

Searching for Time-Reversal Violation with Unstable Molecules

BY RONALD F. GARCIA RUIZ

The matter-antimatter imbalance in the universe

The visible universe is composed almost entirely of matter, while antimatter accounts for less than one part in a billion. Yet according to the known laws of physics, the Big Bang should have produced matter and antimatter in equal amounts. The large imbalance we observe today, with matter vastly prevailing, remains one of the most profound and unresolved questions in modern physics. Explaining this asymmetry likely requires discovering new sources of symmetry violation beyond those already known within the current framework of fundamental physics. These subtle effects may have shifted the balance toward matter in the earliest stages of the universe.

Symmetry principles are fundamental to our understanding of the physical universe, and play an essential role in guiding our understanding of elementary particles and their interactions. In physics, a symmetry means that a system remains unchanged under certain transformations. The fundamental symmetries, illustrated in Figure 1, include parity (P), charge conjugation (C), and time reversal (T). *Parity* involves inverting spatial coordinates, producing a mirror image of a physical process. *Charge conjugation* switches particles with their corresponding antiparticles, reversing the sign of charge. *Time reversal* refers to the invariance of physical laws when the flow of time is reversed. Of the four fundamental forces of nature—electromagnetic, gravitational, strong nuclear, and weak—the weak force is the only one that has been experimentally observed to violate C, P, T, and combined CP symmetries. Although CP violation is theoretically allowed within the framework of the strong interaction, such effects have not been observed. This striking absence is known as the strong CP problem, and it remains one of the outstanding puzzles in modern physics.

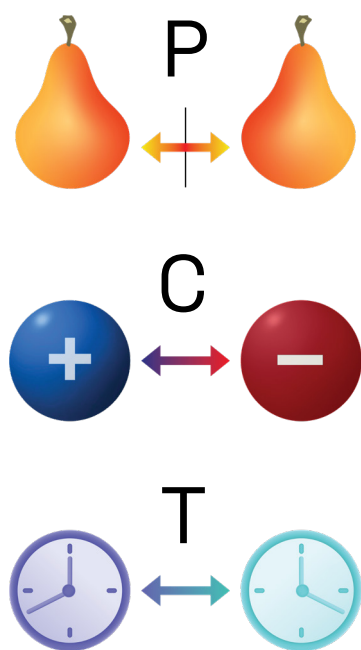
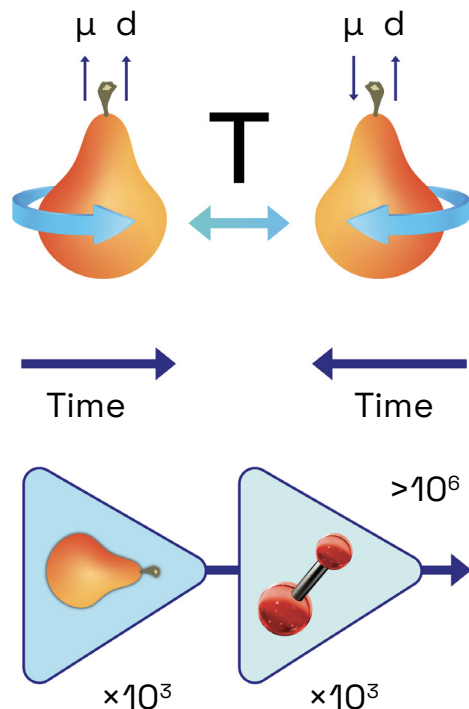


FIGURE 1: Fundamental symmetries—parity (P), charge conjugation (C), and time reversal (T)—are central to particle physics. P creates a mirror image of a process by flipping spatial coordinates, C transforms particles into their antiparticles, and T reverses the direction of time. Credit: Sampson Wilcox

