FIP10604 – Text 15 –
<div align="center">

MAGNETIC ORDER: MACROSCOPIC CHARACTERISTICS
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So far, we have studied magnetism on **ideal** systems, uniform and infinite. We included crystal-field anisotropy, and took into account exchange interactions for localized moments, or explicitly the Coulomb interaction between electrons in the case of itinerant moments, but we left aside dipolar interactions.

If such an ideal system existed, below the Curie temperature (for simplicity, we will refer only to FM order) it would be **uniformly** magnetized, and its response to an applied external field would be perfectly **reversible**. This does not happen in **real** ferromagnetic solids.

Figure 1 shows a typical schematic curve $M(H)$ for a sample of ferromagnetic material. Notice that initially the magnetization is null at zero field, even though the system is ferromagnetic and well below the Curie temperature (this initial state depends on the system's history). The first part (inner curve on the first quadrant) is called *initial branch*, starting with a slope that defines the *initial susceptibility* χ_0, and reaching the saturation magnetization M_s. From there, an **irreversibility** of the $M(H)$ curve is observed, with the other branches completing a **hysteresis** cycle. M_r is the *remanent* magnetization or simply **remanence**, and H_c is the *coercive field* or **coercivity**.

It is also common to plot hysteresis curves as B vs H instead of M vs H. Such curves can be converted into one another in view of the relationship between the three quantities. Magnetic hysteresis can be understood as a consequence of formation of **magnetic domains** and their response to the applied field, as we will discuss in the following.
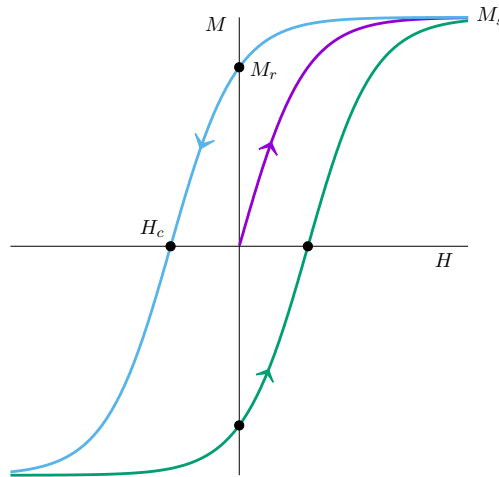
Figure 1: Schematic representation of a typical $M(H)$ hysteresis curve for a ferromagnetic material, showing the remanence (M_r) and the coercivity (H_c).
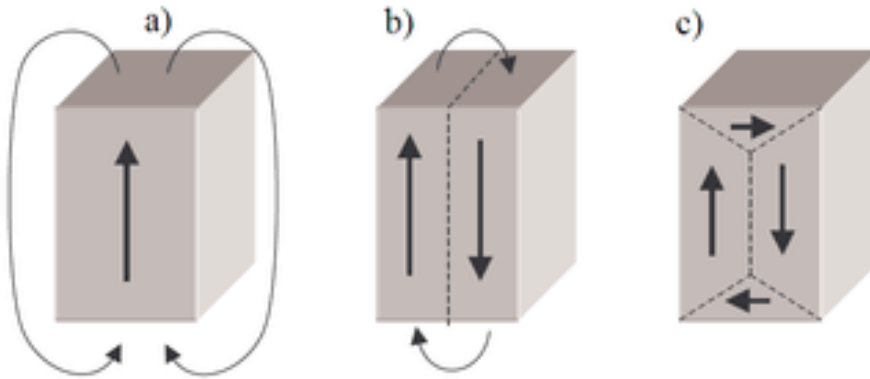
Figure 2: Scheme of changes in the magnetic-field lines by subdivision of a ferromagnetic sample in domains. Internal arrows indicate domain magnetizations.
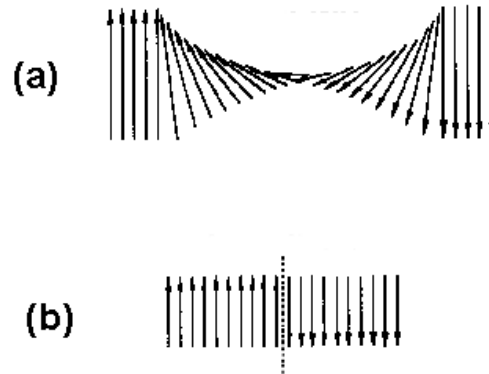
Magnetic domains

A magnetized sample generates a magnetic field. This field can be seen as the sum of fields from different (macroscopic) regions of the sample, which are magnetically ordered due to exchange interactions. Inside the sample, if the magnetization is uniform, different regions are oriented in a way that is energetically unfavorable with respect to the field created by other regions. This is due to the magnetic dipolar interaction, which we previously neglected in comparison to exchange interactions at the microscopic level, but which have a significant effect when considering macroscopic moments associated to large volumes within the material.

