SELECTED TOPICS IN SOLID STATE PHYSICS

Editor E. P. Wohlfarth

Volume XV

MAGNETISM AND METALLURGY OF SOFT MAGNETIC MATERIALS

BY C. W. CHEN

NORTH-HOLLAND

MAGNETISM AND METALLURGY OF SOFT MAGNETIC MATERIALS

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Magnetism and Metallurgy of Soft Magnetic Materials

BY

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To the Memory of My Father CHEN RUENN-LIN (1879–1946) A Dedicated Educator

PREFACE

The development of new materials and novel applications for the computer and telecommunications industries during the past two decades has immensely broadened the scope and altered the nature of soft magnetic materials. No longer is this field dominated by metallic substances in terms of technological importance. Nonmetallic compounds and amorphous thin films share the spotlight, if not steal the show. Consequently, a basic knowledge of this large group of materials involves two major components: (1) traditional magnetism, which imparts magnetization and spin-dependent properties, and (2) metallurgy, which governs the preparation of the products and the attainment of their structure-sensitive properties. The present book has been written in this context and is therefore divided into two parts to cover these two components. The word "metallurgy" in the title is used in a broad sense. It would have been replaced by "materials science" had the word "materials" not already appeared at the end of the title.

Combining magnetism and materials science, the aim of the book is to present, with coherency and consistency and in logical sequence, the physical principles that underlie the intrinsic as well as the applied properties of soft magnets. Discussions on some of the metallurgical phenomena are rather brief in order to hold down the volume of the book. Further information on these phenomena can be found in several texts on physical metallurgy cited in Chapter 5. In discussing many topics of great fundamental importance in Chapters 2, 3 and 4, I have tried to make the presentations as complete as possible without burdening the reader with tedious mathematics and trivial details. Strong emphasis is placed on their physical implications and effects on applications, however. Throughout the book, a policy is adopted and practised as much as possible to refer the theories and experimental studies originated before 1948 to later pertinent publications. Thus the reader will not be distracted by the antiquity of the original work, while the later publications are generally more accessible and understandable.

PREFACE

The book is intended for researchers and development workers who wish to obtain a comprehensive working knowledge of this field. Also, I have twice used it with good results as a textbook for an interdisciplinary graduate course on magnetic materials at the Iowa State University. Therefore I should think, and hope that the book can be used likewise at other universities. For the latter purpose, I should point out that although the book deals exclusively with soft magnetic materials, all principles thus discussed herein apply equally well to hard materials. The only item missing for the permanent magnets is the description of their design, preparation and applications. The missing information can be found in the review articles published in IEEE Trans. Magnetics, Vol. 4, pp. 84–99 and 221–8 (1968).

I am indebted to the Editor of the Series, Professor E. P. Wohlfarth, without whose encouragement and advice I would not have undertaken this time-consuming task. For the preparation of the manuscript, grateful acknowledgment is made to Mrs. Carol Greiner for her editorial assistance, to Miss Verna Thompson and Mrs. Erma Miller for their elegant typing and to D. W. Sailsbury, who did all the photographic reproduction of numerous figures. I am deeply grateful to W. J. Carr, Jr. of the Westinghouse Laboratories, W. H. Hu of the US Steel Research Center, R. R. Coltman of the Oak Ridge National Laboratory and many of my colleagues, M. F. Berard, O. N. Carlson, K. A. Gschneidner. Jr.. F. X. Kayser and M. L. Covault for their kindness in reading portions of the manuscript and suggesting improvements. I also wish to express my appreciation to S. Legvold and to S. H. Liu for helpful discussions on ferromagnetism in lanthanides. I must thank many authors and publishers for permitting me to reproduce many figures and tables from their papers or books. In this connection, I am especially grateful to Dr. R. M. Bozorth, Professor S. Chikazumi and Professor F. Brailsford and their publishers (Litton Educational Publishing International, John Wiley & Sons, and D. Van Nostrand Company Ltd, London, respectively), who granted me their kind permission for reprinting numerous figures from their renowned books. Thanks are due to the Ames Laboratory, US Energy Research and Development Administration, for generous secretarial and graphic services.

