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## Magnetism in Ancient Societies to the Present

Circa 1400 BCE to 2020 CE

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### 1.1 Introduction

Magnetic materials, in the form of lodestone, have been known to ancient cultures for many centuries. Both Greek and Chinese cultures have been widely recognized for their rich heritage in the historical foundations of magnetism for early lodestone instruments and related descriptive literature dating to several centuries before the common era (BCE). Here, we present published findings that supplement these many reports and support the use of magnetic materials by Mesoamerican cultures some 800–1000 years before the Greek and Chinese societies [1].

Lodestone is the descriptor assigned by historians to ancient magnetic ores of igneous, metamorphic, and sedimentary rocks consisting of not only mostly magnetite but also maghemite, hematite, and other more exotic oxide phases of lesser volume fractions. The principal phase, magnetite, having formula  $\text{Fe}_3\text{O}_4$ , is the iron oxide form of spinel ferrite,  $(\text{A})[\text{B}]_2\text{O}_4$ , where A and B are  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  cations occupying tetrahedrally and octahedrally oxygen-coordinated sites in the structure:  $(\text{Fe}^{3+})[\text{Fe}^{2+}\text{Fe}^{3+}]_2\text{O}_4^{-2}$ . Fragments of spontaneously magnetized lodestone have only been found near the earth's surface and not buried below its crust [2]. This observation fostered the belief that its permanent magnetic properties likely derive from lightning strikes.

When a naturally magnetized lodestone fragment is suspended in a fluid, it naturally orients toward a magnetic pole thus enabling the design and development of a magnetic compass.

### 1.2 Discovery and Ancient Applications of Lodestone: 1400 BCE–CE

#### 1.2.1 Magnetism in Ancient Mesoamerican Societies

##### 1.2.1.1 Olmec: Circa 1400 BCE to 400 BCE

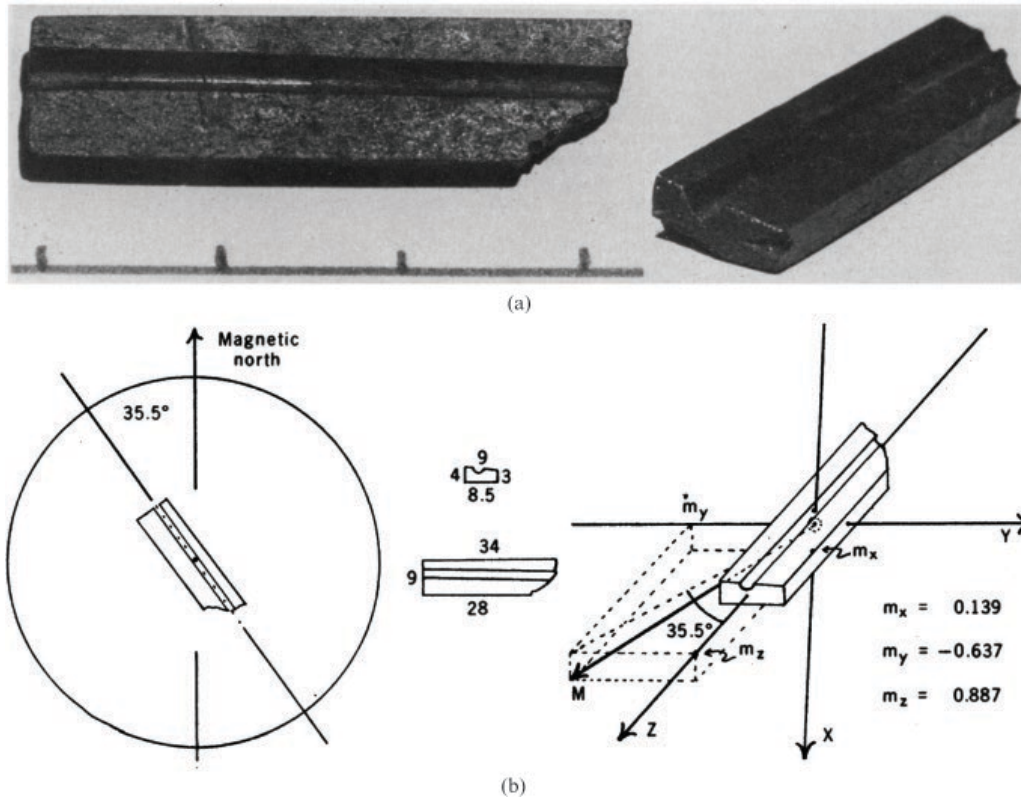
Challenging the general attribution of the origins of magnetism to the ancient Greek and Chinese societies, recent findings support the earliest discovery and applications of magnetism to the ancient Mesoamerican societies, i.e. the Olmec society of North America (c. 1400 BCE to 400 BCE), which is now located in the modern Mexican state of Veracruz [1].

An ancient Olmec magnetic artifact is shaped as an engineered bar composed of hematite impregnated with  $\text{Fe}_{2-x}\text{Ti}_x\text{O}_3$  lamellae [3] and is dated to more than a thousand years before the Greek and Chinese discoveries of magnetism. If true, this artifact would represent the first use of magnetic materials for navigation, or other forms of directional seeking, in world history.

Figure 1.1a contains a photograph of this artifact that shows what appears to be an engineered bar whose functionality remains the subject of much conjecture among historians.

##### 1.2.1.2 Monte Alto: Circa 400 BCE to 200 CE

Another ancient Mesoamerican society, the Monte Alto (c. 400 BCE to 200 CE), flourished in present-day Guatemala and produced sculpted potbellied figures (see Figure 1.2) that resemble those of the earlier Olmec civilization. It is unknown why Monte Alto sculptors incorporated magnetized ores into their work. Fu et al. [4] postulated the ability of such sculptures of leading authorities of the time to deflect a compass needle would have appeared quite impressive to any audience.



**Figure 1.1** (a) Olmec artifact (M-160) from reference 1 (dimensions in centimeters). (b, left) Top face and cross section of M-160 (dimensions in millimeters) and representation of the “floater” experiment showing the observed orientation  $35.5^\circ$  west of magnetic north. (b, right) Total magnetic moment vector  $M$  and components of M-160 [1].

The Monte Alto people were known to trade widely throughout North America. Lodestone tools able to detect magnetic anomalies, which had exotic or even perceived mystical properties, would have proven particularly valuable [4]. It is believed that magnetism had spread broadly throughout North America at this time.



**Figure 1.2** Photograph of a representative potbellied sculpture of the Mesoamerican era. Findings by Fu et al. [4] and others support a magnetic signature of the sculpture. The origins and purpose of the magnetic signature is as yet unclear.

### 1.2.1 Magnetism in Ancient Greek Societies

#### 1.2.2.1 Archaic: Circa 800–480 BCE

Chronologically, following the Mesoamerican societies are reports of the ancient Greek societies, circa 800 BCE to 500 BCE.

In this context, **Thales of Miletus** has been widely attributed as the first reporter of magnetism from these regions and eras. Aristotle attributed the first of what would be called scientific discussions on magnetism to Thales, who lived from about 625 BCE to about 545 BCE [5], is historically recognized as the first individual known to have entertained and engaged in scientific philosophy and is often attributed to the discovery of mathematics and is often referred to as the Father of Science.

Although none of Thales’s writings have been preserved, Aristotle, circa 384 BCE to 322 BCE, noted in *On the Soul* that Thales described the magnet as possessing a “soul” because it moves iron. This description has been corroborated by Laertius, circa 300 CE, in

*Lives of Eminent Philosophers* [5, 6]. The first attempts to provide a rational explanation for magnetism came concomitantly from **Empedocles of Akragas** (c. 495 BCE to 435 BCE), **Democritus of Thracia** (c. 460 BCE to 370 BCE), and from **Diogenes of Apollonia** (c. 450 BCE) [5, 6].

These philosophers attributed magnetism to the movement of iron particles on a “fluid” through air to the surface of lodestone entering surface “pores.” The forces of flow were implied to exist between both iron and lodestone but only iron particles were identified to physically flow to the lodestone and not lodestone particles to the iron. This one-way mechanistic particle flow interpretation of magnetism held for nearly six centuries [6].

During these eras, amber, i.e. fossilized tree resin, had been shown that upon rubbing with a cloth attracted small particles. This was known as the *amber effect*. **Plato**, circa 428 BCE to 348 BCE, speculated incorrectly that the amber effect, an attraction between electrostatic dipoles, and magnetism, an attraction between magnetostatic dipoles, shared a common origin [5, 6].

