## FIP10604 - Text 01 - INTRODUCTION

The first observations of magnetic phenomena date back to ancient Greece, in particular, the attraction of pieces of iron by certain stones which are natural magnets. These stones are generally iron oxides, like magnetite  $- \text{Fe}_3\text{O}_4$  (or FeO·Fe<sub>2</sub>O<sub>3</sub>). In the last centuries BC the Chinese knew about orientation of magnets with respect to Earth's geographical poles, and already around 1000 AD primitive compasses of floating assembly were being used.

A more quantitative description of magnetic phenomena began only in late 18th century, with the statement of a law of interaction between magnetic poles (Coulomb, 1795). Understanding of these phenomena began shortly after, in the first half of the 19th century, with magnetic fields being associated to electric currents (Oersted, 1820, Ampère, 1822; Faraday 1831), and culminating in the unification of electromagnetism (Maxwell, 1864).

Microscopic aspects of magnetism began to be addressed by the end of the 19th century, when diamagnetism, paramagnetism and ferromagnetism were described (Pierre Curie, 1883-5). About 20 years later, a classical statistical theory of paramagnetism was proposed (Langevin, 1905), and the molecular-field hypothesis was stated (Weiss, 1906). These approaches already referred to a collective behavior of microscopic magnetic moments, but without a clear explanation of their origin. This happened only after the experimental (Uhlenbeck and Goudsmit, 1925) and theoretical (Dirac, 1927) discovery of the electron's spin, which allowed to propose the mechanism of exchange interactions (Heisenberg, 1929), followed by elaboration of theories of antiferromagnetism (Néel, 1936) and ferromagnetism (Néel, 1948).

In this course, we will restrict ourselves to the study of basic aspects of magnetism in solids. Our focus will be on understanding its origins at the microscopic level, and the role of interactions that lead to the macroscopic phenomena observed in experiments.

# **Basic concepts**

Let us begin with Maxwell's equations (in the International System of Units – SI).

$$\nabla \cdot \mathbf{D} = \rho , \qquad \nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t} ,$$
$$\nabla \cdot \mathbf{B} = 0 , \qquad \nabla \times \mathbf{H} = \mathbf{j} + \frac{\partial \mathbf{D}}{\partial t} . \qquad (1)$$

The involved quantities are

<b>E</b> : electric field,	<b>D</b> : electric displacement,	$\rho$ : free-charge density
<b>H</b> : magnetic field,	<b>B</b> : magnetic induction,	<b>j</b> : free-current density.

Important relations between some of them are

$$\mathbf{B} = \mu_0(\mathbf{H} + \mathbf{M}) , \qquad \mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P} , \qquad (2)$$

where the **magnetization M** and the electric polarization **P** of the medium are introduced. The constants appearing in Eq. (2) are  $\mu_0 = 4\pi \times 10^{-7}$  N/A<sup>2</sup>, the vacuum's *magnetic* permeability, and  $\varepsilon_0 = 1/(\mu_0 c^2)$ , the vacuum's electric permittivity (c being the speed of light in vacuum).

For *homogeneous* and *isotropic* systems (no spontaneous magnetization or polarization), if the applied fields are sufficiently weak to allow restriction to *linear response*, we have

$$\mathbf{j} = \sigma \mathbf{E} , \qquad \mathbf{M} = \chi \mathbf{H} , \qquad (3)$$

where  $\sigma$  is the *electric conductivity* and  $\chi$  is the *magnetic susceptibility*.

## Generalized susceptibility

Equations (3) are formulated in a very simplified way. The simple form  $\mathbf{M} = \chi \mathbf{H}$ , with a constant  $\chi$ , only applies to the case of a static and uniform field, i.e., independent of position and time. In a more general case, we expect the response (magnetization) at a point  $\mathbf{r}$  and time t to dependent also on the stimulus (magnetic field) at other points in space, and even other (earlier) times. A generalized susceptibility is defined by the relation

$$\mathbf{M}(\mathbf{r},t) = \int_{-\infty}^{\infty} dt' \int d\mathbf{r}' \, \boldsymbol{\chi}(\mathbf{r}-\mathbf{r}',t-t') \cdot \mathbf{H}(\mathbf{r}',t') \,. \tag{4}$$

It is implicitly assumed that  $\chi(\mathbf{r} - \mathbf{r}', t - t') = 0$  for t' > t. We also assume (as is generally verified) that the system is homogeneous and stationary, so that the susceptibility depends on **differences** between positions in space or between instants in time. On the other hand, the susceptibility appears as a **tensor** to take into account the possibility of a non-isotropic system.

