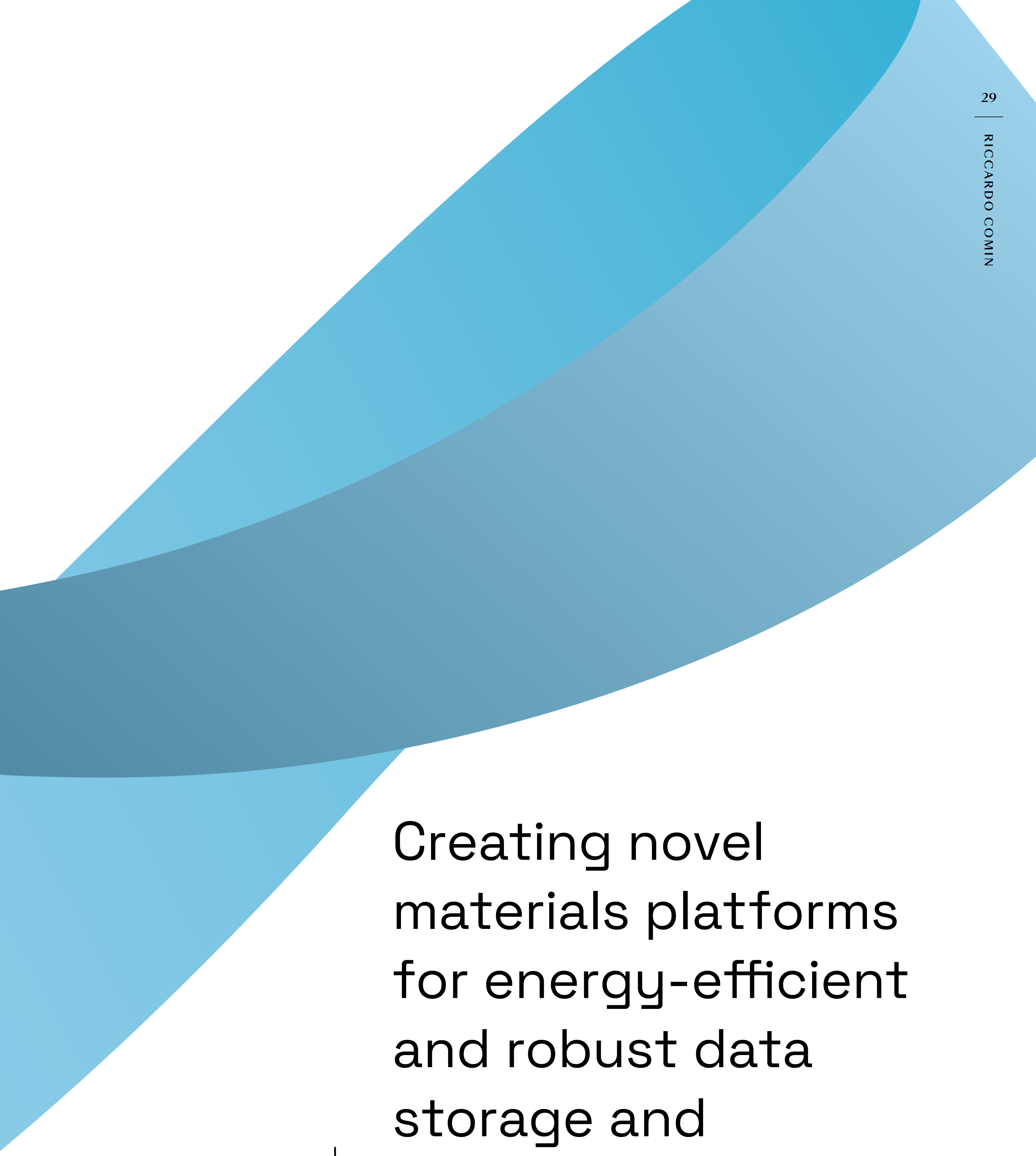
Chiral Quantum Matter



Creating novel materials platforms for energy-efficient and robust data storage and computation