**Plutarch**, circa 46 CE to 119 CE, was first to postulate the attractive powers of magnets and amber derived from different phenomena [6]. Impressively for the day, Plutarch hypothesized the interpretation that would evolve to be termed “magnetic effluvium” and “electric effluvium” where the term effluvium was interpreted as a discharge. The concept of different discharges, one necessitated as Empedocles’s magnetic fluid, another as the electric fluid, was required as the relative strength of magnetism was many times stronger than that created by amber [6].

### 1.2.3 Magnetism in Ancient Chinese Societies

#### 1.2.3.1 Zhou Dynasty: Circa 1046 BCE to 221 BCE

Concurrent with, and independent of the Greek societies, the Chinese reference magnetism in the fourth century BCE. As stated in the *Book of the Devil Valley Master*: “When the people of Cheng go out to collect jade, they carry a south-pointer with them so as not to lose their way.” [7]

The “south-pointer” can only be reasonably interpreted as a lodestone compass. Compasses of that era used lodestone as a natural permanent magnet that aligned itself with Earth’s magnetic field exhibiting north–south polarity.

#### 1.2.3.2 Qin Dynasty: Circa 221 BCE to 206 BCE and Han Dynasty: Circa 206 BCE to 220 CE

The history of the compass was further developed during the Qin (221 BCE to 206 BCE) and Han dynasties (206 BCE to 220 CE). In the Qin dynasty, **Lüshi Chunqiu** explicitly asserted, circa 200 BCE, that “lodestone makes iron come or it attracts it,” and the earliest Chinese mineral diviners had experimented with tools that today would be describe as compasses [8].

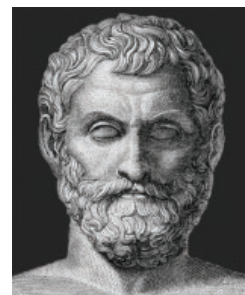
The earliest Chinese compasses include lodestone-shaped spoons or ladles that sat atop a flat, square-shaped plate, made of bronze, which served as a representation of Earth. The lodestone spoon balanced on its curved base and pointed toward the polar south [9] (see Figure 1.3).

The spoon compass was first mentioned by Wang Chong in *Lun Heng*, a book of essays on astronomy and meteorology, written during the Han dynasty. There he described this compass as: “When the south pointing spoon is thrown upon the ground, it comes to rest pointing at the south” [10, 11].

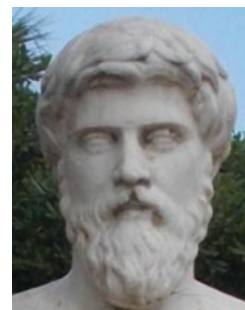
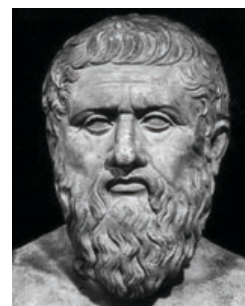
### 1.2.4 Magnetism Ancient Indian Societies

#### 1.2.4.1 Mahajanapadas: Circa 600 BCE–CE

Angutara Nikaya, a Buddhist scripture, mentions 16 great kingdoms, or Mahajanapadas, in India at the beginning of the sixth century BCE. These emerged during the Vedic Age (c. 1500 BCE to 500 BCE). The history of the emergence of Mahajanapadas is linked to the development of eastern Uttar Pradesh and western Bihar where agriculture thrived due to the availability of fertile lands and the abundance of iron ores. This resulted in the development of iron weapons and the subsequent expansion of Janapadas [12]. The ancient Indian text *Sushruta Samhita* was the first to describe the use of lodestone (i.e. magnetite) as a surgical tool used to remove arrow heads embedded in a person’s body. This is the earliest account of employing magnetism in human health care [13, 14].



Thales of Miletus



Plato and Plutarch



Lüshi Chunqiu (Courtesy of the National Palace Museum, Taipei, Taiwan, Republic of China)



Figure 1.3 Photograph of an early spoon compass circa 60 CE [11].

## 1.3 Discovery and Applications of Magnetism: CE to Circa 1800 CE

### 1.3.1 Magnetism in Chinese Tang and Song Dynasties

#### 1.3.1.1 Tang Dynasty (618 CE to 907 CE)



Shen Kuo

By the time of the Tang dynasty, and the beginning of the Northern Song dynasty (960 CE to 1127 CE), Chinese scholars devised a way to magnetize iron needles by rubbing them with magnetite and then suspending them in water. They also observed that needles heated to a high temperature and cooled in the north–south orientation (i.e. along the earth’s magnetic axis) would become magnetic. These more refined compasses could be floated in water as wet compasses, mounted on a shaft as dry compasses, or suspended from a thread, etc. Consequently, due to their portability, they were of greater practicality for purposes of navigation [15].

During the Song dynasty, many ships sailed as far as Saudi Arabia such that the compass was introduced to the Arab and European societies. The spread of the compass to Europe opened travel and led to the discovery of the New World.

#### 1.3.1.2 Song Dynasty (960 CE to 1279 CE)

The eleventh-century Chinese scientist **Shen Kuo** (1031–1095) wrote in the *Dream Pool Essays* in 1088 that the magnetic needle compass improved the accuracy of navigation by employing the astronomical true north [15, 16].

Shen Kuo provided the first detailed description of a magnetized needle compass. This invention enabled global exploration and international commerce. Remarkably, the floating needle compass of 1088 CE has not changed fundamentally in its components and construction to present-day compass manifestations.



Alexander Neckam

### 1.3.2 Magnetism in Ancient Arabic Societies: The Islamic Golden Age

Dry compasses appeared circa 1269 CE in Medieval Europe and 1282 CE in the Islamic Golden Age [17]. Circa 1300 CE, **Al-Ashraf Umar II** (1242–1296), a Yemeni physicist, astronomer, and geographer, and **Ibn Simón**, a Cairene astronomer, wrote individually treatises on the magnetic compass [18]. Al-Ashraf Umar II is known for writing the first description of the use of a magnetic compass for determining the qibla, i.e. the direction toward the Mosque in Mecca used by Muslims to determine the appropriate direction for prayer [18, 19].



Pierre de Mariciurt

### 1.3.3 Magnetism in European Eras: 1000 CE to 1800 CE

Later, compass designs included metallic iron needles. Magnetized needles and compasses were first described in Medieval Europe by the English theologian **Alexander Neckam** (1157 CE to 1217 CE) circa 1190 CE. Neckam preserved the earliest European reports of the magnetized needle as a guide to mariners [20, 21].

In the thirteenth century, **Peter Peregrinus (aka, Pierre de Mariciurt)** [22] wrote the first treatise describing the properties of magnets and pivoting compass needles as *Epistola de*

*magnete*. This tome, divided into two parts, part 1 focusing on inductive reasoning of definite experiences describing fundamental laws of magnetism and discussing for the first time the polarity of magnets and part 2 describing devices of the day utilizing the properties of magnets and their practical applications (e.g. the “wet” floating and “dry” pivoting compass) [23].

The mariner’s compass was further advanced by Italian inventor **Flavio Gioja** circa 1300 CE [24].

### 1.3.3.1 The Renaissance: Circa 1400–1700

**Leonardo Garzoni’s** (1543–1592) work, the *Due trattati sopra la natura, e le qualità della calamita*, is the first known example of a modern treatment of magnetic phenomena. Written circa 1580 CE, but never published, it was widely distributed in its time. In particular, Garzoni was referred to as an expert in magnetism by Niccolò Cabeo, whose *Philosophia Magnetica* (c. 1629 CE) is believed to be a worthy reproduction of Garzoni’s work [25–27].

In 1600 CE, **William Gilbert** (1544–1603) published his *De Magnete, Magneticisque Corporibus, et de Magno Magnete Tellure (On the Magnet and Magnetic Bodies, and on the Great Magnet the Earth)* [28]. In this work, he described many of his experiments from which he concluded that Earth was itself magnetic and that this was the reason compasses pointed north (previously, some believed that it was the pole star, Polaris, or a large magnetic mass at the north pole that attracted the compass) [29].

Although lodestone finds great utility in terrestrial, nautical, and celestial navigation, it was not until the mid-twentieth century that modern variants of lodestone, i.e. spinel ferrites, garnets, and hexaferrites, would be studied for their magnetic and electronic properties for use in power generation, conditioning, and conversion, thus setting society on a course of tremendous technological advance.