Chih-Wen CHEN

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LIST OF MOST IMPORTANT SYMBOLS

- a Lattice parameter
- A Area
- A_{ex} Exchange stiffness
- **B**, B Magnetic induction
- B_r Remanence
- $B_{r,\infty}$ Retentivity
- C Curie constant
- d_w Thickness of domain wall
- E_e Magnetoelastic energy
- E_s Magnetostatic energy
- f Frequency
- **F** Force
- F Helmholtz free energy
- g Landé factor
- G Gibbs free energy
- h Planck's constant
- H, H Applied magnetic field
- H_{a}, H_{k} Anisotropy field
- *H*_c Coercive force
- $H_{c,\infty}$ Coercivity
- H_d Demagnetizing field
- $H_{\rm W}$ Weiss molecular field
- I, I Electric current
- j Total angular momentum quantum number of electron
- J Total angular momentum quantum number of atom
- J_{ex} Isotropic exchange integral
- k Boltzmann's constant
- K_1, K_2 First and second magnetocrystalline anisotropy constants
- *l* Orbital angular momentum quantum number of electron
- L Orbital angular momentum quantum number of atom
- m Total magnetic quantum number, magnetic pole strength

xii	LIST OF MOST IMPORTANT SYMBOLS
mı	Orbital magnetic quantum number of electron
m_0	Rest mass of electron
ms	Spin magnetic quantum number of electron
М , М	Intensity of magnetization, or simply magnetization
М	Total magnetic quantum number of atom
M _A	Magnetization for a mass of the substance corresponding to the atomic weight
Ms	Saturation magnetization
Mo	Saturation magnetization at 0°K
n	Total or principal quantum number of electron
n _{eff}	Effective Bohr magneton number per atom
No	Avogadro's number
Ν	Number of atoms per unit volume
N_{d}	Demagnetizing factor
p , p	Angular momentum
s	Spin quantum number of electron
S	Spin angular momentum quantum number of atom, entropy
Τ	Temperature, torque
Т	Tesla
U	Internal energy
V	Volume, electric potential
$W_{\rm h}$	Hysteresis loss
z	Coordination number or number of nearest neighbors
Ζ	Atomic number
Zn	Number of outer electrons per atom
γ	Magnetomechanical or gyromagnetic ratio
γex	Exchange energy of a domain wall per unit area
γ	Surface energy density of domain wall
$\theta_{\rm C}$	Curie temperature
$\theta_{\rm F}$	Ferrimagnetic Néel temperature
θ_{N}	Néel temperature
λ	Weiss molecular field constant, characteristic length of a bub- ble material
λs	Saturation longitudinal magnetostriction
μ	Absolute normal permeability, magnetic dipole moment
$\mu_{ m i}$	Initial permeability
μ_{\max}	Maximum permeability
μ_{r}	Relative normal permeability
μ_0	Permeability of vacuum
$\mu_{\rm B}$	Bohr magneton

- ξ Mobility of domain wall
- ρ Density, electrical resistivity
- ϕ Magnetic flux
- χ, χ' Absolute and relative magnetic susceptibility per unit volume
- $\chi_{\rm m}, \chi'_{\rm m}$ Absolute and relative mass susceptibility
- χ_A, χ'_A Absolute and relative atomic susceptibility
- χ_M, χ'_M Absolute and relative molar susceptibility
- ω Angular velocity, angular frequency
- $\omega_{\rm R}$ Resonance angular frequency

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CHAPTER 1

INTRODUCTION

§1. HISTORICAL

The history of the development of magnetic materials is undeniably long and splendid. According to Chinese history, a legendary ruler of the ancient kingdom, Huang-Ti (the Yellow Emperor), made use of the compass to direct his victorious battles against barbarian tribesmen before 2600 BC. The history of Asia Minor, another cradle of civilization, claims that lodestone was discovered as a natural magnet in Magnesia more than 3500 years ago. The stone was called magnes lapis, which means magnesian stone. "Magnet" and "Magnetism" were derived from the word "magnesian." Thales of Milestus, the Greek philosopher, stated that the magnetic interaction between magnetite and iron was known before 600 BC.

Magnetic materials also played a prominent role in the discovery of the New World and in the development of modern technology. Without the compass Christopher Columbus would not have made his voyages and discoveries. The outstanding magnetic properties and low cost of iron have made it possible to generate inexpensive electricity on a massive scale since 1886 when Westinghouse Electric Company built the first commercial AC generating station at Buffalo, New York. The use of magnetic materials to perform vital functions is not limited to the utility industry, other industries consuming appreciable amounts of magnetic materials include communications, computer, audio-visual, and home appliances, to name just a few.