BY RICCARDO COMIN

Chirality in nature

The term CHIRAL is used to describe a structure that cannot be mapped onto its mirror image by rotation or translation. Consider the old primary school trick: the Mobius strip, a strip of paper that, when twisted once around itself, miraculously only has one side. A Mobius strip is chiral: there are two possible ways to create it, depending on which way one twists the paper (Figure 1a). Try it yourself. No matter how hard you try, the left- and right-handed Mobius strips cannot fit together. The presence of chirality manifests in countless spheres of science, known by different names and carrying slightly different physical implications, but all due to the observation that somewhere, somehow, the universe is not symmetric, and we are still not entirely sure why.

This asymmetry is perhaps most well-studied in biology. There are numerous examples of molecules with the same chemical formula that, when the chirality is switched, become ineffective toward their intended purpose, and can sometimes even be harmful. Just like Mobius strips, molecules with incompatible chirality cannot fit together and participate in their intended reactions.

Deoxyribonucleic acid (DNA), the molecule that holds the genetic code of all living things, is chiral. In most organisms, DNA and ribonucleic acid (RNA), as well as their constituent molecules, exist only in the right-handed form, a phenomenon known as HOMOCHIRALITY.

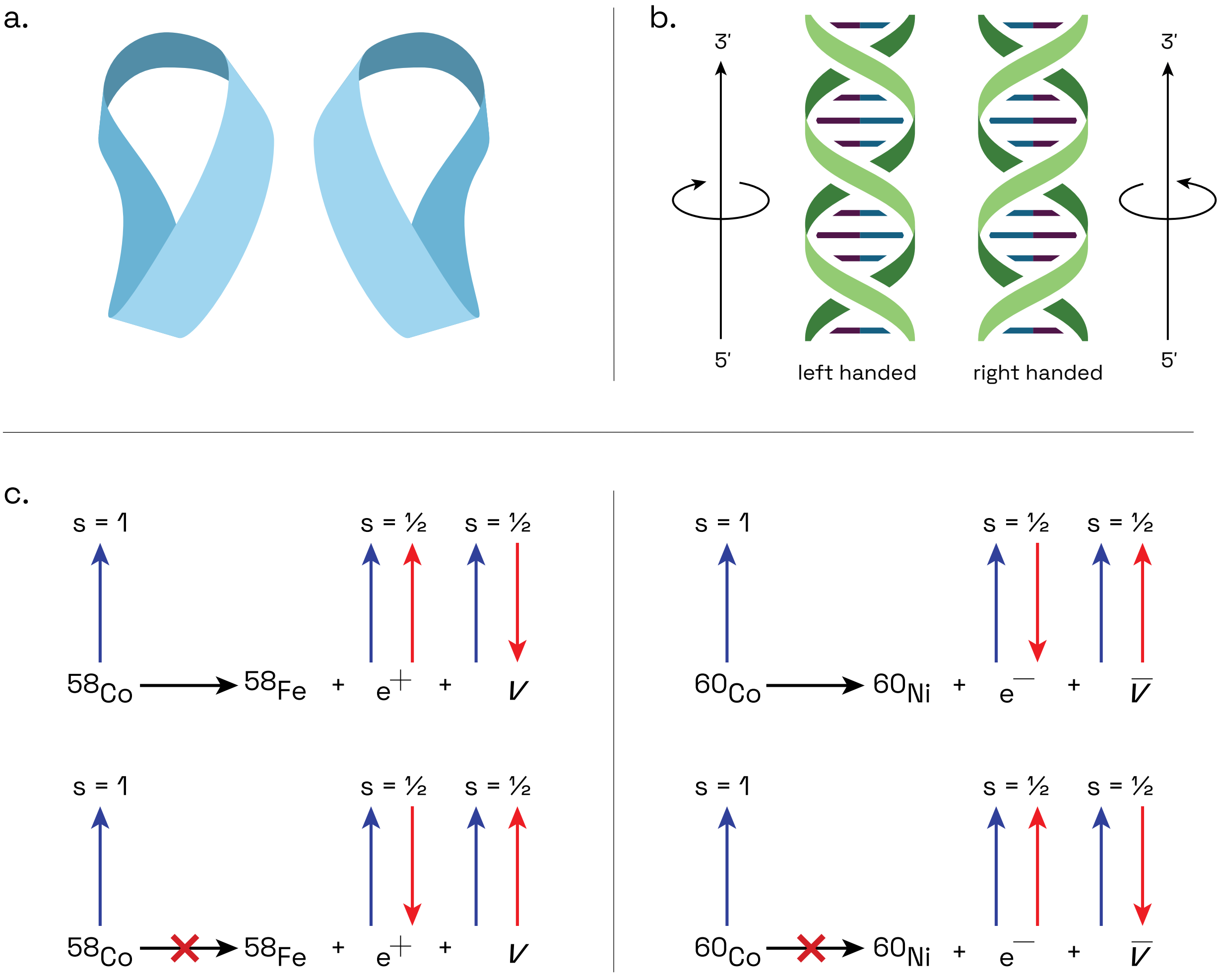