As seen in Text 05, the dipolar interaction favors parallel ordering of magnetic moments along a line connecting them but antiparallel if oriented perpendicularly to that line. Fig. 2 illustrates the fact that the magnetostatic energy is reduced if the system is subdivided into *domains* with opposite magnetization. However, the number of domains cannot increase indefinitely, because there must be a balance between the magnetostatic (dipolar) energy, reduced by the formation of domains, and the energies of exchange and anisotropy, which increase upon creation of domain walls. Depending on the relative importance of these latter two interactions, we have the situations described below.

Strong exchange interaction – Microscopic moments change orientation smoothly, resulting in a wide domain wall. It basically costs anisotropy energy, since magnetic moments form unfavorable angles with the anisotropy axis throughout the wall, while neighboring moments are almost parallel [Fig. 3 **(a)**].

Strong anisotropy – Microscopic moments tend to remain aligned with the anisotropy axis, resulting in a narrow wall. The energy cost is essentially of exchange, since moments on opposite sides of the domain wall become antiparallel [Fig. 3 **(b)**].

Figure 3: Schematic figures representing how magnetic moments reverse their orientation across a wide (a) or narrow (b) domain wall.

The number of domain walls is determined by a balance between reduction of magnetostatic energy and cost in anisotropy/exchange energy. It clearly depends on the sample size. Another important factor appears when the sample is polycrystalline, which is the most common situation. In this case the system is composed of *grains* that, in general, have anisotropy axes with different orientations, and whose interfaces (grain boundaries) are quite disordered. Depending on the grain size, we have a single domain within it or more than one. We will return to this point later.

The existence of domains explains the fact that we can have a null global magnetization although the system is in the FM phase. The remaining details of the $M(H)$ curve will be discussed later.

Types of anisotropy

Considering the important role played by anisotropy in the formation of magnetic domains, it is important to review and complement what we know about it. We have essentially three types of anisotropy, depending on its origin.

1. **Magneto-crystalline anisotropy** – We have already studied its microscopic origin, the combined effect of crystal-field and spin-orbit interaction to generate anisotropy in spin space. Macroscopic approaches usually involve a phenomenological free energy dependent on the angles between magnetization and symmetry axes.

2. **Magnetoelastic anisotropy -** It is due to the phenomenon of *magnetostriction*, i.e., strains or stresses in response to applied magnetic fields; or, conversely here, the effect of these strains and stresses on the magnetic interactions. From a microscopic point of view, it has its origin also in the spin-orbit interaction.

3. **Shape anisotropy -** It is due to the *demagnetizing field*, which originates from dipolar interactions (see below) and depends on sample shape and size.

Demagnetizing field

The *demagnetizing field* is defined as the difference between the magnetic field existing within the material and the applied (external) magnetic field. To understand this concept, let us consider a simple case: a film, i.e., an infinite system in two of its dimensions and finite in the third. Assuming that the film is uniformly magnetized in the presence of an applied field $\mathbf{H}_a$ perpendicular to its plane, we have the situation sketched in Fig. 4.

$\mathbf{H}_a$ is the applied field; $\mathbf{B}_o$ is the magnetic induction outside the material; $\mathbf{M}$, $\mathbf{B}_i$ and $\mathbf{H}_i$ are the three magnetic vectors within the material, and $\mathbf{H}_d$ is the demagnetizing field. Starting with the basic (SI) relations

$$\mathbf{B} = \boldsymbol{\mu}_0(\mathbf{H} + \mathbf{M}) \,, \qquad \nabla \cdot \mathbf{B} = 0 \,, \tag{1}$$

we have

$$\mathbf{B}_o = \mathbf{B}_i \quad \Rightarrow \quad \mathbf{H}_a = \mathbf{H}_i + \mathbf{M} \,. \tag{2}$$

Therefore,

$$\mathbf{H}_i = \mathbf{H}_a - \mathbf{M} \quad \Rightarrow \quad \mathbf{H}_d \equiv \mathbf{H}_i - \mathbf{H}_a = -\mathbf{M} \,. \tag{3}$$

The $+$ and $-$ signs in Fig. 4 are a representation of "magnetic charges" attributed to the poles (respectively, N and S) of a magnet (magnetized object). In this type of representation, lines of $\mathbf{H}$ begin in positive "charges" and end in negative ones, unlike the lines of $\mathbf{B}$, which are always continuous. The demagnetizing field can be seen as the magnetic field created by these charges inside the material.