Nearly a thousand years after the discovery of the floating lodestone needle compass, ferrites were rediscovered and developed to address other technological needs ranging from communication, computation, transportation, energy and medical applications, and consumer electronics.

## 1.4 Development of Modern Magnetism: 1800 CE to Circa 2020 CE

### 1.4.1 Development of Classical Electromagnetism (1800 CE to 1900 CE)

The modern understanding of the relationship between electricity and magnetism began in 1819 with the work by **Hans Christian Ørsted** (1777–1851) who serendipitously noticed the twitching of a compass needle by a nearby electric current and inferred an induced magnetic field was generated by the current. **André-Marie Ampère** (1775–1836) followed in 1820 with the postulate that any induced magnetic field was related to an electrical current, provided that the electric field does not change with time [30]. **Johann Friedrich Carl Gauss** (1777–1855) and **Jean-Baptiste Biot** (1774–1862) and **Félix Savart** (1791–1841), also in 1820, derived the Biot–Savart law that quantitatively relates the magnetic field amplitude to a current-carrying wire (for example) and is analogous to Coulomb’s law for electricity [31]. **Michael Faraday** (1791–1867), in 1831, reported that a time-varying magnetic flux through a loop of wire induces a voltage. Faraday also established that magnetism could affect the polarization of light providing the foundation for nonreciprocity in today’s RF and optical devices [32]. He similarly discovered the principles of electromagnetic induction (near simultaneously with the American scientist Joseph Henry who also popularized the electromagnet as a practical device) [31, 33, 34] and mutual induction – the basis for modern power converters and transformers [35], as well as diamagnetism, and the fundamentals of electrolysis. His inventions of rotary machines included the first ever generator of electricity and electric motor technology that enabled and accelerated the industrial revolution [36, 37].

These founding fathers of electromagnetism laid the early foundational stones for examination by Maxwell.



William Gilbert



Hans Christian Ørsted



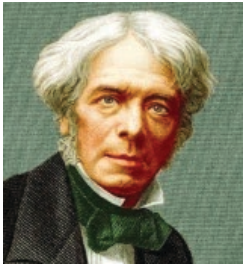
André-Marie Ampère



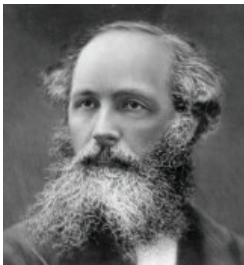
Johann Friedrich Carl Gauss



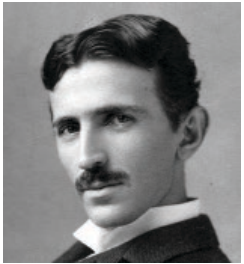
Jean-Baptiste Biot and Félix Savart



Michael Faraday



James Clerk Maxwell



Nikola Tesla



Hendrik Lorentz

**James Clerk Maxwell** (1831–1879), circa 1861, expanded upon those ideas and produced his astonishing revolutionary formulas, unifying all electricity, magnetism, and optics into the modern field of electromagnetism. With the development of his four principal equations for electromagnetism, Maxwell demonstrated that electric and magnetic fields travel through space as waves moving at the speed of light. He proposed that light is an undulation in the same medium as that hosting electric and magnetic phenomena [38]. The unification of light and electrical phenomena led to his prediction of radio waves. Maxwell is regarded as the founder of the modern field of electrical engineering [39].

In more practical developments, in 1887, **Nikola Tesla** (1856–1943) developed an induction motor that ran on alternating current, a power system that was rapidly expanding in Europe and the United States because of its advantages in long-distance, high-voltage transmission. The motor used polyphase current, which generated a rotating magnetic field that turned the motor, a principle that Tesla conceived of in 1882 [40–42]. His electric motor was a simple self-starting design that did not require a commutator, thus avoiding sparking and high maintenance [43, 44].

Approaching the turn of the century, **Hendrik Lorentz** (1853–1928) derived the modern formula for electromagnetic force that included contributions from both electric and magnetic fields. Lorentz began by distinguishing between matter and the luminiferous aether and applying Maxwell's equations at a microscopic scale. Using Oliver Heaviside's version of Maxwell equations for a stationary ether and applying Lagrangian mechanics, he derived the now widely used Lorentz law for electromagnetic force [45].

In 1892, Lorentz further introduced a new time concept, i.e. local time, that depended on universal time and a person's particular location to describe electromagnetic phenomena in reference frames that moved relative to the said luminiferous aether [46]. In 1900, Henri Poincaré called local time Lorentz's "most ingenious idea" [47]. In 1892, attempting to explain the Michelson–Morley experiment (which disproved that light waves were carried on the luminiferous aether) [48], Lorentz proposed that moving bodies contract in the direction of motion [49]. In 1899, Lorentz added time dilation to his transformations and published what Poincaré in 1905 named Lorentz transformations [50]. Lorentz's covariant formulation of electrodynamics, in which electrodynamic phenomena in different reference frames are described by identical equations with well-defined transformations, recognized that the outcomes of electrodynamic experiments do not depend on the relative motion of the reference frame. These ideas proved foundational to the work of Albert Einstein in his development of the theories of special and general relativity [51].

#### 1.4.2 Classical Magnetism: Enter the Electron (1900–1925)

**Pierre Curie** (1859–1906) was a pioneer in crystallography, magnetism, piezoelectricity, and radioactivity. Before his studies on magnetism, he designed and perfected sensitive torsion balances for measuring magnetic coefficients. With these tools, Curie studied ferromagnetism, paramagnetism, and diamagnetism and discovered the effect of temperature on paramagnetism which is known as Curie's law. He also discovered that ferromagnetic materials exhibit a critical temperature transition, above which the materials lose ferromagnetic behavior. This is now known as the Curie temperature. In addition to ferromagnetic materials, the Curie temperature

is used to study plate tectonics, treat hypothermia, and understand extraterrestrial magnetic fields, among other effects [52].

**Pierre Weiss** (1865–1940) developed the domain theory of ferromagnetism in 1907 [53]. Weiss also developed the Weiss domains, the Weiss mean field theory (MFT), and the Curie–Weiss law. Of these, MFT was perhaps his most significant contribution. The main idea of MFT is to replace all interactions relative to any one body with an average molecular field. This reduces any many-body problem to an effective one-body problem. The ease of solving MFT problems means that insight into the behavior of a complex system can be readily obtained with little effort. MFT has been widely applied to fields outside of physics, including statistics, graphical models, neuroscience [54], artificial intelligence, epidemic models [55], queueing theory [56], and computer-network performance and game theory [57]. Alongside Auguste Picard, Pierre Weiss is considered one of the discoverers of the magneto-caloric effect (1917) [58].

**Joseph John (J. J.) Thomson** (1856–1940) published a number of influential papers addressing problematic issues of electromagnetism at the turn of the twentieth century. He examined the electromagnetic theory of light of James Clerk Maxwell, introduced the concept of electromagnetic mass of a charged particle, and demonstrated that a moving charged particle increases in mass [59]. In 1897, Thomson suggested that one of the fundamental units contributing to the structure of atoms was more than 1000 times smaller than the atom then thought to be a singular unit. Thomson discovered this through his careful experiments on the properties of cathode rays following his discovery that such rays travel much further through air than expected if the rays consisted of atom-sized particles [60, 61]. His experiments suggested that cathode rays were more than 1000 times lighter than the hydrogen atom and that their mass did not vary from their atomic source. He concluded the rays were composed of very light, negatively charged particles that later scientists would name the electron [62]. The charge of the electron was measured with precision by Robert A. Millikan’s oil drop experiment in 1909 [63].

### 1.4.3 Electron Spin

An essential property of an electron is its spin. Spin is an intrinsic form of angular momentum necessary to the origins of magnetism. Spin is one of two types of angular momentum in quantum mechanics, the other being orbital angular momentum. Where the spin angular moment refers to the intrinsic spin of the electron (spin  $\pm \frac{1}{2}$ ), the orbital angular moment component derives from a classical analog of a charge orbiting about its nucleus akin to a charge traveling through a loop of wire, giving rise to an axial magnetic field through its center.

**Wolfgang Pauli** (1900–1958) in 1924 proposed the doubling of available electron states by introducing two-valued “hidden rotation” [64]. In 1925, **George Uhlenbeck** and **Samuel Goudsmit** suggested the physical interpretation of a particle spinning on its own axis [65]. The mathematical theory was worked out by Pauli in 1927 [66].