FIGURE 1:
(a) Left and right chiral Mobius strips, which cannot be superimposed with only rotation and translation.
(b) Left and right helical DNA, where the directional axis of the helix is defined by the 5'–3' axis.
(c) Beta decay of two isotopes of cobalt in the Wu experiment, including the directions of spin and linear momentum of each decay product. The top decay in each column is the only one observed: neutrinos are always emitted antiparallel to the spin of the cobalt nucleus, while antineutrinos are always emitted parallel. If parity is conserved in the weak interaction, both the top and bottom decay of each column should happen equally often.

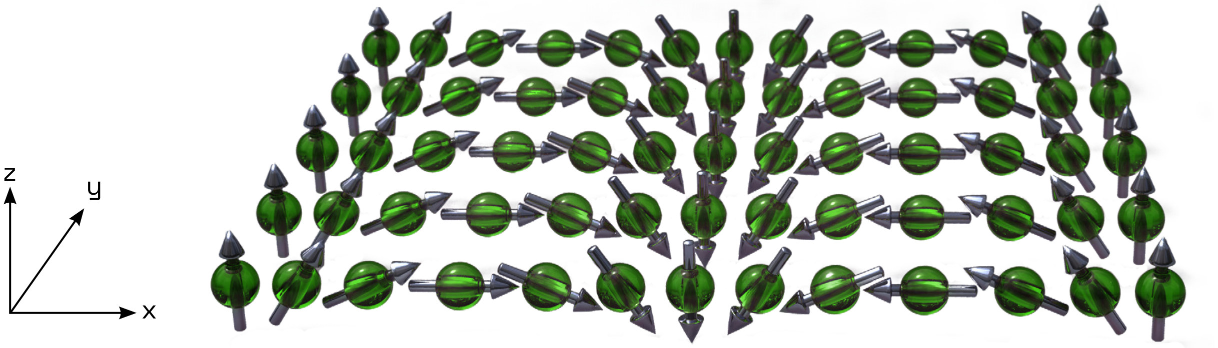


FIGURE 2:
Illustration of spin helix. This figure depicts a two-dimensional lattice characterized by a spiraling arrangement of electron spins. Atoms are represented by green dots, while the grey arrows indicate the local spin orientation at each atom. The spins are confined to the xz-plane, with propagation occurring along the x-axis.

By contrast, proteins and their building blocks—amino acids—naturally exist almost exclusively in the left-handed form. It is reasonable to assume that the homochirality of DNA should go hand-in-hand with the homochirality of amino acids, since they must interact very closely. However, the underlying reason for this imbalance is not known, and homochirality in life is a well-established topic of study for evolutionary biologists [1].