In general, for a system with **uniform** magnetization $\mathbf{M}$, we have

$$\mathbf{H}_d = -N_d\mathbf{M} \,, \tag{4}$$
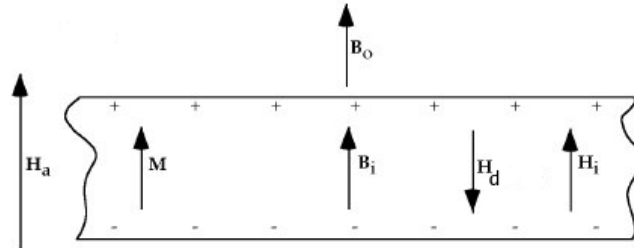
Figure 4: Scheme of internal and external magnetic vectors for a ferromagnetic film with perpendicular magnetization, including the *demagnetizing field* $\mathbf{H}_d$.

where N_d is the *demagnetization factor*. Thus, a film has $N_d = 1$ for perpendicular magnetization.

In general, one cannot define a single demagnetization factor for objects of arbitrary shape because the magnetization is not uniform. Objects in the form of a revolution **ellipsoid** may be uniformly magnetized, and the demagnetization factor can be calculated **exactly** (solving the field equations with the appropriate boundary conditions at the object's surface). Calling a, b and c the three orthogonal semi-axes of the ellipsoid (with a and b of the same length), one obtains the exact relation

$$N_d^a + N_d^b + N_d^c = 1 \ . \tag{5}$$

Strictly speaking, the relationship between demagnetizing field and magnetization should be written as

$$\mathbf{H}_d = -\mathbf{N}_d \cdot \mathbf{M} \ , \tag{6}$$

with the different demagnetization factors defining a tensor.

Some relevant geometries may be viewed as special cases of an ellipsoid:

- **Sphere -** The three semi-axes are equal, which results in a single demagnetization factor $N_d = 1/3$.

- **Large diameter disc -** We have an "ellipsoid " with $a = b \to \infty$ and c finite, which is equivalent to the film that we analyzed before. We then have $N_d^c \simeq 1$, $N_d^{a,b} \simeq 0$.

- **Long cylinder -** It can be viewed as an "ellipsoid" with $c \to \infty$ and $a = b$ finite, which leads to $N_d^c \simeq 0$, $N_d^a = N_d^b \simeq 1$.

Note the appearance of **shape anisotropy**: magnetization is favored along directions for which $N_d = 0$, e.g., parallel to the axis of a long cylinder or in the plane of a flat film.

In addition to shape anisotropy, the demagnetization factor is important in determining the initial susceptibility (as we will see later), and must be taken into account when extracting microscopic information about a system from magnetic measurements.

Hysteresis curve and irreversibilities

We have seen that the absence of a net magnetization in a macroscopic FM system is due to the formation of domains. We also discussed the energy balance for this in terms of magnetostatic energy, exchange interaction, and various types of anisotropy. But why is there a hysteresis curve?

A hysteresis indicates the presence of **irreversibilities**, which occur in the magnetization processes under the external field and relaxation when it is removed.

In a perfect crystal, domains naturally formed by the competing interactions yield zero net magnetization at zero field. Let us suppose that an external magnetic field is then applied,

and its intensity is slowly increased. Domains with favorably oriented magnetization grow, while those with other orientations shrink in size. We may also have rotation of the magnetization away from a possible anisotropy axis to align itself with the field when it becomes strong enough. If experimentally possible, the entire sample becomes a single domain, with all magnetic moments parallel to the field when saturation is reached. Now, reducing the field intensity, these processes should revert, and the zero-magnetization state should be recovered at zero field. For such a system, the $M(H)$ curve would be reversible (no hysteresis). This does not happen in real systems due to crystal-structure defects that cause *pinning* of domain walls, preventing their movement. Therefore, the ideal domain pattern is not recovered, and a non-zero magnetization remains when the field is turned off.

From the above analysis we see that the characteristics of natural magnets depend on their history, like the occurrence of variations in temperature and magnetic fields to which the rocks have been subjected.

Soft and *hard* magnets

In most cases, the grains of a polycrystalline sample develop independent domain structures. According to the size of a given grain, we have two different scenarios:

- **Large-size grain ⇒ multidomain (MD) -** When the field is applied, favorably oriented domains grow at the expense of size reduction of the others, which takes place by wall motion. When the magnetization approaches saturation, the grain has been reduced to a single domain. Then the final stage follows what happens in the next scenario.

- **Small grain size ⇒ single-domain (SD)** – The external field causes **rotation** of the magnetization from an initial orientation (defined by the anisotropy) towards alignment with the field.

The first process (domain growth) is, in general, easier, leading to a *soft* behavior, in which saturation is quickly reached. The second process (magnetization rotation) requires more energy, resulting in a *hard* behavior in which very intense fields are needed to approach saturation.