In 1967, physicist Andrei Sakharov proposed a set of conditions that must be satisfied to explain the origin of the matter–antimatter asymmetry in the universe, requiring three necessary ingredients: violation of baryon number conservation, departure from thermal equilibrium, and violation of the combined charge conjugation and parity (CP) symmetry. CP symmetry was once thought to be universally conserved, until this view was overturned in 1964 with the discovery of CP violation in the decay of neutral kaons. This landmark result later earned the Nobel Prize in Physics in 1980 [James Watson Cronin and Val Logsdon Fitch], and more recently, CP violation has also been observed in baryon decays at CERN. However, the level of CP violation observed to date is too small to account for the vast predominance of matter in the visible universe. This points to the possible existence of new sources of CP violation beyond those described by the Standard Model of particle physics. Searching for such sources is one of the central goals of modern physics. One possibility is the existence of unknown, heavy particles at energy scales well beyond the reach of current particle accelerators like the Large Hadron Collider (LHC) at CERN. Although these particles may be too massive to be produced directly, they could leave detectable signatures by subtly altering the properties of known particles such as neutrons, protons, and atomic nuclei. To probe these effects, researchers are developing increasingly precise experiments that can measure tiny symmetry-violating properties using “table-top” experiments. In particular, molecules containing heavy, reflection-asymmetric nuclei have emerged as powerful systems for testing these ideas, due to their exceptional sensitivity to CP-violating effects [1].

Unstable molecules as amplifiers of time-reversal violation

Some atomic nuclei exhibit reflection-asymmetric shapes that deviate from spherical symmetry, resembling the shape of a pear, as illustrated in Figure 2. These nuclei also possess pairs of quantum states with the same total angular momentum (spin) but opposite parity, with nearly degenerate energies. If the fundamental laws of physics include even a slight violation of time-reversal symmetry (T), meaning they do not behave identically when time is reversed, these near-degenerate states can mix, and would mix more strongly than they would in symmetric nuclei. Smaller energy separation results the larger the mixing, and this enhanced mixing leads to a dipole-like asymmetry in the nuclear charge distribution. As the combined CPT

**FIGURE 2:**

Top row: a pear-shaped nucleus with both a magnetic dipole moment (μ) and a permanent electric dipole moment (d) has enhanced sensitivity to time-reversal (T) violation due to its large asymmetry of the charge distribution. Under time reversal, the spin and magnetic dipole moment reverse direction, while d remains unchanged, indicating a violation of time-reversal symmetry. *Bottom row:* by combining the molecular enhancement from the intense internal electromagnetic fields of polar molecules with the nuclear amplification offered by heavy, pear-shaped (octupole-deformed) nuclei, we can achieve over six orders of magnitude greater sensitivity to potential time-reversal symmetry violation compared to atoms composed of non-octupole-deformed nuclei. Credit: Sampson Wilcox

symmetry is a fundamental principle in the framework of quantum field theory, any violation of time-reversal symmetry is interpreted as a violation of CP symmetry to preserve overall CPT invariance. Consequently, the observation of a permanent electric dipole moment (EDM)—denoted as d —in a fundamental particle or a composite system like an atomic nucleus, implies the existence of CP violation. But how can one measure an electric dipole moment, d ? One approach is to apply an external electric field, E , and measure the interaction energy between the field and the dipole, given by the product $d \cdot E$. To maximize the observable effect, it is desirable to apply the strongest possible electric field. In a vacuum, electrostatic fields are typically limited to about 100 kV/cm. However, certain diatomic molecules can be fully polarized, generating internal effective electric fields on the order of 10–100 GV/cm, up to six

orders of magnitude larger than what can be applied in the laboratory [5, 8]. These large internal fields effectively amplify the interaction, making molecules powerful tools for probing fundamental properties of the particles they contain. The use of such molecular systems has enabled the most stringent experimental limits on the EDM of the electron [5, 8]. Similarly, atomic nuclei can also possess an EDM, arising from CP-violating properties of the protons, neutrons, and their constituent quarks. Moreover, certain unstable nuclei, such as radium-225, which has a large imbalance between protons ($Z = 88$) and neutrons ($N = 137$), can exhibit reflection-asymmetric shapes. These pear-shaped nuclei can lead to an enhancement of CP-violating effects by more than three orders of magnitude compared to spherical, symmetric nuclei. When such nuclei are embedded in polar molecules, the moments of the nuclear charge distribution interact with the large internal effective electric fields of the molecule, producing tiny shifts in molecular energy levels. Therefore, the energy shifts caused by CP-violating nuclear properties are effectively amplified at the molecular quantum levels. The resulting enhancement can exceed six orders of magnitude compared to atoms composed of symmetric nuclei, dramatically increasing the sensitivity for CP-violation searches.