DNA is a special example of chirality because not only is it chiral, but it is also a helix. In fact, all helices are chiral, because there are two possible ways they can twist around an axis. To illustrate this example, the old rhyme to operate screws comes to mind: “right tight, left loose.” And when you come across a screw that tightens the other way, it is perplexing indeed. This raises a question: Why have we decided that DNA, in its most common form, is “right-handed”? What prevents us from saying that all DNA is “left-handed”? One must necessarily define conventions to decide on an absolute definition of right- and left-handed; DNA and amino acids are well studied enough that such conventions exist.

Accordingly, a chiral object with a well-defined propagation direction possesses HELICITY. In particle physics, helicity has a more subtle definition, but the symmetries that are broken as a result share interesting parallels. Fundamental particles, such as quarks, protons, and the exotic neutrino, have one of two possible assigned parities, just like they have an assigned mass and charge, which depends on their spin (intrinsic angular momentum). Helicity is defined as the product of their spin and direction of motion. Intriguingly, neutrinos always have a left-handed helicity, while antineutrinos always are right-handed. This phenomenon is known as PARITY VIOLATION and occurs in particle decays governed by the weak interaction, as discovered by Prof. Chien-Shiung Wu and colleagues in 1957 [2].

Chirality exists from macroscopic objects to the lightest known particles in existence. But why is DNA always right-handed in nature, and why are neutrinos always left-handed? Yes, it is true that the asymmetry in neutrinos is a product of the weak interaction, while prevailing theories for DNA homochirality err on the (relatively) more macroscopic side, but what causes these asymmetries? Could they be due to the same fundamental mechanism? As condensed matter physicists, we cannot study individual particles or molecules, but we can realize chirality from a collection of interacting electrons and spins in quantum materials. The *emergent* nature of chirality in quantum materials is rooted in the many-body nature of electronic phases of matter. Within the playground quantum materials, we can realize and study a diverse array of chiral phenomena, including many which cannot be found or engineered in any other physical system.

Magnetic chirality and magnetoelectric phenomena

MAGNETIC CHIRALITY, characterized by a geometrically nontrivial arrangement of electron spins, is a fascinating aspect of condensed matter physics that relates to the deeper symmetry properties of materials. This emergent phenomenon is characterized by spin moments that precess in space in a manner that defines a “handedness,” or, chirality. The resulting texture of spins can have right- or left-handedness, analogous to the helical twist of DNA. By contrast, simple collinear magnetic arrangements, such as those found in ferromagnets and antiferromagnets, exhibit zero chirality.

Notable examples of systems displaying spin chirality include helical, conical, and cycloidal spin structures. These configurations are characterized by a spin rotation plane and a spin propagation vector: the former defines the plane in which the spins lie, while the latter denotes the direction along which the spins twist. Helical spin textures exhibit a spiral plane perpendicular to the propagation vector; cycloidal structures have these elements aligned (Figure 2); and conical spins combine helical arrangements with a nonzero net magnetization.

To explore chirality in materials, the Comin group uses a technique called MAGNETIC CIRCULAR DICHROISM (MCD). This technique takes advantage of the natural helicity present in light, which comes in the form of left and right circular polarization. The interaction between light and matter enables the helicity of light to couple to the helicity (or handedness) of the chiral spin arrangement, leading to variations in the amount of light that is reflected or absorbed (Figure 3). In a similar vein, X-RAY MAGNETIC CIRCULAR DICHROISM (XMCD) provides a deeper understanding of chirality by harnessing the selective absorption of circularly polarized X-rays. This method has the additional benefit of relying on X-rays tuned at special energies where one can selectively probe the magnetic ions in the material. This allows for the precise determination of the spin texture. Furthermore, when XMCD is combined with X-ray scattering techniques, it becomes possible to quantitatively measure the pitch of a chiral magnet's structure, in a similar fashion as the original X-ray discovery of the double-helix structure of DNA by Rosalind Franklin in 1953 [3].