MD grains, being *soft*, tend to have low coercivity. The schematic behavior of coercivity as a function of the grain size (often called "diameter") is shown in Fig. 5, exemplifying with grain sizes in two iron compounds (notice that the length scales are quite different).

Two **new** situations appear:

Pseudo single domain (PSD) - It happens in the transition region between SD and MD, where a metastable SD configuration exists in a region preferably MD (and vice versa). The possibility of changing from one to the other by effect of the applied field can lead to low coercivity with high remanence.
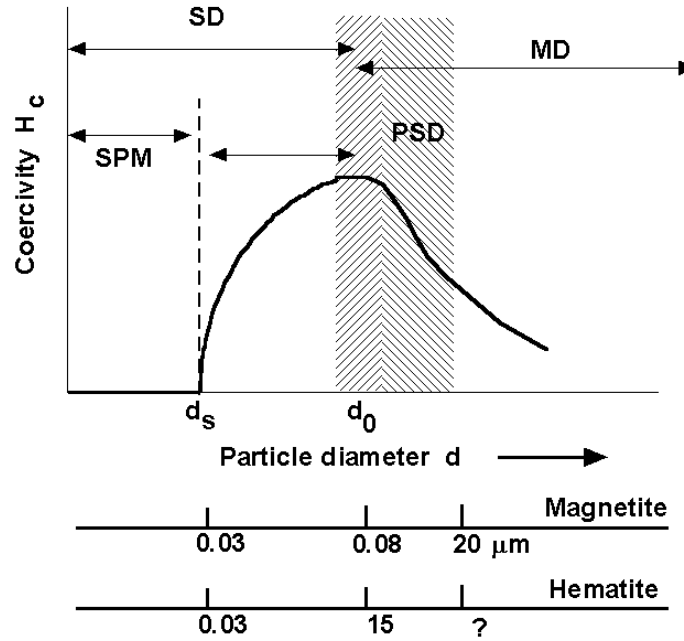
Figure 5: Behavior of the coercive field as a function of grain size, indicating the length scales for two naturally occurring iron compounds.

Super-paramagnetism (SPM) - When the grain is very small it tends to be roughly spherical. Then the anisotropy can be easily overcome by temperature, allowing the grain's (nearly saturated) magnetic moment to rotate instead of having a fixed orientation. It thus behaves as a (**very large**) paramagnetic moment. The leading role here is played by the dependence of relaxation time on anisotropy, grain volume, and temperature. The temperature dependence is exponential, producing large variations of relaxation time for not very large temperature changes. A reversible M vs H curve, characteristic of paramagnetic behavior, is observed for high temperatures (but still less than T_C), while a hysteresis is obtained at lower temperatures. The boundary between these two regimes defines a *blocking temperature*.

Initial susceptibility

On the initial branch of the M vs H curve (Fig. 1), we begin with a linear regime, which defines the initial susceptibility $\chi_0 = M/H$. Notice, however, that this is **not** a paramagnetic response since the moments are aligned inside domains. The magnetization processes involve, as we have seen, displacement of domain walls (rotation of the spins in these walls) or rotation of saturated single-domain magnetizations. Therefore, the initial susceptibility is closely related to the **transverse** component of the microscopic susceptibility in the ferromagnetic phase. Inside the material we must relate response, i.e., magnetization, with **internal** magnetic field. Then, in the linear regime one should have a relationship of the

type $\mathbf{M} = \chi_i \mathbf{H}_i$, where χ_i is an intrinsic susceptibility **of the material** and $\mathbf{H}_i$ is the internal magnetic field. To determine the initial susceptibility χ_0, which measures the sample's response to the **external** field, we must take into account the demagnetizing field. We have, therefore,

$$\chi_0 = \frac{M}{H} = \frac{\chi_i H_i}{H_i + N_d M} = \frac{\chi_i H_i}{H_i + N_d \chi_i H_i} ,\tag{7}$$

resulting in

$$\chi_0 = \frac{\chi_i}{1 + N_d \chi_i} .\tag{8}$$

Materials of high intrinsic susceptibility ("strong" ferromagnets) present $\chi_0 \simeq 1/N_d$. This does not occur, however, when the field is applied along a direction for which $N_d \simeq 0$, showing that the initial susceptibility is strongly dependent on sample geometry.

On the other hand, given that χ_0 is the measured quantity, we can invert Eq. (8) to obtain the intrinsic susceptibility as

$$\chi_i = \frac{\chi_0}{1 - N_d \chi_0} .\tag{9}$$

It is worth remarking that this last equation holds not only in relation to the initial susceptibility of a hysteresis cycle, but also to direct measurements of susceptibility in the case of a system that is not in the ordered phase.