From a beaker to precise quantum control

Nuclei exhibiting pear-shaped deformation are extremely rare in nature, and their large proton-to-neutron asymmetries make them unstable, typically possessing lifetimes of only a few days or less. Consequently, they must be artificially produced and are available only in trace quantities, often less than a microgram. This extreme scarcity poses significant experimental challenges, as their study requires highly sensitive and precise techniques. Moreover, even when such nuclei can be produced, the unambiguous identification of the molecules that they form remains difficult due to the lack of well-characterized spectroscopic signatures. Prior to our work, direct experimental studies of these exotic species were not feasible. Only recently did our group and collaborators develop the experimental methods that have, for the first time, enabled laser spectroscopy studies of molecules composed of unstable, short-lived nuclei [3, 6, 7].

An additional complication arises from the fact that these rare nuclei are produced alongside contaminants that can be more than eight orders of magnitude more abundant within a given sample. Consequently, a major

challenge is to begin with a compound—illustrated schematically in Figure 3 as a beaker—containing only a few atoms of the nucleus of interest, and from that starting point, efficiently form the desired molecule, separate it from the overwhelming background of contaminants, prepare it in a specific quantum state, trap it, and ultimately perform precision measurements. Over the past few years, our group has been steadily overcoming these challenges [2, 3, 6, 7, 9].

One of the most powerful techniques for studying individual atoms is laser cooling. In atomic systems, this method is well established and routinely applied to many elements that can be approximated as isolated two-level quantum systems. In contrast, laser cooling of molecules is a relatively recent and rapidly developing field. The presence of complex vibrational and rotational degrees of freedom in molecules makes it exceedingly rare to find species that can be effectively approximated as closed two-level systems. When a molecule is excited to an electronic state by a laser, it can decay into a wide range of vibrational and rotational levels, thereby complicating the implementation of efficient optical cycling and quantum control.

In a few rare cases, such as molecules formed by alkaline earth atoms bonded with fluorine, the molecular structure exhibits properties that are favorable for laser cooling. Motivated by this, our group and collaborators have performed precise spectroscopic measurements of radium monofluoride (RaF), enabling an experimental determination of its laser cooling scheme [7]. These exciting results open the door to achieving precise quantum control and interrogation of molecules, comparable to what has long been possible with atoms.

In collaboration with colleagues from Harvard, Caltech, and the new world-class Facility for Rare Isotope Beams (FRIB) at Michigan State University, we are developing an experimental platform to extract radium-containing molecules from liquid sources (such as the beaker illustrated in Figure 3) and to establish, step by step, the full sequence of techniques required to laser cool and trap molecules made of unstable, pear-shaped nuclei. We are optimistic that the unprecedented amplification of CP-violating effects offered by these molecules will enable the discovery of new sources of CP violation, potentially pointing to the existence of previously unseen heavy particles and shedding light on the matter–antimatter asymmetry of our visible universe.