Ultrathin chiral quantum materials

An example of a spin chiral quantum material is the two-dimensional (2D) magnet nickel iodide (NiI_2). The atoms in this material are organized in a 2D triangular lattice. Each layer is composed of Ni^{2+} ions surrounded by six iodine ions forming a network of edge-sharing octahedral units (Figure 4a). These layers are stacked parallel to each other and are bonded weakly by van der Waals (vdW) forces. The vdW force is weak enough that atomically thin NiI_2 sheets can be simply separated by peeling them off using scotch tape, much like graphene.

In NiI_2, the Ni^{2+} ions possess unpaired spins within their 3d orbitals, for a total spin state of $S = 1$ (from two aligned spin-½ electrons). These spins can interact with adjacent spins, favoring either parallel (ferromagnetic) or antiparallel (antiferromagnetic) alignment. However, on a triangular lattice, where each spin is connected to two neighboring sites, this antiparallel alignment cannot be satisfied for all pairs of neighboring spins simultaneously. If two spins are antiparallel, the third spin cannot align antiparallel to both, due to the geometry of a triangle. To resolve this frustrated state, spins choose neither a parallel nor antiparallel configuration, and rather prefer to form a noncollinear state, *e.g.*, a spin helix.

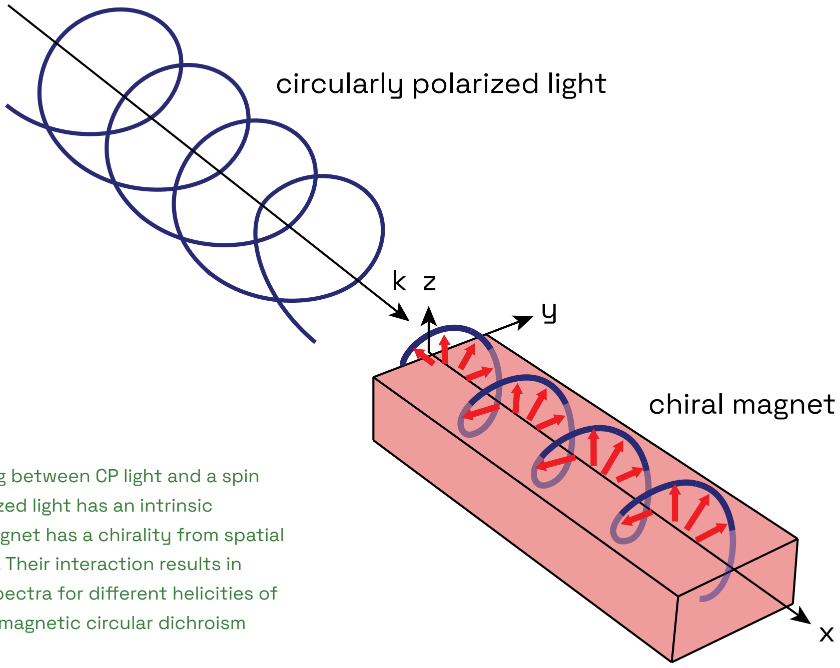


FIGURE 3:
Illustration of coupling between CP light and a spin helix. Circularly polarized light has an intrinsic helicity, and chiral magnet has a chirality from spatial alignment of its spins. Their interaction results in distinct absorption spectra for different helicities of light, showcasing the magnetic circular dichroism phenomenon.