Pierre Curie and Pierre Weiss



J. J. Thomson



Wolfgang Pauli, George Uhlenbeck, and Samuel Goudsmit



Louis de Broglie and Erwin Schrödinger

#### 1.4.4 Wave Mechanics

**Louis Victor Pierre Raymond, 7th Duc de Broglie** (1892–1987), commonly referred to as Louis de Broglie, was a French physicist and aristocrat who made groundbreaking contributions to quantum theory. In 1924, he postulated the wave nature of electrons and suggested that all matter possessed wave properties. This concept is known as the de Broglie hypothesis, an example of wave–particle duality, and forms a central tenet of the theory of quantum mechanics. de Broglie won the Nobel Prize for Physics in 1929, after the wave-like behavior of matter was experimentally verified [67].

The de Broglie hypothesis was later used by **Erwin Schrödinger** (1887–1961) in his formulation of wave mechanics. In 1926–1927, Schrödinger published a series of influential papers on wave mechanics and presented what is known as the Schrödinger equation and derived it for time-independent systems and showed that it gave correct energy eigenvalues for the hydrogen atom. He went on to show the equivalence of this approach with that of Heisenberg’s and extended his approach to time-variant systems, introducing complex solutions to wave equations in order to prevent higher order (i.e. fourth and sixth orders) differential equations and ultimately reducing them to first-order differential equations, making quantum mechanics accessible to common practitioners [68].

#### 1.4.5 Quantum Theories of Magnetism

In 1927, **Walter Heitler** and **Fritz London** successfully applied Schrödinger’s theory to address bonding in dihydrogen (i.e.  $H_2$ ). This theory introduces the first principles of how two hydrogen atom wave functions join to form a covalent bond [69]. **Linus Pauling** built upon the HL theory and the work of Lewis (of ionic bonding theory) to establish *resonance* and *orbital hybridization*, two key foundations of valence bond theory (VBT) that led to his Nobel Prize in Chemistry in 1954. Today, modern VBT is complemented by molecular orbital theory (MOT) as pillars of quantum chemistry. The principal difference between these theories is that VBT treats electron pairs as localized to the parent atoms, whereas MOT treats electron pairs as molecular orbitals that may extend over the entire molecule. Importantly, MOT is more readily adaptable to predicting magnetic properties.

MOT was developed through the efforts of **Friedrich Hund**, **Robert Mulliken**, **John C. Slater**, and **John Lennard-Jones** [70]. The first quantitative use of MOT was the 1929 work of Lennard-Jones [71] who predicted a triplet ground state for the dioxygen molecule that explained its paramagnetism.

The success of MOT spawned ligand field theory, which was developed during the 1930s and 1940s as an alternative and enhancement to crystal field theory.

**Werner Karl Heisenberg** (1901–1976) contributed significantly to quantum magnetism in calculating the exchange term,  $J_{jk}$ , which appears in the context of chemical bonding, spectroscopy, and MFT (see Curie–Weiss) to be of central import in explaining ferromagnetism. In order to calculate the effect, he used the many-electron wave function to be a Slater determinant so that it is antisymmetric, thus making sure all electrons obey Pauli’s principle. Heisenberg proceeded to calculate the magnetization of such systems such that  $J_{jk} > 0$ , with the triplet state having the lowest energy. This state, in which the two neighbor spins are aligned in the same direction, is energetically favorable and represents the case for all ferromagnetic materials such as Fe, Co, Ni, and some rare earth elements.



Werner Karl Heisenberg

Modern theories of magnetism extensively use a Hamiltonian called the “Heisenberg exchange Hamiltonian” to investigate the magnetic properties of materials. The exchange term,  $J$ , was put to use by many notable physicists, most notably Néel who postulated that the exchange could take on negative values and thus give rise to antiferromagnetic (or ferrimagnetic) order in which nearest-neighbor spins antialign [72]. Other theories that employ common concepts include superexchange [73]. These phenomena have their origin in the Heisenberg exchange mechanism that have been validated by neutron scattering developed by contributions of Shull et al. [74].



The first step in the development of a new quantum theory was taken in 1925 when **Paul Adrien Maurice Dirac** (1902–1984) received a paper from Werner Heisenberg who revisited the old quantum theory of Bohr and Sommerfeld and changed the equations so that they were constrained by directly observable quantities, leading to a matrix formulation. Dirac's attention was drawn to an enigmatic mathematical relationship of noncommuting dynamical variables.

In 1928, Dirac derived his relativistic equation of motion for the wave function of the electron [75]. This work led Dirac to predict the existence of antiparticles such as the positron, the electron's antiparticle [76], which was later observed by Carl Anderson in 1932 [77]. Dirac's equation also contributed to explaining the origin of quantum spin as a relativistic phenomenon.

Dirac is regarded as the founder of quantum electrodynamics (QED), being the first to use that term. He also introduced the idea of vacuum polarization (1930) [78]. This work was key to the development of QED by the next generation of theorists, in particular **Julian Schwinger** [198], **Richard Feynman** [79], **Shin'ichirō Tomonaga** [80], and **Freeman Dyson** [81], in their formulation of modern QED.

In 1931, Dirac proposed that the existence of a single magnetic monopole in the universe would explain the quantization of electrical charge [82]. To date, no direct evidence supports their existence.



Paul Adrien Maurice Dirac

#### 1.4.6 Density Functional Theory (1970s–Present)

Density functional theory (DFT) is a powerful computational quantum mechanical modeling method used in physics, chemistry, and materials science to investigate the electronic structure (or nuclear structure) principally of the ground state of many-body systems, in particular atoms, molecules, and the condensed phases. Using this theory, the properties of many-electron systems can be determined by using functionals, i.e. functions of other functions. In the case of DFT, these are functionals of spatially dependent electron density.

DFT is among the most popular and versatile methods available in condensed-matter physics, computational physics, and computational chemistry.

Although DFT has been commonly applied to problems in solid-state physics as the 1970s, it was not considered sufficiently accurate for calculations in quantum chemistry until the 1990s, when approximations were greatly refined to better approximate exchange and correlation interactions.

Recent advances in DFT applied to magnetic oxide systems derive from basic observations in applying the Hubbard model. The Hubbard model, introduced in 1963 [83], is based on the tight-binding approximation in which electrons are viewed as occupying standard orbitals “hopping” between atoms during conduction. The competition between the hopping integral and the onsite repulsion accounts for the transition from conductor to insulator in many transition metal oxide systems, including many magnetic oxides such as the ferrites. As previously mentioned, the superexchange interaction ( $J$ ) between cations mediated by an anion is often found to be negative. As  $J$  is negative, the ground state is the antialignment of near-neighbor cation spins [84], i.e. antiferromagnetic or ferrimagnetic states. This has been confirmed by neutron diffraction experiments [85].

DFT was put on a firm theoretical footing by **Walter Kohn** and **Pierre Hohenberg** with the conception of the two Hohenberg–Kohn (HK) theorems [86]. The original HK theorems applied only to nondegenerate ground states in the absence of a magnetic



Walter Kohn, Pierre Hohenberg, and Lu Jeu Sham

field [87, 88]. The first HK theorem demonstrated that ground-state properties of many-electron systems are uniquely defined by electron densities that depend on spatial coordinates. This theorem identifies a path to reducing the many-body problem of  $N$  electrons through the use of functionals of the electron density. The second HK theorem defined an energy functional and proved the ground-state electron density for minimization of that functional. In a later work, the HK theorem was further developed to produce the Kohn–Sham DFT to address the intractable many-body problem of interacting electrons in a static potential [89]. The effective potential includes the external potential and the effects of Coulomb interactions between the electrons, e.g. the exchange and correlation interactions. Modeling the latter two interactions become the challenge of KS DFT.

To date, challenges remain when vector potentials are introduced to DFT such as in the case of magnetic fields. Other challenges include accounting for strongly correlated systems, spin–orbital coupling, van der Waals bonding, and finite temperature effects and phase transitions.

#### 1.4.7 Development of Commercially Viable Magnetic Materials (1900–2020)

At the beginning of the twentieth century, circa 1909, **Hilpert** patented the first descriptions detailing the processing and application potential of ferrites as insulating magnetic materials for “Electric and Magnetic Apparatus.” However, this early attempt to develop ferrites for commercialization did not meet with immediate success. The reason was that the intrinsic magnetic properties of ferrites were not competitive with existing ferromagnetic steels for low frequency technologies under development at the time. The value of insulating magnets at the turn of the century had yet to be made clear and Hilpert’s invention was far ahead of its time.