From the scientific viewpoint, it is understandable that magnetic materials have contributed vitally to the history of civilization and technology. In the first place, all substances, whether solid, liquid or gas, display certain magnetic characteristics at all temperatures. Hence magnetism is one of the basic properties of materials. Secondly, although only three elements (Fe, Co, and Ni) out of a total of 106 are ferromagnetic at room temperature, we are fortunate that iron constitutes approximately 3.3% of the earth's crust, and hence, it is

INTRODUCTION

abundant in natural resources and relatively inexpensive. Thirdly, metallic meteorites are known to consist largely of pure iron. It is logical to assume that, by the time ancient man appeared on the earth, pure iron had already existed for millions of years. The natural occurrence of Fe_3O_4 as lodestone is just as old as meteorites and probably more widespread than the latter. Thus it is not surprising that the magic stone and pure iron were used so early in our history.

Magnetism appears in various forms, as will be discussed in §4; but the kinds that have practical value, and therefore interest us here, are known as ferromagnetism and ferrimagnetism. Traditionally, only those elements that exhibit ferromagnetic properties are called magnetic. Besides Fe, Co and Ni, six elements in the lanthanide series (Gd, Tb, Dy, Ho, Er and Tm) become ferromagnetic at $\approx 20^{\circ}$ C (Gd) and subzero temperatures. The list of magnetic materials, nevertheless, is infinitely long because an unlimited number of magnetic solid solutions and compounds can be formed of magnetic and certain nonmagnetic elements.

Soft magnetic material is more a technical term than a scientific one. The word "soft" means temporary in the sense that the ferromagnetism emerges only when a magnetic field is applied. In contrast, hard or permanent magnets display ferromagnetism in the absence of an external field*. All magnetic elements in the pure form are soft; whereas magnetic solid solutions and compounds can be either soft or hard, depending on the composition and metallurgical treatments. In this book, we are concerned mainly with soft magnetic materials. However, the physical principles discussed in this book are also applicable to hard magnets.

§2. UNITS

In accordance with the 1960 adoption by the Conférence Générale de Poids et Mesures (Chiswell and Grigg [1971]), the Système International (SI) d'Unités is used throughout the book. This system is based on the metre-kilogram-second-ampere (MKSA) system. It includes such practical units as ampere, volt and ohm; hence, it is particularly convenient when eddy currents and other phenomena relating magnetism to elec-

^{*}Although the earth produces a magnetic field, its strength (from $32 \text{ A} \cdot \text{m}^{-1}$ or 0.4 Oe at the equator to $64 \text{ A} \cdot \text{m}^{-1}$ or 0.8 Oe at the poles) is too small to be considered as an effective external field.

tricity are discussed. The system offers another advantage in that the cumbersome factor 4π is eliminated in (1.6), which links magnetic induction **B** with magnetization **M**. The various SI units for magnetic quantities will be discussed in the next section.

The adoption of the SI units may cause inconvenience to some readers who are more accustomed to the CGS units. Furthermore, the CGS system has thus far been the choice of most scientists in their publications. For these reasons, the equivalent values in CGS units are given in parentheses following many of the quantities expressed in SI units. A table of two-way conversion between major units of the two systems is given in appendix 1.

§3. FUNDAMENTAL QUANTITIES OF MAGNETISM

The quantities discussed in this section are considered fundamental because they deal with elemental magnetic properties covering all kinds of magnetism.

§3.1. Magnetic poles

Experiment shows that a natural magnet of lodestone in the shape of a long and thin bar sets up a magnetic field at each end. This field is radially outward at one end (N or positive) and radially inward at the other end (S or negative). The magnet may then be considered as possessing two equal and opposite poles at the ends. The strength of each pole m is defined in the Coulomb interaction that the force F exerted on a magnetic pole of strength m_2 by another pole of strength m_1 at a distance r is

$$F = K \frac{m_1 m_2}{r^2} r_0,$$
(1.1)

where r_0 is a unit vector along *r*. In the SI, the magnitude of *F* is expressed in newton $(1 \text{ N} = 1 \text{ kg} \cdot \text{m} \cdot \text{s}^{-2})$ and *m*, a scalar quantity, in A · m when the constant of proportionality *K* takes the rationalized value of $1/\mu_0 c^2$, where $\mu_0 \equiv 4\pi \times 10^{-7}$ henry/metre $(\text{H} \cdot \text{m}^{-1})$ is the magnetic permeability of free space (vacuum) and $c = 2.998 \times 10^8 \text{ m} \cdot \text{s}^{-1}$ is the speed of light in a vacuum.