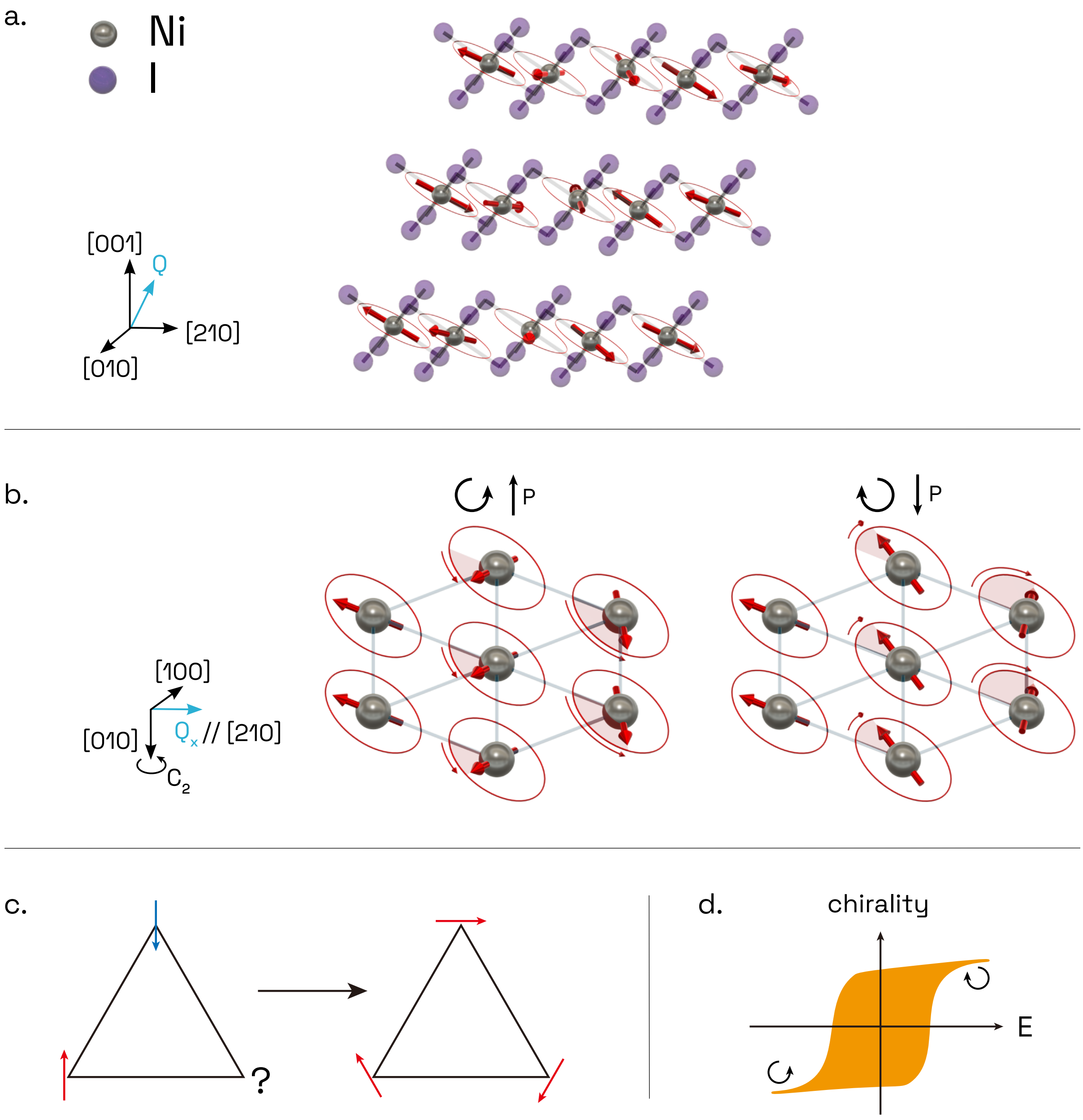


FIGURE 4:
(a) Crystal and magnetic structure of a spin chiral quantum magnet NiI_2. (b) Magnetoelectric coupling between spin chirality and electric polarization. (c) Geometric frustration of antiferromagnetic exchange in a triangular lattice. (d) Schematics of a multiferroic hysteresis loop. Spin chirality can be switched by an external electric field.