In 1917, **Kotaro Honda** invented KS steel that was 0.4–0.8% carbon, 30–40% cobalt, 5–9% tungsten, and 1.5–3% chromium. KS steel was a permanent magnetic steel with three times the magnetic resistance of tungsten steel. He later invented NKS steel whose magnetic resistance is several times that of KS steel [91, 92].

Metallurgist **Tokushichi Mishima** in 1931 invented MK steel. MK steel is regarded as the precursor to AlNiCo magnets. AlNiCo magnets are regarded as the first generation of permanent magnets. Mishima discovered that aluminum restores magnetism to nonmagnetic nickel steel. MK steel, which is an extremely inexpensive magnetic alloy, is closely related to modern AlNiCo magnets [92].

In 1934 and 1935, approximately 25 years after Hilpert, **Yogoro Kato and Takeshi Takei** rediscovered spinel oxides of zinc and iron had unique and attractive high-frequency magnetic properties. Tokyo Denki Kagaku Kogyo, now known as the TDK Corporation, was founded in 1935 to commercialize these ferrite materials. The collaborative works of Tokyo Institute of Technology and TDK led to the development of CuZn-ferrite cores well suited for practical applications at frequencies up to  $\sim 1$  MHz [95].

TDK began mass production of ferrite cores in 1937 under the product name “Oxide Core.” Most of the shipped product were used in oscillators, mixers, and transformers for radios to meet the needs of the rapidly escalating Pacific Rim war that would become World War II. These were the world’s first practical uses of ferrite materials [96].

#### 1.4.8 Snoek and the Philips Company

Ferrite materials are a unique class of insulating magnetic oxides that possess moderate values of magnetization, high permeability, high permittivity, and low losses at frequencies up to and beyond millimeter wavelengths.

These properties afford great value to high-frequency devices that require strong coupling to electromagnetic signals while experiencing low losses. Additionally, due to their intrinsic magnetism, these materials also provide time-reversal symmetry breaking that leads to nonreciprocal behavior essential to many device applications in radar and communications systems as isolators, circulators, phase shifters, etc.

The value of ferrites as materials for high-frequency applications continues to grow.

In the 1940s–1950s, ferrites for RF applications were systematically studied by **J. L. Snoek** [98] of Philips for applications in devices that send, receive, and manipulate electromagnetic signals at RF frequencies as well as for power conversion and conditioning applications.

Snoek, worked primarily on refining the process conditions for optimized performance of high-frequency spinels, developed products under the tradename *Ferroxcube* (c. 1941, type I CuZn-ferrite, type II MgZn-ferrite, type III MnZn-ferrite, type IV NiZn-ferrite, and type V Mn-ferrite). These product lines allowed enhanced performance and reduced form factors for communication coils, transformers, flyback transformers, and deflection yokes. MnZn ferrites, operating from 1 kHz to 1 MHz, profoundly influenced international markets of these technologies and emerging commercial electronics.

Although he lived a relatively short life, dying as a result of an automobile crash in 1950 at the age of 48, perhaps no one had advanced ferrite processing and product development more in fewer years of active research than J. L. Snoek.

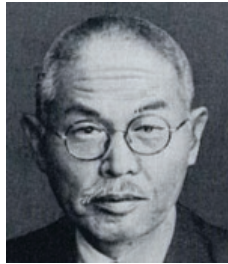
From 1970 to 2000, led by experts such as **Alex Goldman** of Spang & Company, new spinel ferrite compositions, having permeabilities as high as 20 000 to 30 000, were developed to be used at frequencies up to and exceeding 1 MHz [128].

Today, ferrites play an essential role in modern society. Nearly every consumer product has one or more ferrites as embedded components. And nearly all future products considered today are anticipated to contain ferrites.

Major technological market growth of ferrites is enabled by the development of high-quality crystals (by several techniques), polycrystalline compacts (including LTCC), films (as pulsed laser deposition [PLD], spin-spray, liquid-phase epitaxy [LPE], sputtering, and molecular beam epitaxy [MBE]), and nanoparticles (by a large variety of techniques reviewed by Willard et al. [156]).

Many of these breakthroughs and major developments in magnetism during the twenty-first century are given in Table 1.1. Technologies range from magnetic storage, medical (as MRI contrast agents and magnetic hyperthermia), power generation, conditioning, and conversion, RF and millimeter-wave devices for the sending, receiving, and manipulating electromagnetic signals for communication, radar, and sensing.

**Table 1.1** History of key technological and theoretical developments in twentieth-century magnetism.

Era	Major events	Development and key citations	Inventor
<b>1900</b>			
1895	<b>Hendrik Lorentz</b> developed the Lorentz law, local time, time dilation, Lorentz transformation, and other concepts.	[45–50]	Hendrik Lorentz
1897	<b>John J. Thomson</b> postulated moving charged particles increase in mass. He suggested that cathode rays are composed of very light, negatively charged particles labeled electrons.	[59–62]	J. J. Thomson
1906	<b>Pierre Curie</b> established thermal response of ferromagnetic materials defining Curie temperature ( $T_C$ ). $T_C$ is used to study ferromagnetic materials and plate tectonics, treat hypothermia, and understand extraterrestrial magnetic fields, among other effects.	[52]	Pierre Curie
1907	<b>Pierre Weiss</b> developed the domain theory of ferromagnetism, Weiss domains, the Weiss mean-field theory, and the Curie–Weiss law, among other key concepts.	[53]	Pierre Weiss
1909	<b>Hilpert</b> proposed that iron oxide–based materials are suitable for electric machine applications.	[90]	S. Hilpert
1917	<b>Kotaro Honda</b> invented KS steel that is 0.4–0.8% carbon, 30–40% cobalt, 5–9% tungsten, and 1.5–3% chromium.	KS steel is a permanent magnetic steel with three times the magnetic resistance of tungsten steel. [91, 92]	 Kotaro Honda
1924–1925	Quantum spin was proposed to double the available electron states by introducing two-valued “hidden rotation.”	<b>George Uhlenbeck</b> and <b>Samuel Goudsmit</b> suggested the physical interpretation of the electron spinning on its axis [65]. <b>Wolfgang Pauli</b> developed the underlying theory in 1924–1925 [66].	Wolfgang Pauli

(Continued)

**Table 1.1** (Continued)

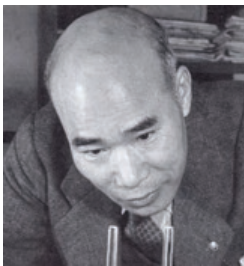







Era	Major events	Development and key citations	Inventor
<b>1900</b>			
1931	Metallurgist <b>Tokushichi Mishima</b> invented MK steel. MK steel is regarded as the precursor to AlNiCo magnets. AlNiCo magnets are regarded as the first generation of permanent magnets.	Mishima discovered that aluminum restores magnetism to nonmagnetic nickel steel. MK steel, which is an extremely inexpensive magnetic alloy, is closely related to modern AlNiCo magnets [92].	 <p data-bbox="1161 600 1349 621">Tokushichi Mishima</p>
1933–1934	<b>Kotaro Honda</b> invented NKS steel whose magnetic resistance is several times that of KS steel.	[91, 92]	
1935 (1946)	<b>Lev Landau</b> and <b>Evgeny Lifshitz</b> predicted the existence of ferromagnetic resonance (FMR) of the Larmor precession [93].	FMR was verified experimentally by Griffiths [94].	 <p data-bbox="1161 972 1268 993">Lev Landau</p>
			 <p data-bbox="1161 1266 1300 1287">Evgeny Lifshitz</p>
1935	Tokyo Denki Kagaku Kogyo (now <b>TDK Corporation</b> ) was founded to commercialize ferrite cores of <b>Yogoro Kato</b> and <b>Takeshi Takei</b> .	Establishment of TDK Corporation and commercial products of spinel ferrites as “Oxide Core” products [95, 96].	 <p data-bbox="1161 1566 1284 1587">Takeshi Takei</p>  <p data-bbox="1161 1871 1268 1892">Yogoro Kato</p>

Table 1.1 (Continued)