The convolution form (both in space and in time) of Eq. (4) makes it convenient to use Fourier transforms of the quantities appearing there. Introducing the Fourier components of a generic function  $F(\mathbf{r}, t)$  as

$$F(\mathbf{k},\omega) = \int dt \int d\mathbf{r} F(\mathbf{r},t) e^{-i(\mathbf{k}\cdot\mathbf{r}-\omega t)} , \qquad (5)$$

Eq. (4) assumes the simple form

$$\mathbf{M}(\mathbf{k},\omega) = \boldsymbol{\chi}(\mathbf{k},\omega) \cdot \mathbf{H}(\mathbf{k},\omega) .$$
(6)

Note that a uniform field has a non-zero Fourier component only for  $\mathbf{k} = 0$ , and that a static field has a non-zero Fourier component only for  $\omega = 0$ . Therefore, the constant

scalar susceptibility  $\chi$  of Eq. (3) can be seen as the limit of  $\chi(\mathbf{k}, \omega)$  for  $\mathbf{k} \to 0$  and  $\omega \to 0$  in the case of an isotropic system, when the tensor  $\chi$  reduces to a scalar (multiplied by the identity matrix).

So far we have considered that a linear relation between  $\mathbf{M}$  and  $\mathbf{H}$  is valid. More generally, one should have a function  $\mathbf{M}(\mathbf{H})$ . In this context, the generalized forms (4) and (6) should be written for the variations  $\delta \mathbf{M}$  and  $\delta \mathbf{H}$ , with a field-dependent susceptibility. Then the susceptibility tensor has components given by

$$\chi_{\alpha\beta}(\mathbf{H}) = \frac{\partial M_{\alpha}(\mathbf{H})}{\partial H_{\beta}} \,. \tag{7}$$

This definition is also appropriate for a system with spontaneous magnetic order, as we will see later, when the magnetization is non-zero even in the absence of applied external field.

#### Magnetization and magnetic moment

In the domain of classical electromagnetism, a planar wire loop enclosing an area A and carrying an electric current i has a **magnetic dipole moment** (or, simply, magnetic moment) of module  $\mu = iA$ . Its application point is the loop center, and its direction (perpendicular to the loop's plane) is determined by the "right-hand rule". More generally, the magnetic moment at the center  $\mathbf{r}$  of a current distribution with density  $\mathbf{j}(\mathbf{r}')$  confined to a volume  $V'_{\mathbf{r}}$  is given by

$$\boldsymbol{\mu}(\mathbf{r}) = \frac{1}{2} \int_{V'_{\mathbf{r}}} \mathbf{r}' \times \mathbf{j}(\mathbf{r}') d\mathbf{r}' .$$
(8)

The magnetization is defined as the magnetic moment per unit volume. So, if  $d\mu(\mathbf{r})$  is the magnetic moment associated with an elementary volume dV centered at point  $\mathbf{r}$ , then the magnetization at that point is

$$\mathbf{M}(\mathbf{r}) = \frac{\mathrm{d}\boldsymbol{\mu}(\mathbf{r})}{\mathrm{d}V} \ . \tag{9}$$

In this equation, the magnetization appears as a (local) position-dependent quantity, but it is implied that the "elementary" magnetic moment  $d\mu(\mathbf{r})$  is *macroscopic*. In measurements of magnetic properties of materials, the magnetization is generally determined for an entire sample, thus corresponding to the ratio between the sample's net magnetic moment and its total volume ( $\mathbf{M} = \mu_{tot}/V$ ).

#### Units

From the "definition" of magnetic moment, Eq. (8), we see that its SI units are  $Am^2$ . It then follows that the magnetization is measured in A/m. These are also the units of magnetic field, since **M** and **H** have the same physical dimension, as seen in Eq. (2). On the other hand, the magnetic induction **B** is measured in tesla (T), with  $1 \text{ T} = \frac{\mu_0}{4\pi} \times 10^7 \text{ A/m}$ .

It is quite usual in the field of Magnetism (partly for historical reasons) to employ the so-called *Gaussian* units, based on the CGS system. Then, Maxwell's equations take the form

$$\nabla \cdot \mathbf{D} = 4\pi\rho , \qquad \nabla \times \mathbf{E} = -\frac{1}{c}\frac{\partial \mathbf{B}}{\partial t} ,$$
  
$$\nabla \cdot \mathbf{B} = 0 , \qquad \nabla \times \mathbf{H} = \frac{4\pi}{c}\mathbf{j} + \frac{1}{c}\frac{\partial \mathbf{D}}{\partial t} , \qquad (10)$$

and relations (2) are replaced by

$$\mathbf{B} = \mathbf{H} + 4\pi \mathbf{M} , \qquad \mathbf{D} = \mathbf{E} + 4\pi \mathbf{P} .$$
(11)

Therefore, the Gaussian quantities **B**, **H**, and **M** all have the same physical dimension. Nevertheless, units with distinct denominations are employed: gauss (G) for **B**, oersted (Oe) for **H**, and emu/cm<sup>3</sup> for **M**, with emu (*electromagnetic unit* – equivalent to erg/gauss) being the unit of magnetic moment  $\mu$ . The following table summarizes the magnetic units introduced above for both unit systems (we use H, B, M, and  $\mu$  to denote magnitudes of the respective vectors).