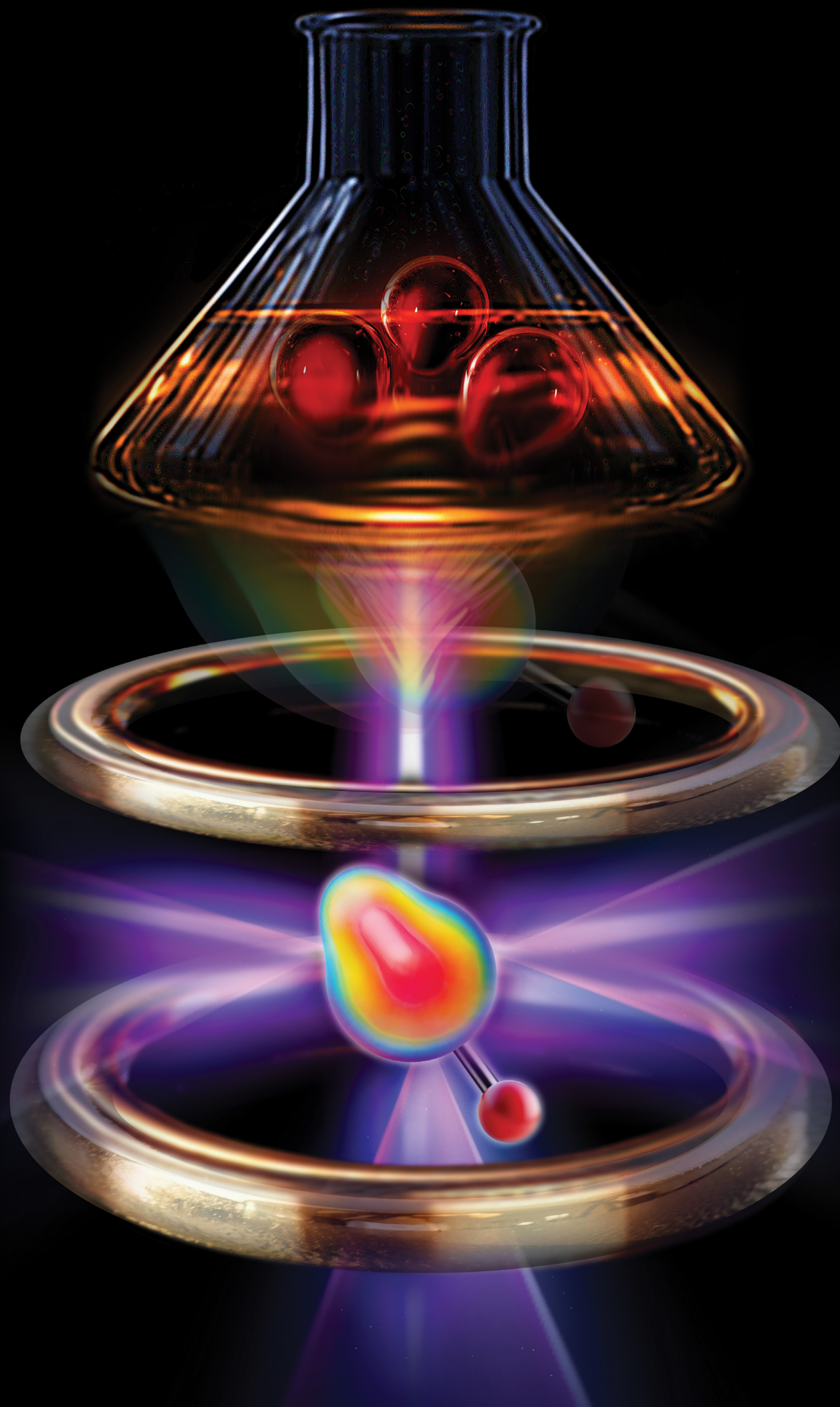
The existence of a heavy particle can induce a measurable electric dipole moment. Conversely, the absence of such a signal can be interpreted as excluding the presence of new heavy particles up to energy scales set by the precision of the experiment. The more precise the measurement, the higher the energy scale that can be probed. Owing to the large amplification provided by a heavy, pear-shaped nuclei, even a single trapped molecule could place constraints on new physics at mass scales exceeding 100 TeV, well beyond the reach of current high-energy colliders.

Molecules as an angstrom-scale laboratory for nuclear and particle physics

In addition to their extreme sensitivity to the dipole-like deformation of the nucleus, molecules can also be highly sensitive to a range of nuclear properties and fundamental interactions. A non-negligible overlap between the electron cloud and the nucleus, combined with the asymmetry of the molecular electron orbitals, enhances the sensitivity of the molecular quantum levels to short-range electron–nucleon and nucleon–nucleon interactions. This is especially relevant for probing yet unexplored electroweak nuclear properties, which remain extremely challenging to access in accelerator-based experiments.

Figure 4 illustrates a diatomic molecule such as RaF and highlights its hierarchical structure, from the molecular scale down to the subatomic level. In such a system, each atom consists of a nucleus surrounded by a cloud of electrons. These electrons interact with the protons and neutrons in the nucleus primarily via the electromagnetic force. In certain cases, particularly involving heavy nuclei, the weak interaction can also contribute significantly, especially in mediating rare processes or symmetry-violating effects. These fundamental forces influence the structure and energy levels of the molecule, which can be precisely probed using advanced laser spectroscopy techniques.