Such chiral magnetic order is stabilized in NiI_2 below T_N = 60 K. Figure 4a on page 33 depicts the spatial texture of this long-range helimagnetic order: it has periodicity of around seven Ni atoms, the spin helix propagation vector **Q** is oriented at 35 degrees from the planes, and the spins rotate in a plane normal to **Q**. Interestingly, these spin helices can induce an electric polarization **P**, which is akin to an internal electric field in the material. Such a polarization is a consequence of inversion symmetry breaking. As shown in Figure 4b, the counterclockwise rotation of spin creates an electric polarization pointing up, while the clockwise rotation creates an electric polarization pointing down. This strong coupling between magnetic order and electric polarization in NiI_2 exemplifies a magnetoelectric effect, which is very rare in natural magnetic materials.

While the helimagnetic order is the ground state of bulk NiI_2, whether it can survive in the two-dimensional (2D) limit of ultrathin samples has been a subject of debate. It is well-established that isotropic magnetic order cannot exist in the 2D limit because thermal fluctuations destroy such long-range order between spins. Against all odds, the multiferroic state persists in single atomic layers of NiI_2. The inherent multiferroic properties of

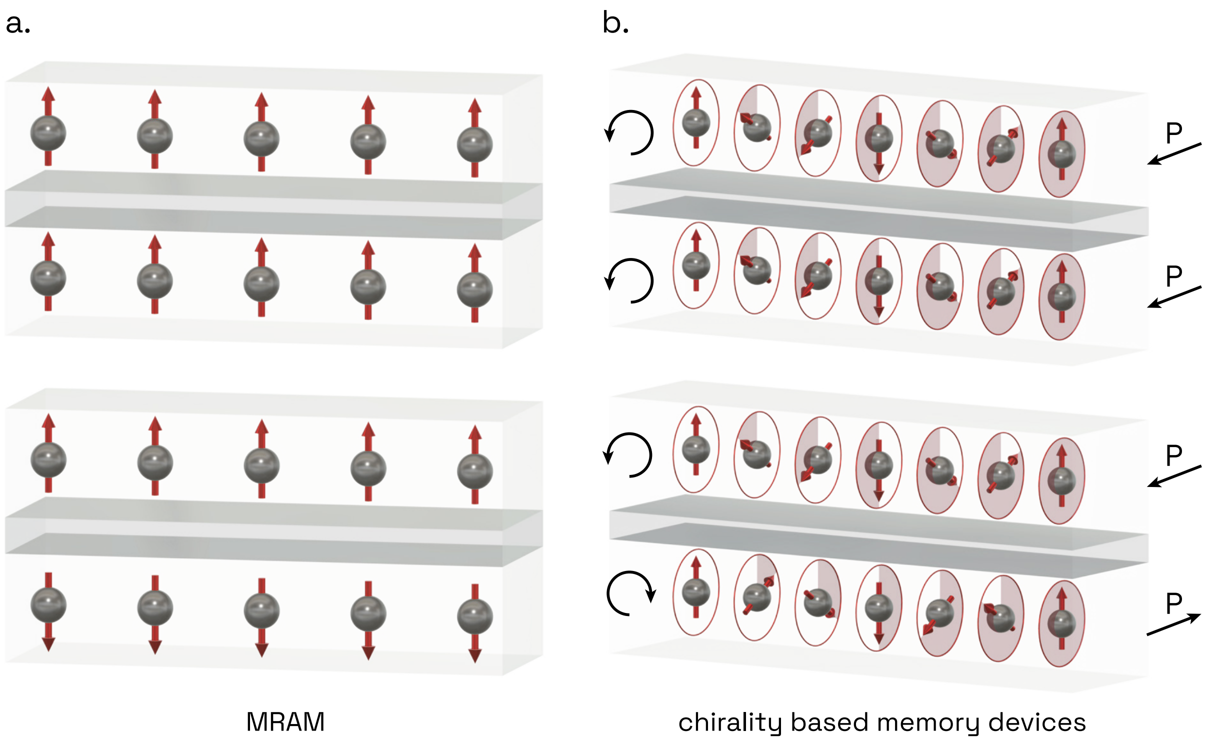


FIGURE 5:
(*Left*) Magnetoresistive random-access memory (MRAM) consists of two ferromagnetic layers sandwiching an insulating barrier. Parallel and antiparallel spin arrangement in the ferromagnetic layers generate two resistive states. (*Right*) Chirality-based memory device is composed of two multiferroic layers. Parallel and antiparallel spin chirality between two layers encode binary states. Left and right spin chirality are labeled ↺ and ↻.

