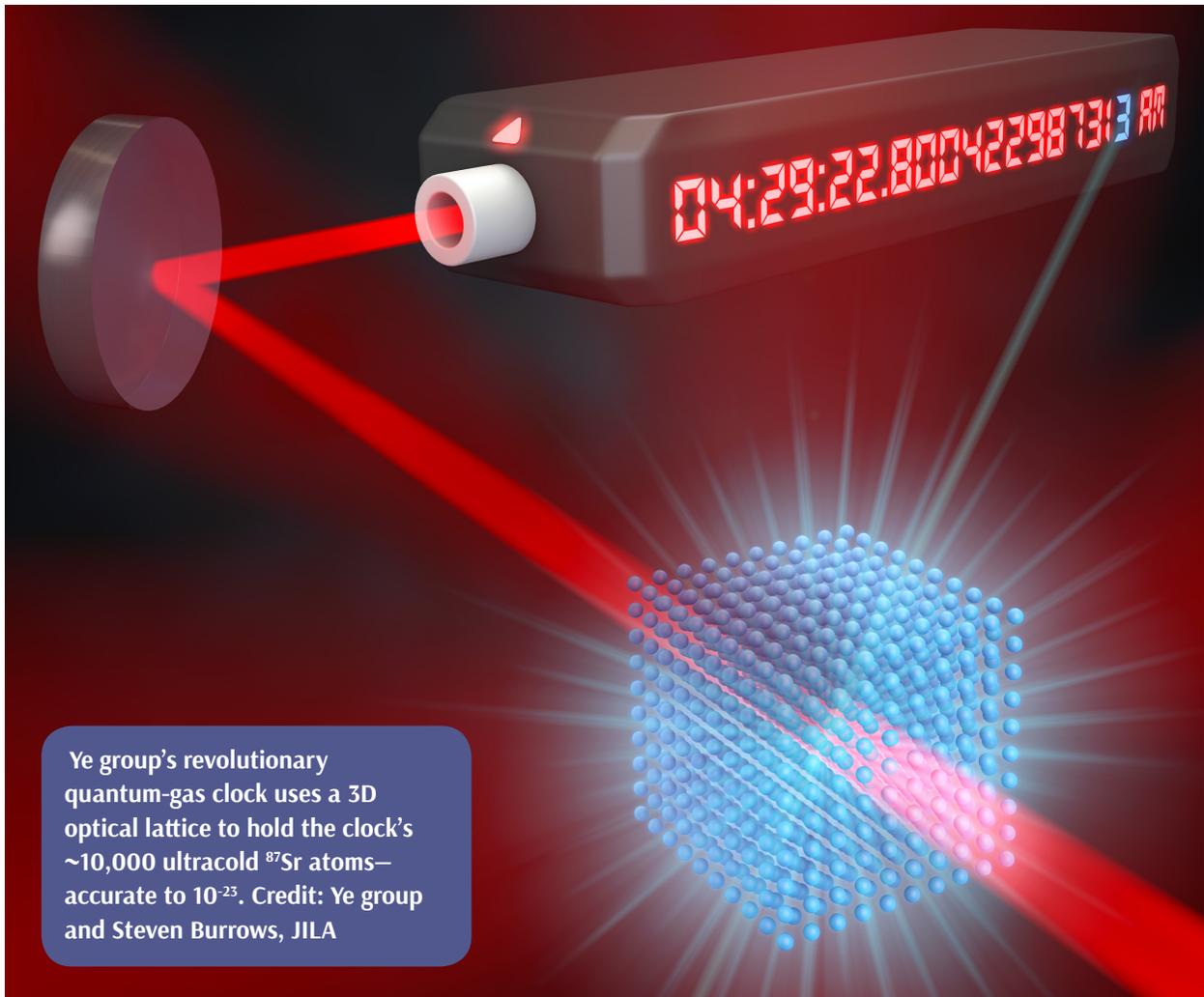
atoms' quantum correlations to significantly improve both the accuracy and speed of measurement.