FIGURE 3:
From beaker to trapped molecule. A compound, represented by the beaker, contains only a few atoms of the rare nucleus of interest. These must be efficiently converted into the desired molecule. Multiple lasers are then used to prepare the molecule in a specific quantum state, trap it, and perform precision measurements. Credit: Sampson Wilcox



At higher levels of precision, molecular energy levels become sensitive to the internal structure of the atomic nuclei, where protons and neutrons themselves are composed of quarks bound by gluons via the strong nuclear force. Consequently, precision spectroscopy of molecular quantum states enables the investigation of diverse nuclear and particle physics phenomena. These include the possible existence of dark matter candidates and CP-violating effects in the strong nuclear forces, both of which are unresolved questions in modern physics.

Motivated by the exceptional sensitivity of molecules to subtle nuclear and particle physics properties, our group

at MIT is actively developing a series of tabletop precision experiments using different molecular systems [4]. These experiments aim to establish powerful new platforms for probing electroweak nuclear properties, testing the violation of fundamental symmetries, and exploring the existence of new physics beyond the Standard Model.

A new era of precision physics

In recent years, the development of highly precise experimental techniques, combined with advanced theoretical tools for describing fundamental particles, nuclei, atoms, and molecules, has opened up exciting

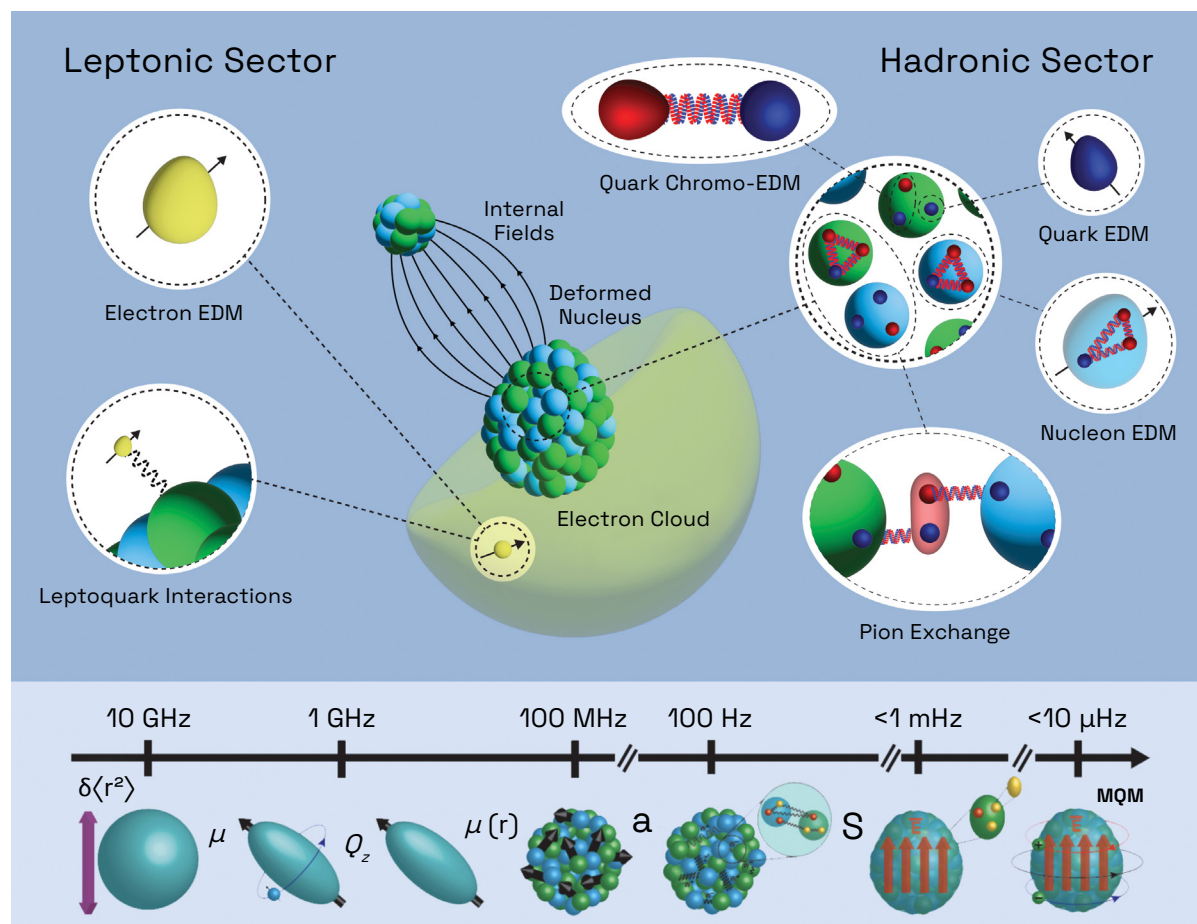


FIGURE 4:

Top: Schematic representation of a diatomic molecule and interactions among its fundamental constituents. Electrons interact with the protons and neutrons within each atomic nucleus primarily via the electromagnetic and weak forces. Inside the nuclei, additional complex interactions occur among protons and neutrons, as well as their fundamental building blocks—quarks and gluons—through a combination of the electromagnetic, weak, and strong forces. Within certain molecules the effective electromagnetic fields experienced by their constituents can be millions of times stronger than those achievable in the laboratory, leading to enhanced sensitivity to their intrinsic properties. *Bottom:* Illustration of how various nuclear features can affect the energy levels of molecules. From left: energy effects caused by changes in nuclear size, magnetic dipole moment, electric quadrupole moment, internal magnetization structure, effects of the weak nuclear forces, and the time-reversal violating dipole and magnetic quadrupole moments. The greater the precision of the measurements, the finer the resolution with which we can probe the microscopic structure of the nucleus and its fundamental constituents. Credit: Sampson Wilcox