§3.2. Magnetic dipole and magnetic moment μ

Magnetic poles have never been observed in isolated form, but occur in pairs. Such a pair is called a magnetic dipole. The magnetic moment of

a dipole is defined as

$$\boldsymbol{\mu} = \boldsymbol{m}\boldsymbol{l},\tag{1.2}$$

where *l* is the vector pointing from the S pole to the N pole and μ is expressed in A \cdot m².

§3.3. Magnetic field H

The strength of the magnetic field H induced by either pole of a bar magnet of pole strength m_1 , at a distance r, is defined as the magnetizing force acting on a unit test pole ($m_2 = 1$) placed at that position. Accordingly,

$$H = \frac{F}{m_2} = K \frac{m_1}{r^2} r_0.$$
(1.3)

The popular oersted (Oe) is the unit of H in the CGS system. In the latter system, since K has the rationalized value of unity, one oersted corresponds to a repulsive force of one dyne acting on a unit test pole by another unit pole of like kind at a distance of one centimeter. The SI unit of H is based on the fact that a magnetic field is often produced by a current of electricity. For a long solenoid having N turns per metre and carrying a current of I amperes, the magnetic field produced inside the solenoid is given by

$$\boldsymbol{H} = \boldsymbol{N}\boldsymbol{I}.\tag{1.4}$$

Hence the SI unit of H is the ampere-turn per metre, or simply, the ampere per metre (A \cdot m⁻¹). One ampere per metre is equivalent to 0.0126 oersted and one oersted is roughly 80 A \cdot m⁻¹.

§3.4. Magnetization M and magnetic induction B

Magnetization, or more completely, the intensity of magnetization M, is the total magnetic moment of dipoles per unit volume in units of $A \cdot m^2$ per m^3 or $A \cdot m^{-1}$. Considering a bar magnet of pole strength m, length land cross-sectional area A, the vector quantity M has its direction pointing from the S pole to the N pole and its magnitude given by

$$M = \mu/V = m/A. \tag{1.5}$$

So magnetization is also the pole strength per unit area in units of $A \cdot m^{-1}$, which is equivalent to 10^{-3} gauss (G) in the CGS system.

Magnetic induction or magnetic flux density B is the flux per unit

area and expressed in units of $Wb \cdot m^{-2}$ or tesla (T). By flux is meant the number of lines of induction crossing a given area at a right angle. The weber is the magnetic flux that, linking a circuit of one turn, produces in the circuit an electromotive force of one volt as the flux is reduced to zero at a uniform rate in one second. The idea of using endless flow lines between the opposite poles of a magnet to represent magnetic induction was first conceived by M. Faraday. Indeed when a magnet is brought underneath a piece of cardboard on which iron filings are sprinkled, the magnet will force the filings to form endless flow lines emanating from the N pole and converging at the S pole, thus depicting the pattern of the magnetic induction, as shown in fig. 1.1. Hence **B** also is called the magnetic flux density.

In free space a magnetic field produces a magnetic induction given by $B = \mu_0 H$. If the space filled with any magnetic substance in which the induced magnetization is $\mu_0 M$, the total induction now becomes

$$\boldsymbol{B} = \boldsymbol{\mu}_0(\boldsymbol{H} + \boldsymbol{M}). \tag{1.6}$$

Thus both the magnetizing field and the magnetization contribute to the induction. For ferromagnetic materials, however, the contribution of M usually dominates B.

In the CGS system, since $\mu_0 = 1$, (1.6) becomes

$$\boldsymbol{B} = \boldsymbol{H} + 4\pi \boldsymbol{M}. \tag{1.7}$$

The factor 4π arises from the fact that a unit pole produces a unit field everywhere on the surface of a sphere of unit radius (1 cm) enclosing the pole, and the surface area of this sphere is 4π cm². The unit of **B** in



Fig. 1.1. Lines of magnetic induction, formed by iron filings, depicting the magnetizing force of a field near a group of magnets (Bozorth [1951]).