Era	Major events	Development and key citations	Inventor
<b>1900</b>			
1938	Crystal structure of magnetoplumbite (M-type, hexaferrite) was determined to be hexagonal with the composition $\text{PbFe}_{7.5}\text{Mn}_{3.5}\text{Al}_{0.5}\text{Ti}_{0.5}\text{O}_{19}$ .	Structure of magnetoplumbite hexaferrite was determined [97].	
1946	Patent and commercialization of <i>Ferroxcube</i> at Philips Company led by <b>J. L. Snoek</b> . Development of $\text{AB}_2\text{O}_4$ -spinel ferrites (A = Zn,Fe,Mn; B = Cu,Mg,Ni) resulted in size reduction of inductor cores.	Patent submitted on Ni-Fe oxide ferrite developed under tradename <i>Ferroxcube</i> . [98] J. L. Snoek made several key contributions to the development of spinel and hexaferrite magnetism and products during the course of his life.	 J.L. Snoek
1949	Tensor notation of permeability was proposed by <b>Dirk Polder</b> as necessary for ferrites as ferrimagnetic materials are shown to become anisotropic in the presence of magnetizing fields.	Introduction of the Polder tensor was essential to understanding ferrite gyromagnetic properties [99].	 Dirk Polder
1948	Theoretical studies of magnetic properties of ferrites by <b>Néel</b> , following <b>Kramer's</b> postulate on antiferromagnetism, led to the discovery of ferrimagnetism and a broader understanding of ferrite magnetism based on multiple molecular fields.	Superexchange theory was first proposed [100–102].	 Hans Kramer  Louis Eugène Néel

(Continued)

Table 1.1 (Continued)





Era	Major events	Development and key citations	Inventor
<b>1900</b>			
1950	Superexchange theory was developed in a robust mathematical framework by <b>Phillip Anderson</b> .		 Phillip Anderson
1950–1952	Commercialization of <i>Ferroxdure</i> by Philips Company was carried out.	Development of first hexaferrite commercial products [103]	
1950	Industrial activity leads to the growth of the power ferrite industry stemming from demand for mass production of large-scale deflection yokes for the burgeoning TV market.	Introduction of power ferrites	
1951	First neutron scattering studies of ferrites. <b>Clifford G. Shull</b> shared in 1994 the Nobel Prize in Physics with <b>Bertram Brockhouse</b> for the development of neutron scattering.	Neutron scattering was applied to ferrites by Shull et al. [104].	 Clifford G. Shull
1952	NiZn and MnZn ferrites for use in TV tube deflection yokes and high-voltage flyback transformers.	Philips Company advanced commercialization of spinel ferrites.	
1953	<b>George Rado</b> described frequency spectra of ferrites and identifies resonances related to domain wall and spin dynamics.	Domain wall and spin resonances explained [105].	
Circa 1955	<b>John B. Goodenough–Junjiro Kanimori</b> proposed rules for superexchange that provided guidelines related to local bonding angles, covalency, and ligand field theory for Me–O–Me molecular triad arrangements.	[106–108]	 John B. Goodenough  Junjiro Kanimori

Table 1.1 (Continued)

Era	Major events	Development and key citations	Inventor
<b>1900</b>			
1956	Nonlinear behavior in ferrites at high microwave power established by <b>Suhl</b> .	[109]	 Harry Suhl
1956–1957	First development of magnetic garnets.	Bertaut and Forrat [110] Geller and Gilleo [111]	
1957	First demonstration of nanoparticle ferrites for cancer therapeutics by magnetic hyperthermia. Later developed for human trials circa 2010.	Gilchrist et al. [112]	
1960–1961	First single crystal magnetic garnets processed.	Van Hook [113] Rudness and Kebler [114]	
1967	Mössbauer spectroscopy was employed to describe the magnetic and nuclear structures of spinel $\text{NiFe}_2\text{O}_4$ (and $\text{Ni}(\text{Cr})\text{Fe}_2\text{O}_4$ ) as a collinear Néel ferrimagnet. <b>Rudolf Ludwig Mössbauer</b> shared in 1961 the Nobel Prize in Physics with Robert Hofstadter. Mössbauer was cited for his “...research concerning the resonance absorption of gamma radiation and his discovery in this connection of the effect which bears his name.”	Chappert and Frenkel [115]	 Rudolf Ludwig Mössbauer Karl Strnat and Alden Ray
1967–1972	<b>Karl Strnat</b> and <b>Alden Ray</b> developed $\text{SmCo}_5$ and $\text{Sm}_2\text{Co}_{17}$ alloys having then record energy products and excellent high-temperature performance.	[116–118]	
1970	Nobel Prize in Physics was awarded to <b>Louis Eugène Néel</b> and Hannes Olof Gösta Alfvén. Néel was cited for his “...fundamental work and discoveries concerning antiferromagnetism and ferrimagnetism...”	Noble Prize, 1970.	Louis Eugène Néel
1970–1972	Microwave relaxation mechanisms in ferrites is advanced by <b>Carl Patton</b> .	Carl Patton [119].	 Carl Patton

(Continued)

Table 1.1 (Continued)








Era	Major events	Development and key citations	Inventor
<b>1900</b>			
1963, 1969	Pure phases BiFeO <sub>3</sub> determined to be a multiferroic material later proven to be of great research interest and potential value.	[120–122]	S.V. Kiselev et al. J.R. Teague et al.
Circa 1972–1978	Ferrite-based magnetoelectric heterostructures/composites of magnetostrictive ferrites and piezoelectrics.	Initial magnetoelectric heterostructures reported [123, 124].	J. Van Suchetelene J. Van den Boomgaard and R.A.J. Born
1971, 1972	Liquid-phase epitaxy (LPE) growth of garnet thick films demonstrated.	LPE growth of ferrites by Blank, Glass, and others demonstrated. [125–127]	
1976, 1977	LPE growth of spinel ferrite thick films demonstrated.		
1978	LPE growth of hexaferrite thick films demonstrated.		
1975	RF applications of spinels demonstrated and advanced.	<b>Goldman</b> advanced NiZn ferrites and others for RF applications at Spang & Company [128].	 Alex Goldman
1977	Nobel Prize in Physics awarded to <b>Phillip W. Anderson, Melville Mott, and John van Vleck</b> . van Vleck has been called the Father of Quantum Magnetism.	Nobel Prize (1977) [129]	 John Van Vleck
1981–1982	Aqueous coprecipitation of ferrite nanoparticles demonstrated.	Massart [130, 131]	R. Massart
1982	Barium hexaferrite thin film–based perpendicular recording media proposed by <b>Hideo Fujiwara</b> .	Ferrite-based perpendicular high-density storage media proposed by Hideo Fujiwara [132]	 Hideo Fujiwara





Table 1.1 (Continued)


Era	Major events	Development and key citations	Inventor
<b>1900</b>			
1982–1984	The high costs of raw materials for SmCo permanent magnets led to the development of Nd <sub>2</sub> Fe <sub>14</sub> B by Sumitomo, NRL, and General Motors (GM) and others. Patent conflicts throughout the 1980s–1990s resulted in priority granted to Sagawa of Sumitomo and Koon of the US Naval Research Laboratory. Today, China maintains a near-global monopoly over rare earth ores and finished magnets due to having the largest mines and processing plants.	[133, 134]	 Masato Sagawa
			 Norman C. Koon
Circa 1989	Barium hexaferrite thin film–based longitudinal recording media proposed by <b>Dennis Speliotis</b> .	Longitudinal high-density storage media proposed by Speliotis [135].	 Dennis E. Speliotis
Circa 1990–1995	Ferrites are used as bead line filters for many commercial electronics.	Bead line filters commercialized.	
1980–2000	Research into the synthesis and characterization of nanostructured ferrite systems and nanoparticles grew rapidly.	Nanostructured ferrite systems rapidly developed.	
Circa 1980	Investigation of physics and devices of spinwave solitons in ferromagnetic films.	[136]	 Boris Kalinikos

(Continued)

Table 1.1 (Continued)

Era	Major events	Development and key citations	Inventor
<b>1900</b>			
1988	Giant magnetoresistance effect discovered near simultaneously by <b>Albert Fert</b> and <b>Peter Grünberg</b> .	[136, 137]	 <p>Albert Fert</p>  <p>Peter Grünberg</p>
1997	GMR hard disk head introduced by IBM.	[138]	
2007	The Nobel Prize in Physics awarded to <b>Albert Fert</b> and <b>Peter Grünberg</b> for discovery of GMR.	[129]	
<b>2000</b>			
Circa 2000	Ferrite nanoparticles developed for biomedical applications.	Ferrite NPs for use in cellular therapy such as cell labeling, targeting, and a tool for cell biology research to separate and purify cell populations; tissue repair; drug delivery; magnetic resonance imaging (MRI); hyperthermia; magnetofection; etc. [139].	
Circa 2010	Ferrite nanoparticles developed for theragnostics applications.	Ferrite NPs used for concomitant multifunctional biomedical diagnostic-therapeutic treatments [140].	
Circa 2000	First demonstration of laminated piezoelectric–ferrite magnetoelectric heterostructures.	[141]	
Circa 2010–present	Magnonics: The study of physics and devices based on quantum spin dynamics.	Brillouin light scattering experiments in 1966 confirmed the existence of spin waves with nonzero wave vectors [142]. YIG magnonics of the last two decades discussed the transferability of concepts and ideas learned in ferrite materials to modern nanoscale systems [143].	