new opportunities to explore previously inaccessible physical phenomena. In parallel, several major facilities in North America and Europe have been established to produce and deliver rare radioactive isotopes for research and industrial applications. As a result, experiments with these exotic systems are now becoming feasible in university laboratories such as ours at MIT.

Technological advances in cryogenics, buffer gas cooling, laser cooling, and ion trapping are making it possible to manipulate and study molecules made of unstable nuclei with unprecedented precision. At the same time, theoretical efforts are advancing *ab initio* calculations of molecular and nuclear structure, guiding experimental developments and enabling the interpretation of potential discoveries within the framework of particle physics.

While significant challenges remain, from the availability of unstable nuclei to the complexity of achieving precise quantum control and interrogation of these molecules, the field is advancing rapidly. As these techniques continue to mature, radioactive molecules are poised to enable a new experimental platform to push the boundaries of our understanding of the universe.

We anticipate that over the next decades, experiments with molecular systems conducted in university laboratories will uncover previously unknown properties of fundamental particles and interactions. These efforts have the potential to discover new fundamental particles and forces. In doing so, these unstable molecules serve as a unique bridge, connecting the properties of the smallest building blocks of matter with open questions about our macroscopic, asymmetric universe.

RONALD FERNANDO GARCIA RUIZ is the Thomas A. Frank Career Development associate professor in the Department of Physics at MIT. His research is focused on the development of laser spectroscopy techniques to investigate the properties of subatomic particles using atoms and molecules made up of short-lived radioactive nuclei. His experimental work provides unique information about the fundamental forces of nature, the properties of nuclear matter at the limits of existence, and the search for new physics beyond the Standard Model of particle physics.

Garcia Ruiz grew up in a small town in the Colombian mountains. As a teenager he moved to Bogota, where he obtained a bachelor's degree in physics in 2009 at Universidad Nacional de Colombia. After earning a Master's degree in Physics in 2011 at Universidad Nacional Autónoma de México, he moved to Belgium to start his PhD degree at KU Leuven. Garcia Ruiz was based at CERN during most of his PhD, working on laser spectroscopy techniques for the study of short-lived atomic nuclei. After his PhD, he became a research associate at the University of Manchester, UK (2016–2017). In 2018, he was awarded a CERN Research Fellowship to lead the local CRIS team. At CERN, he led several experimental programs motivated by modern developments in nuclear science, atomic physics and quantum chemistry. In January 2020, Garcia Ruiz joined the MIT Physics Department as an assistant professor and promoted to associate professor in July 2025.

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