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the CGS system is the popular gauss. One gauss is equal to one line of induction per cm². In the SI, as indicated by (1.6), H and M have the same unit in $A \cdot m^{-1}$. In the CGS system, since B = H in free space, gauss frequently is used to express the magnitude of H in place of oersted. For conversion it is well to remember that one tesla is equivalent to 10^4 G.

§3.5. Susceptibility χ and permeability μ

To compare the magnetic response of various materials, we define the volume susceptibility, or simply susceptibility, χ , and the absolute permeability μ by

$$\chi = M/H \quad \text{and} \quad \mu = B/H, \tag{1.8}$$

where H is the applied magnetic field. Thus in SI, since $\mu_0 = 4\pi \times 10^{-7} \,\mathrm{H} \cdot \mathrm{m}^{-1}$, not unity, the absolute value of permeability divided by μ_0 gives the relative permeability, $\mu' = \mu/\mu_0$. While χ and μ' are dimensionless, μ is expressed in henry per metre (H $\cdot \mathrm{m}^{-1}$), or appropriate SI units per m³. (In the CGS system, since $\mu_0 = 1$, the absolute and relative values of μ are identical, and they are numerics as χ .)

In the literature, the susceptibility has two other forms: (A) mass susceptibility, χ_m , which is related to χ by

$$\chi_{\rm m} = \chi/\rho, \tag{1.9}$$

where ρ is the density expressed in kg \cdot m⁻³ in SI and χ_m is based per kg; and (B) atomic susceptibility, χ_A , which is related to χ_m by

$$\chi_{\mathsf{A}} = \chi_{\mathsf{m}} \cdot w, \tag{1.10}$$

where w is the atomic or molecular weight and χ_A is based per gram-mole. Whenever the material is monatomic, the atomic susceptibility is also the molar susceptibility, χ_M , which refers to one molecular gram-weight (one mole) of the material dissolved in sufficient solvent to make one cubic decimeter $(=10^{-3} \text{ m}^3)$ of solution. Experimenters favor the volume susceptibility because it can be measured directly, whereas theoreticians prefer the mass and the atomic or molar susceptibilities in their analysis. For copper at 18° C, the volume susceptibility is $\chi = -9.24 \times 10^{-6}$ per m³. With $\rho = 8.96 \times 10^3 \text{ kg} \cdot \text{m}^{-3}$ and $w = 63.54 \times 10^{-3} \text{ kg} \cdot \text{g-mole}^{-1}$ for Cu, we have $\chi_m = -9.24 \times 10^{-6}/8.96 \times 10^3 = -1.03 \times 10^{-9}$ per kg and $\chi_A = \chi_M = -1.03 \times 10^{-9} \times 63.54 \times 10^{-3} = -6.55 \times 10^{-11}$ per g-mole.

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From (1.6) and the definition of μ' , we obtain

$$\boldsymbol{\mu}' = 1 + \boldsymbol{\chi}. \tag{1.11}$$

If other forms of χ are used in (1.11), there will be corresponding changes in μ' .

§4. VARIOUS KINDS OF MAGNETISM

We have stated at the outset that all substances display certain magnetic properties at all temperatures, regardless of their composition and state. Now we shall review the various kinds of magnetism that have been observed in solids. Broadly speaking, there are five basic kinds of magnetism; namely, (1) diamagnetism, (2) paramagnetism, (3) ferromagnetism, (4) ferrimagnetism and (5) antiferromagnetism. In this listing, metamagnetism is not included because it is merely a phenomenon in which antiferromagnetism is replaced by ferromagnetism upon the application of a strong magnetic field or a decrease in temperature. Nor are superparamagnetism, parasitic ferromagnetism and mictamagnetism included, which may be viewed as variations of the ones listed.