Quantity	symbol	SI unit	Gaussian unit	conversion factor
magnetic induction	В	tesla $(T)$	gauss (G)	$1 \text{ T} = 10^4 \text{ G}$
magnetic field	H	A/m	oersted (Oe)	$1 \text{ A/m} = 4\pi \times 10^{-3} \text{ Oe}$
magnetic moment	$\mu$	${ m A}{ m m}^2$	emu	$1 \text{ Am}^2 = 10^3 \text{ emu}$
magnetization	M	A/m	$emu/cm^3$	$1 \text{ A/m} = 10^{-3} \text{ emu/cm}^3$

Note that the **numerical value** of a magnetic field measured in oersted is the same as that of the corresponding magnetic induction measured in gauss. It is also usual to quote a measured magnetic field by the corresponding value of  $\mu_0 H$  in tesla. For conversion purposes, it is worth keeping in mind that  $\mu_0 H = 1$  T corresponds to H = 10 kOe.

It is also worth remarking that the magnetic susceptibility is **dimensionless** in both unit systems, but the numerical value of  $\chi$  in SI is  $4\pi$  times larger than its Gaussian value when the susceptibility in the latter is expressed in its "natural" units (emu/cm<sup>3</sup> Oe). However, in experimental measurements of susceptibility it is usual to quote the *molar susceptibility*, with its value in m<sup>3</sup>/mol (SI) or emu/mol Oe (Gaussian)), which corresponds to defining the magnetization as the magnetic moment per mol of the substance and not per unit volume.

In this course, especially when dealing with microscopic models, we will use the Gaussian formulation, since in the absence of a magnetization it allows to exchange the vectors **B** and **H** without worrying about factors of  $\mu_0$ .

#### **Diamagnetism and Paramagnetism**

The denomination *diamagnetic* applies to substances that, not presenting magnetization at zero field, exhibit a magnetization **opposite** to an applied field, that is, a **negative** magnetic susceptibility. On the other hand, substances that do not present spontaneous magnetization but become magnetized **parallel** to the applied field, i.e., have a **positive** magnetic susceptibility, are called *paramagnetic*.

Intuitively, the existence of microscopic electric charges in a substance (although globally neutral), and taking Lenz's Law into account **seems** to explain diamagnetism. In fact, the situation is more complex, since the diamagnetic response is observed for **static** magnetic fields. Paramagnetism can only be understood if we consider the existence of **permanent microscopic magnetic moments**, even though the substance does not show an overall magnetization at zero field. It is known from classical electromagnetism that a magnetic dipole minimizes its potential energy when oriented parallel to the applied field. We will see later how a quantum-mechanical treatment and the existence of the electron spin allow us to understand both kinds of behavior.

### Magnetic order

The very origin of Magnetism as a "discipline", as we have mentioned earlier, is due to the existence of substances that exhibit *spontaneous magnetization*. These substances are called *ferromagnetic*, and their best-known example is obviously iron. However, ferromagnetic substances do not always have a net non-zero magnetization, but remain magnetized after application and removal of a sufficiently intense magnetic field. To understand these phenomena we have to assume the presence of **interactions** between microscopic magnetic moments favoring their parallel alignment, as we will see during this course.

The behavior of another type of substances may be understood only by considering that they have locally oriented magnetic moments, as in the case of ferromagnetism, but that **alternately** reverse their orientations throughout the system, so that the net magnetization is zero. Such substances are called *antiferromagnetic*.

Other types of magnetic order are also observed, as *ferrimagnetism* (similar to antiferromagnetism, but with non-equivalent opposing moments), *helimagnetism* (ferromagnetic order in "layers" with a non-zero angle between the magnetizations of successive layers), etc.

It is worth mentioning a kind of magnetic system know as *spin glass*. In such a system, there exist permanent microscopic magnetic moments but they are **not ordered**. However, they do not individually average to zero, as it happens in paramagnetism, but are frozen at randomly oriented directions with respect to one another.

These briefly described phenomenological aspects of magnetism will be explored in more detail throughout the course, starting from a microscopic point of view, and seeking to understand the observed phenomena at the macroscopic level.