**Table 1.1** (Continued)

Era	Major events	Development and key citations	Inventor
<b>1900</b>			
2000–present	Room temperature, low-field magnetoelectric response reported in long-wavelength hexaferrite magnetic structures.	Magnetoelectric effect in hexaferrites by Kimura [144].	
2000–present	Research and applications focus on microwave ferrites, magneto-optical properties, ferrite films, nanostructured ferrites, and low temperature co-fired ceramics (LTCC) multilayer chip inductors.	Microwave and magneto-optical applications pursued.	
2000–present	Development of self-biased hexaferrites.	Self-biased hexaferrite ceramics developed by Harris et al. [145–149].	
Circa 2008–present	Harris et al. demonstrated successful integration of ferrite with semiconductor substrates (SiC and GaN) for integrated system-on-a-wafer technology.	Underlying materials science for integrated system-on-a-wafer technology demonstrated.	
2010–present	Demonstration of self-biased microstrip circulators and commercialization.	Self-biased devices demonstrated and commercialized [150, 151].	
2016–present	Metamagnetics Inc. commercialized the first broadband passive frequency-selective limiters as <i>Autotune filters</i> .	<i>Autotune Filters</i> products introduced based on original designs of John Douglas Adam [152, 153].	
2012, 2020	Ultra-high coercive fields measured in $\epsilon$ -Fe <sub>2</sub> O <sub>3</sub> and hexaferrites with FMR frequencies occurring beyond 240 GHz.	Namai et al. – $\epsilon$ -Fe <sub>2</sub> O <sub>3</sub> [154] Gorbachev et al. – hexaferrites [155]	John Douglas Adam
Twenty-first-century advances	Advances in the synthesis and characterization of ferrites continues. Research focuses on crystal growth, thin films, nanoparticles, nanostructured ferrites, ferrite-based composites, and multilayer chips inductors, ferrofluids, Spintronics, soft magnetic materials, magneto-optics, EMI suppression and shielding, as well as EVs, energy, biomedical, environmental applications, and RF technologies.	Materials for RF, sub-millimeter wavelength, and optical applications continue to grow. New fields of Spintronics and magnetoelectronics emerge and evolve. Medical application of multifunctional NPs continues to develop.	

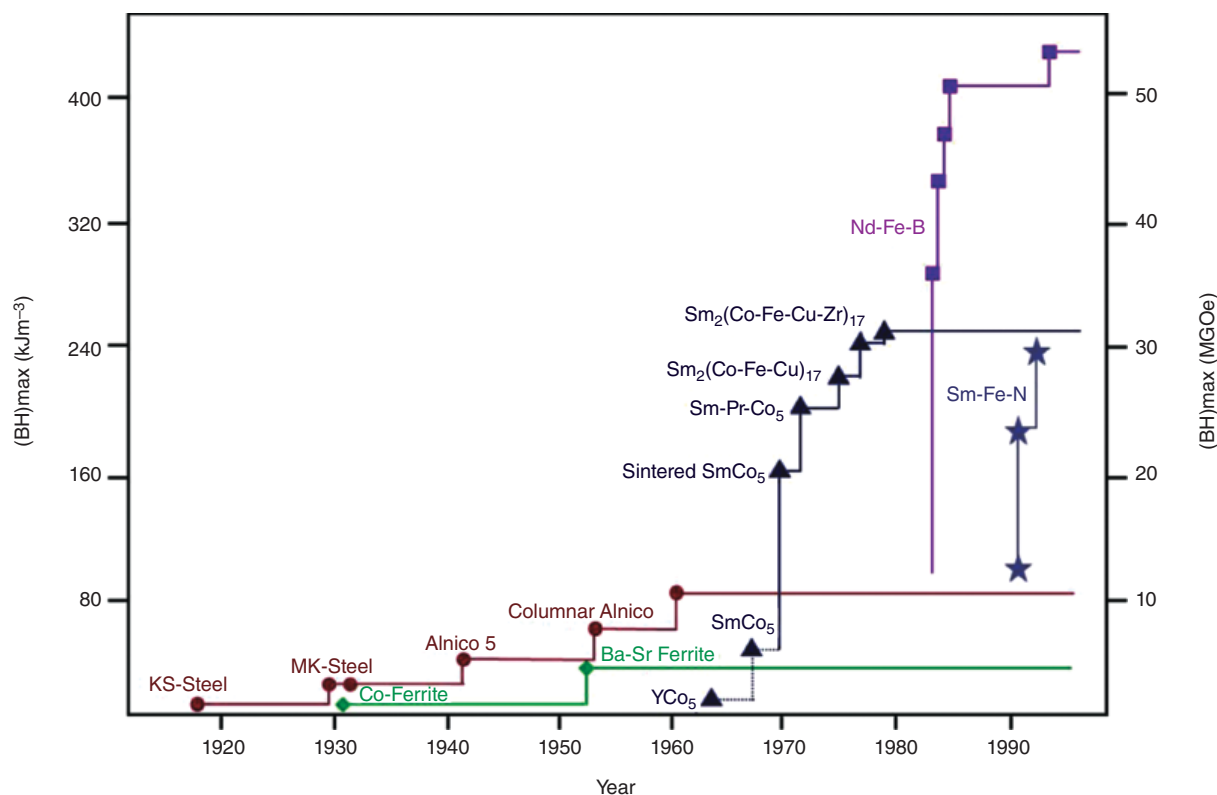
### 1.4.9 Development of Permanent Magnet Materials (1920–2020) [157]

#### 1.4.9.1 AlNiCo Magnets

Figure 1.4 provides a graphic detailing the evolution over time of the magnetic energy product from early in the twentieth century to near the present day. As one sees, AlNiCo magnets were developed in the 1930s by metallurgist **Tokushichi Mishima** as MK steel. Their properties derive from shape anisotropy associated with its microstructure that consists of ferromagnetic Fe–Co needles in an Al–Ni matrix. Due to their high Curie temperature  $\sim 850$  °C, they remain useful even today for many high-temperature applications [157].

#### 1.4.9.2 Hard Ferrite Magnets

Circa 1950, barium hexaferrite (BaO•6Fe<sub>2</sub>O<sub>3</sub>) of the magnetoplumbite structure was discovered at Philips Physics Laboratory and advertised under the tradename *Ferroxdure*, was shown to have both a high coercivity (i.e.  $\sim 170$  kA/m) and low materials cost relative to metallic permanent magnets.



**Figure 1.4** Historical evolution of permanent magnet maximum energy product from 1900 to 2000 [157].

Soon after, Philips developed strontium hexaferrite (i.e.  $\text{SrO} \cdot 6\text{Fe}_2\text{O}_3$ ), with superior properties to barium hexaferrite. Barium and strontium hexaferrites have since dominated the market for hard ferrites due their excellent performance and low costs [103].

Hexaferrites exist in several structural variants, i.e. M, Y, Z, X, U, and W, in which the metal and oxygen stoichiometries vary. In comparison to the cubic crystalline ferrites, crystal symmetry breaking in the hexaferrites provide them with extremely high magnetic anisotropy fields, allowing for the shifting of ferromagnetic resonance (FMR) frequency leading to the operation of hexaferrites at microwave, millimeter-wave, quasi-optical frequencies [158, 159].

#### 1.4.9.3 Samarium–Cobalt

Developed in the late 1960s, this group of alloys combine cobalt, iron, and samarium. Samarium–cobalt magnets are available in two phases, namely,  $\text{SmCo}_5$  and  $\text{Sm}_2\text{Co}_{17}$ , with  $\text{SmCo}_5$  the phase of greatest proven commercial value. Samarium–cobalt magnets have maximum energy products, which relate to the magnetic flux output per unit volume, ranging from 14 to 33  $\text{MG} \cdot \text{Oe}$ , i.e. approximately 112 to 264  $\text{kJ/m}^3$ ; their theoretical limit is 34  $\text{MG} \cdot \text{Oe}$ , or 272  $\text{kJ/m}^3$ . Additionally, these alloys have a strong resistance to corrosion and oxidation and can be widely used in high-temperature environments [116–118].