In simple terms, a solid is said to be diamagnetic if it is repelled by a permanent magnet and to be paramagnetic, ferromagnetic or ferrimagnetic if it is attracted. The classification of magnetism can be made more precisely, of course. A convenient way to define the first four kinds of magnetism is to use the susceptibility or relative permeability as follows:

Criterion for			Ferro- or
classification	Diamagnetism	Paramagnetism	ferri-magnetism
Susceptibility, χ Relative	< 0	≥0	≥ 0
permeability, μ'	< 1	≥1	≥ 1

Table 1.1 lists some outstanding examples of each kind and typical values of their susceptibility or relative permeability at room temperature. In the Periodic Table, various groups of elements are shown to display certain kinds of magnetism. For instance, inert gases and noble metals (Cu, Ag and Au) are diamagnetic, alkali metals (Li, Na, K and Rb) and all transition metals except Fe, Co and Ni are paramagnetic, iron group transition metals and heavy lanthanide metals are ferromagnetic, etc. Ferrimagnetism exists only in compounds and alloys

Kind of magnetism and examples	Susceptibility, χ					
(I) Diamagnetism						
Copper	-1.0×10^{-5}					
Zinc	-1.4×10^{-5}					
Gold	-3.6×10^{-5}					
Mercury	-3.2×10^{-5}					
Water	-0.9×10^{-5}					
Hydrogen (n.t.p.)**	-0.2×10^{-8}					
(II) Paramagnetism						
Lithium	$+4.4 \times 10^{-5}$					
Sodium	$+0.62 \times 10^{-5}$					
Aluminum	$+2.2 \times 10^{-5}$					
Vanadium	$+38 \times 10^{-5}$					
Palladium	$+79 \times 10^{-5}$					
Neodymium	$+34 \times 10^{-5}$					
Air† (n.t.p.)	$+36 \times 10^{-8}$					
	Approximate highest value of					
	relative permeability, $\mu'(=1+\chi)$					
(III) Ferromagnetism						
Iron crystals	1.4×10^{6}					
Cobalt crystals	$\approx 10^3$					
Nickel crystals	$\approx 10^{6}$					
3.5% Si-Fe (transformer						
sheets)	7×10^4					
(single crystals)	3.8×10^{6}					
(IV) Ferrimagnetism						
Fe ₃ O ₄	100					
Ferrites	5000					

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Examples of diamagnetism, paramagnetism, ferromagnetism and ferrimagnetism and their susceptibility or relative permeability values* at 18°C

*References: Brailsford [1966a] and Bates [1963a].

**Normal temperature (18°C) and pressure (1 atmosphere).

[†]Oxygen is paramagnetic and nitrogen is diamagnetic.

because it involves at least two magnetic moments, μ_A and μ_B (the origin of which is discussed in ch. 2, §1) carried by ions of different species. While all moments of the same ions may be coupled together



Fig. 1.2. Spin arrangement in a crystal or domain illustrating (a) paramagnetism, (b) ferromagnetism, (c) antiferromagnetism and (d) ferrimagnetism.

ferromagnetically, the coupling between μ_A and μ_B is antiferromagnetic (see fig. 1.2d). The alignment of magnetic moments in a simple antiferromagnet is shown in fig. 1.2c in comparison with those of paramagnetic and ferromagnetic materials. Several metals show antiferromagnetism, notably Cr, Mn and some of the light lanthanides.

Next we shall examine the magnetic susceptibility for diamagnetism and paramagnetism. A prominent group of diamagnetic substances is the inert gases. These gas atoms have their outermost subshell completely filled. Therefore, their ground states all have the common term ${}^{1}S_{0}^{*}$, of which the magnetic moment is zero because the spin, orbital and total angular momentum quantum numbers are all zero. (See ch. 2, §1 for detailed explanation.)

The observed diamagnetic susceptibility may be explained, however, by drawing an analogy to the Lenz law in electrodynamic theory. This law states that, when the flux through an electrical circuit is changed, an induced current is produced in such a direction as to oppose the flux change. The application of a magnetic field H to an inert gas would then cause the orbital motion of each electron in the atom to precess about H, thus inducing a magnetic moment μ_a for the collection of Zelectrons in each atom. The volume susceptibility of a rare gas and other diamagnetic substances can be shown (Kittel [1976a]) as

$$\chi = \frac{N\mu_{\rm a}}{H} = -\frac{NZ\mu_0 e^2}{6m_0} \langle r^2 \rangle, \qquad (1.12)$$

where N is the number of atoms per unit volume, m_0 is the rest mass of electron, e is the electronic charge and $\langle r^2 \rangle$ is the mean square radius of

^{*}For metals, the ${}^{1}S_{0}$ ground state term is neither a necessary condition (e.g. Cu with ${}^{2}S_{1/2}$, In with ${}^{2}P_{1/2}$ and Pb with ${}^{3}P_{0}$ are diamagnetic) nor a sufficient condition (e.g. Mg, Ca, Ba and Yb are paramagnetic) for the occurrence of diamagnetism.