The quantum-gas clock makes its measurements about 10 times faster (with 10 times less averaging time required) than previous clocks that use thermal atomic gases. Looking forward, as the Ye team further masters the details of quantum-gas clock technology (expected to be challenging, but doable), both the quantum-gas clock's accuracy and measurement speed should continue to significantly improve.

One new feature of the quantum gas clock was the placement of single  $^{87}\text{Sr}$  atoms in the energy wells of a three-dimensional optical lattice created by laser beams. This configuration allowed the researchers to control the quantum interactions of the atoms when they were in their lowest energy state. For instance, the 3D lattice prevented the atoms from colliding or interfering with each other. And, through their quantum interactions, the atoms collectively prevented any one atom from disturbing the others. These features ensured that all the atoms ticked at exactly the same rate, making it possible to make faster measurements.

This first quantum-gas optical clock is just the beginning of a revolution in precision measurement! By carefully honing every aspect of this first clock's performance, the Ye group hopes to someday leverage the clock's superior precision to achieve new levels of clock accuracy. Such a super-accurate clock would be useful not just for measuring time, but for precisely measuring an exceptionally

1. Because  $^{87}\text{Sr}$  atoms are independent-minded fermions, when they occupy their lowest energy state, they cannot form a Bose-Einstein condensate. Rather the atoms fill in lowest energy states like rungs on a ladder, with two atoms, one spin up and one spin down, on each rung.



Ye group's revolutionary quantum-gas clock uses a 3D optical lattice to hold the clock's ~10,000 ultracold  $^{87}\text{Sr}$  atoms—accurate to  $10^{-23}$ . Credit: Ye group and Steven Burrows, JILA

broad range of physical quantities that cause changes in the clock's frequency, i.e., ticking rate.

"I believe Jun's quantum gas clock is a transformational discovery," said Tom O'Brian, chief of JILA's Quantum Physics Division. "This is the beginning of an era of understanding, controlling, and applying quantum interactions to the science of all kinds of precision measurement and to using quantum interactions to probe some of the key challenges of fundamental physics.

"Ye group scientists have not just made a better clock. They are also helping lead a new revolution in quantum mechanics and measurement science."

The researchers responsible for this breakthrough in the fastest, most accurate measurement of time include research associates Sara Campbell, G. Edward Marti, and Aki Goban, graduate students Ross Hutson, Lindsay Sonderhouse, and John Robinson, JILA Ph.D. Ben Bloom, CU Boulder graduate Rees McNally, NIST colleague Wei Zhang, and Fellow Jun Ye. ✨

S. L. Campbell, R. B. Hutson, G. E. Marti, A. Goban, N. Darkwah Oppong, Rees McNally, L. Sonderhouse, J. M. Robinson, W. Zhang, B. J. Bloom, J. Ye, *Science* **358** 90–94 (2017).



## About JILA

JILA was founded in 1962 as a joint institute of CU-Boulder and NIST. JILA is located at the base of the Rocky Mountains on the CU-Boulder campus, next to the Duane Physics complex.

JILA's faculty includes two Nobel laureates, Eric Cornell and John Hall, as well as two John D. and Catherine T. MacArthur Fellows, Margaret Murnane and Ana Maria Rey. JILA's CU members hold faculty appointments in the Departments of Physics; Astrophysical & Planetary Science; Chemistry and Biochemistry; and Molecular, Cellular, and Developmental Biology as well as in the School of Engineering. NIST's Quantum Physics Division members hold adjunct faculty appointments at CU in the same departments.

The wide-ranging interests of our scientists have made JILA one of the nation's leading research institutes in the physical sciences. They explore some of today's most challenging and fundamental scientific questions about quantum physics, the design of precision optical and x-ray lasers, the fundamental principles underlying the interaction of light and matter, and processes that have governed the evolution of the Universe for nearly 14 billion years. Research topics range from the small, frigid world governed by the laws of quantum mechanics through the physics of biological and chemical systems to the processes that shape the stars and galaxies. JILA science encompasses seven broad categories: Astrophysics, Atomic & Molecular physics, Biophysics, Chemical physics, Laser Physics, Nanoscience, Precision Measurement, and Quantum Information.

To learn more visit:  
[jila.colorado.edu](http://jila.colorado.edu)