#### 1.4.9.4 NdFeB

$\text{Nd}_2\text{Fe}_{14}\text{B}$ , and variations of the same, are today's strongest magnets by far with theoretical magnetic energy products of  $BH_{\text{max}} \approx 512 \text{ kJ/m}^3$ , or 64  $\text{MG} \cdot \text{Oe}$ , and those readily available for commercial sale at 52  $\text{MG} \cdot \text{Oe}$ .

The strength of neodymium magnets results in part from their tetragonal crystal structure that contributes high magnetocrystalline anisotropy.  $H_A$  provides an internal resistance to the turning of its magnetization vector and gives rise to high coercivity, which directly affects the energy product. The neodymium atom has a large dipole moment due to its four unpaired electrons providing  $\text{Nd}_2\text{Fe}_{14}\text{B}$  a high-saturation magnetization ( $J_s \approx 1.6 \text{ T}$  or 16 kG) and a remnant magnetization of typically 1.3 T. Its maximum energy density, or potential for storing amounts of magnetic energy, is 64  $\text{MG} \cdot \text{Oe}$  [160].

It has been widely reported since their inception that **Croat** of General Motors (GM) and **Sagawa** of Sumitomo Special Metals independently discovered the  $\text{Nd}_2\text{Fe}_{14}\text{B}$  compound almost simultaneously in 1984 [161]. However, as established in

later years through careful historical investigation, the earliest US research was carried out (and patented) by **Norman Koon** at the US Naval Research Laboratory (NRL) [162, 163]. In the late 1990s, appellate courts found in favor of companies who licensed Koon's patent in litigation initiated by GM. Immediately thereafter, GM sold their Nd<sub>2</sub>Fe<sub>14</sub>B company, Magnequench, and abandoned the permanent magnet market.

A carefully researched account of the history of permanent magnet materials by Stadmaier is presented in [164]. There, he attributes the key to expanding the scope of binary rare earth–Fe-based metallic magnetic alloy research with the addition of boron to Koon. Further, Stadmaier states:

At Sumitomo the evidence for the idea of the boron addition also points to Koon, as already noted earlier [165]. Here a Japanese patent application [166] was entered before the priority patent application [167] and might have established an earlier priority date on composition but was too obviously derivative from Koon and Das [134] and not mentioned at all in Ref. [167]. The application [166] involves melt spinning as a process and is simply an improvement on the rapidly solidified material of Koon and Das [134]. The composition of a ternary rare earth alloy was given in Ref. [134] as (Fe<sub>0.82</sub>B<sub>0.18</sub>)<sub>0.9</sub>Tb<sub>0.05</sub>La<sub>0.05</sub>, and it had an undesirable constricted hysteresis loop. In the Japanese application Ref. [166], two compositions appear in formulations that are obviously derived from Koon, namely (Fe<sub>0.82</sub>B<sub>0.18</sub>)<sub>0.9</sub>(La<sub>0.4</sub>Pr<sub>0.3</sub>Tb<sub>0.3</sub>)<sub>0.1</sub> and (Fe<sub>0.82</sub>B<sub>0.18</sub>)<sub>0.9</sub>(La<sub>0.4</sub>Pr<sub>0.3</sub>Nd<sub>0.3</sub>)<sub>0.1</sub> now having proper loops. [164]

Based on these reports, it appears that Koon is a principal, if not *the*, principal inventor of NdFeB magnets. Commercially, GM successfully advanced the development of melt-spun nanocrystalline Nd<sub>2</sub>Fe<sub>14</sub>B magnets, and Sumitomo developed full-density sintered Nd<sub>2</sub>Fe<sub>14</sub>B magnets. As Koon lamented before his tragic death that occurred in a skiing accident circa 1997 as the cause for GM's success in establishing their fame as inventors of NdFeB: "...the Navy's State school law team were no match for GM's Armani-clad, Harvard law team" [168].

For far too long, Koon has been denied proper credit for his brilliant discovery that has changed modern technology.

#### 1.4.10 Spin-Based Electronics: Spintronics

In contrast to traditional electronics, where charge carriers, as holes and electrons, are integrated in solid-state devices such as diodes and transistors, *spintronics* is based on the measurement of spin-dependent electron transport. The origins of spintronics can be traced to the early experiments of spin-polarized tunneling between ferromagnetic metals by Julliere [169] and those of superconductors into metals by Meservey and Tedrow [170]. In 1985, Johnson and Silsbee demonstrated spin-polarized electron injection from a ferromagnetic metal into a normal metal [171] while the use of semiconductors was first proposed by Datta and Das in 1990 for a novel spin field-effect transistor [172]. Famously, in 1988, Fert et al. [137] and Grünberg et al. [173] discovered giant magnetoresistance (GMR) that paved the way for GMR principles of spin-dependent devices.

Sinova and Žutić recently reviewed the field of spintronics and identified five emerging subfields that uniquely build upon the quantum discoveries and engineering breakthroughs of the last 75 years; these include: spin-transfer torque (STT), spin Hall effect (SHE), spin caloritronics (SCAL), silicon-based spintronics (SBS), and graphene + topological insulators (G+TI) [174].

##### 1.4.10.1 Spin-Transfer Torque (STT) Technologies

STT has its origins in the conservation of angular momentum [175–177] and is a key component enabling next-generation magnetic random-access memories (MRAM), logic-in-memory architectures, and high-density memory devices [178].

##### 1.4.10.2 SHE Technologies

SHE originates from relativistic spin–orbit coupling (SOC) interactions predicted by Dyakonov and Perel in 1971 [179, 180]. It manifests as the appearance of spin accumulation on lateral surfaces of an electric current-carrying sample, the signs of the spin directions being opposite on opposing boundaries [181]. SHE has been used to create spin FETs [182], as a means to measure spin currents [183], and to generate spin currents large enough to produce STT effects [184].

##### 1.4.10.3 SCAL Technologies

Discovery of the spin Seebeck effect [185], thermal gradients driving heat currents are now being exploited to generate spin currents. The origin of the effect appears to be rooted in the coupling between collective spin modes and lattice excitations, i.e. magnon–phonon coupling [174].

#### 1.4.10.4 Silicon-Based Spintronics

Silicon has been for decades the central material for twenty-first century electronics but due to its indirect bandgap and weak SOC has not been pursued for spin injection and detection. However, recent breakthroughs [186, 187] have demonstrated that spin injection efficiency and coherence can be maintained over large distances calling for a reexamination of its value for many spintronic applications.

#### 1.4.10.5 G+TI Technologies

The spintronic aspects of  $G+TI$  originate from band-structure properties that create an effective topological *knot* that protects the surface and/or interfacial states from nonmagnetic disorder effects, making them promising materials as spin generators (TIs) or spin conductors (graphene) [188]. Topological insulators can also be coupled with superconductors, enabling error-protected quantum computation [189].

#### 1.4.11 Spin Wave–Based Electronics: Magnonics

*Magnonics* is based upon long-range transport of quantized spin waves called magnons [190–192]. To realize practical magnonics devices, high-crystal-quality ultralow Gilbert damping ferrite materials are required. Yttrium iron garnet has been shown to be the only acceptable magnonics media at the time of this publication [193, 194].

Spin waves manifest as either short-wavelength exchange spin waves or long-wavelength magnetostatic waves. To date, magnon signal processing devices operate upon magnetostatic waves that can be readily excited and detected by inductive antennas or simple transducers. More recently, research has focused on exchange waves that allow for implementation as nanostructured devices.

To achieve the integration of magnonics, devices with electronic circuitry require exciting magnons by charge currents. This has been demonstrated by STT-induced spin precession [195, 196]. An alternative approach involves the creation of spin-polarized currents from SHE spin-dependent scattering of electrons in a nonmagnetic metal or semiconductor having large SOC.

Based on these recent advances, magnonics-based THz communication technologies in support of 6G wireless communications and beyond is under consideration [197]. Such breakthroughs would require new low-Gilbert-loss ferrites that would enable long-distance magnonics currents to be realized and concomitantly allow decreases in energy consumption.

In summary, magnonics is a burgeoning field of spintronics that is anticipated to deliver breakthroughs enabling advances in computation, data processing, and communication in the coming years.

This brings us to the present-day developments on magnetism and magnetic materials. Of course, inevitably, several, if not many, important discoveries have been omitted due to space constraints or the ever rapidly development of new technologies occurring presently. Omissions are inadvertent.

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