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the electron charge cloud about each atom. This is the formula originally derived from the classical treatment, but also arrived at in the quantum-mechanics analysis. It worked well for the inert gases. For example, an atomic susceptibility of -2.39×10^{-11} in SI units per g-mole was obtained for helium as compared with measured value of -2.40×10^{-11} .

From (1.12) we conclude that diamagnetic susceptibility is due entirely to the orbital motion of electrons associated with Larmor precession. For metals, it is important to remember that their atomic cores, having completely filled subshells, are normally associated with a small, negative susceptibility. Hence the negative diamagnetic effect is always present in metallic materials even though it may be overwhelmed by the positive paramagnetic or ferromagnetic effect. This explains why many metals other than the noble ones, notably Be, Zn, Cd, In, Hg, Pb and Bi also are diamagnetic.

There are three forms of paramagnetism. Traditionally, however, paramagnetism refers to a system in which each atom carried a magnetic moment, but the atomic moments point in all directions, thereby generating no net magnetization, as shown in fig. 1.2a. The complete disorder of atomic moments comes about because there is no magnetic interaction between neighboring atoms on one hand and because the thermal agitation causes the moments to be aligned randomly on the other. This form of paramagnetism was first treated classically by Langevin (see Bates [1963a]) who considered a system of atoms in which each atom has a magnetic moment μ in Bohr magnetons, but the moments are not coupled. When a magnetic field H is applied, the moments will tend to align as close to the field as the thermal effect at temperature T permits. Applying the Maxwell-Boltzman law, Langevin arrived at the expression for the average magnetic moment $\bar{\mu}$ of the atom in the direction of H

$$\tilde{\mu}/\mu = \coth a - 1/a = L(a),$$
 (1.13)

where $a = \mu_0 \mu H/kT$ and L(a) is known as the Langevin function.

Usually *a* is very small. A typical value for μ is $1 \mu_B = 9.2732 \times 10^{-24} \text{ A} \cdot \text{m}^2$. For $H = 8 \times 10^5 \text{ A} \cdot \text{m}^{-1}$ (~10⁴ Oe) and at room temperature ($T \sim 300^{\circ}$ K), $\mu_0 \mu H/kT \approx 2 \times 10^{-3}$, thus $\mu_0 \mu H \ll kT$. Under these conditions,

$$L(a) = \frac{a}{3} - \frac{a^3}{45} + \frac{2a^5}{945} - \dots \approx \frac{a}{3}.$$
 (1.14)

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Hence, in weak to moderate fields and at high enough temperatures,

$$\bar{\mu}/\mu \simeq \mu_0 \mu H/3kT. \tag{1.15}$$

The magnetization per atomic mass (i.e. the mass corresponding to the atomic weight) is given by

$$M_{\rm A} = N_0 \bar{\mu} = \mu_0 N_0 \mu^2 H/3kT, \qquad (1.16)$$

where N_0 is the Avogadro number, and the atomic susceptibility is given by

$$\chi_{\rm A} = M_{\rm A}/H = \mu_0 N_0 \mu^2 / 3kT = C/T, \qquad (1.17)$$

where $C = \mu_0 N_0 \mu^2 / 3k$ is known as the Curie constant. The relation $\chi_A = C/T$ is known as the Curie law. This is the essential result of the classical theory of paramagnetism of the Langevin or spin type. In this theory, the magnetic axis was assumed to take any angular position with respect to the field direction.

Another type of paramagnetism was proposed by Pauli (see Kittel [1976b]) to account for the magnetic behavior of the conduction electrons. Take copper as a typical example. This metal may be envisaged as consisting of ionic cores with closed orbits up to the 3d subshell and the conduction electrons, one per atom, in the 4s band. Hence, this is a system in which the diamagnetic ionic cores are immersed in an electron gas. Set the ions aside and consider only the conduction electrons. In the absence of a magnetic field, the band structure of the 4s electrons is shown by a density-of-states diagram in fig. 1.3a. When a field is applied and remembering that the inherent magnetic moment per



Fig. 1.3. Pauli paramagnetism of the conduction electrons: (a) H = 0, (b) and (c) $H \neq 0$ applied in the upward vertical direction (Brailsford [1966]).