two-dimensional NiI_2 opens doors to innovative technological applications, notably the possibility to use chirality to store information. The strong coupling between the spin chirality and electric polarization in NiI_2 provides a clear route to electrically switch chirality, or equivalently, to write a chiral 'bit.' As shown in Figure 4d, by reversing the direction of electric polarization, one can switch the spin chirality, making NiI_2 a potential candidate for advancing next-generation spintronic memories.

Applications of chiral quantum materials

This electrical control of spin chirality not only enriches the fundamental understanding of multiferroic materials but also paves the way for their utilization in future memory storage solutions, sensors, and transducers where electrical control of magnetic state is critical. Here, spin chirality can be used directly to encode information. The conventional method of encoding binary information ('0' and '1') in silicon-based devices uses charged and discharged states, a principle that underlies the operation of most semiconductor devices today. While silicon-based devices have revolutionized computing and electronic technologies, they are subject to several inherent physical limitations such as limited switching speed (in the gigahertz frequency range), poor energy efficiency, and volatile storage. The recent development of Magnetoresistive Random-Access Memory (MRAM) relies on spin as the information carrier, with an individual bit being composed of two ferromagnetic layers, with parallel and antiparallel spin alignment to encode information (Figure 5a). MRAM offers several advantages over charge-based storage mechanisms, such as non-volatility and high durability, but it is still limited by low energy efficiency and slow operation speeds. The adoption of spin chirality for information storage and manipulation has the potential to overcome both the challenges faced by charge-based (Silicon) and spin-based (MRAM) devices.

Most chiral spin systems have magnetic resonance frequencies over 1 terahertz, which enables fast electrical switching and high read/write speeds, potentially orders-of-magnitude faster compared to silicon-based devices. In addition to writing speed, spin chirality also has clear advantages in energy efficiency. The spin induced electric polarization in a chiral multiferroic is around four orders of magnitude lower than the conventional charge-based devices, resulting in ultralow energy consumption for the writing process. The energy density required to switch spin chirality is around 10^{-23} J/nm², or around six orders of magnitude lower than silicon based and MRAM (10^{-17} J/nm²) devices. With the recent explosive growth in data centers and energy consumption related to new and increasingly power-hungry technologies such as artificial intelligence, the development of novel materials platforms for energy-efficient and robust data storage and computation is a challenge of critical importance for modern society.

We designed a new type of highly energy-efficient memory device using a pair of spin chiral layers, separated by a thin insulating barrier, as shown in Figure 5b. Configurations of parallel and antiparallel spin chirality between the two layers encode the binary states '0' and '1.' In this sense, the spin chirality can be manipulated in the same way as the electron spin. However, with spin chirality being an emergent property of many spins, it is inherently robust. Further, spin chirality is rigidly coupled to the electric polarization, thus it can be switched by simply applying an in-plane voltage. The tunneling magnetoresistance across the two chiral multiferroic layers is used to read out the binary state, with parallel spin chirality giving lower resistance, and antiparallel spin chirality giving higher resistance. This mechanism is analogous to the tunnelling magnetoresistance phenomenon that is used in the read heads of MRAM devices.

While spin chirality-based memory devices hold exciting potential, there are important challenges that must be addressed for its practical applications. The largest issue is the low temperatures at which the chiral states are established. To date, the highest reported transition temperature at ambient pressure for a chiral multiferroic is 230 degrees Kelvin (-45 F), observed in cupric oxide (CuO). The pursuit of chiral multiferroic materials with stable properties at room temperature is a timely challenge. If such a material is discovered, these proposed spin chirality-based memory devices can be integrated into current electronic devices to improve both the computing power (high speed) and, perhaps most importantly, the energy efficiency of today's electronics.

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