

Chapter 3

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ELEG/PHYS667 Magnetism & Spintronics
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Now that we have an understanding of how magnetic moments due to orbital angular momentum and spin manifests itself in single-body systems, let's see how *macroscopic* magnetism occurs. We are considering here an ensemble of non-interacting moments.

1 Types of Magnetism

Magnetism is the acquisition of a magnetic moment by matter. In the solid state, there are many types of magnetism, which can be classified by the response of a material to an external field. We define the *magnetic susceptibility* χ as the ratio of the acquired magnetic moment M of the macroscopic body to the applied magnetic field H per unit volume:

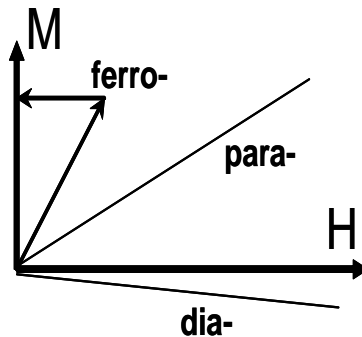
$$\chi = \frac{M}{H} \quad (1)$$

Here, since we are now talking about macroscopic physics, we make the distinction between B (used previously) and H . These two are related via the definition

$$B = \mu_0(H + M) = \mu_0(1 + \chi)H = \mu H. \quad (2)$$

where μ_0 is the magnetic permeability of free space.

We can easily characterize different forms of magnetism based on our definition of χ : Paramagnetism ($\chi > 0$), Diamagnetism ($\chi < 0$), Ferromagnetism ($\chi \rightarrow \infty$), and Antiferromagnetism (more complicated).



2 Diamagnetism

Based on Lenz's law (a consequence of Faraday's $\nabla \times E = -\frac{dB}{dt}$) we would expect that all matter is diamagnetic. In fact, most materials are weakly diamagnetic, except when the other forms of magnetism are present; they usually far outweigh the weak diamagnetic response. Since diamagnetism is of little practical use, we'll discuss it here but no further.

Actually, diamagnetism is interesting from the "gee-whiz" standpoint, because it enables magnetic levitation. The force between a magnetized body and a magnetic moment can be calculated as

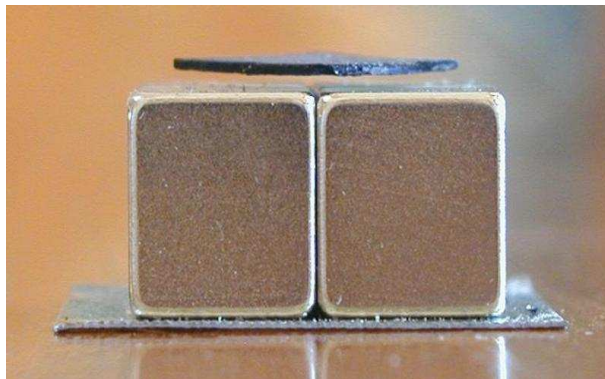
$$F = -\frac{dE}{dx} = -\frac{d}{dx}(-M \cdot H) = \frac{dM}{dx}H + M\frac{dH}{dx} \quad (3)$$

First of all, the field H decreases away from the magnetized body, so $\frac{dH}{dx} < 0$. If M is induced by H ($M = \chi H$), and $\chi < 0$, $M < 0$.

$$F = \frac{dM}{dx}H + M\frac{dH}{dx} = 2\chi H\frac{dH}{dx} > 0 \quad (4)$$

Therefore, $F > 0$ and the moment is repelled.

This phenomenon can be observed by using pyrolytic graphite in a field obtained by four permanent magnets (NdFeB-based magnets are cheap and can be bought on the internet in many appropriate sizes). The magnets are arranged to obtain a field minimum in the center, confining the graphite laterally, and levitated above the surface.



3 Paramagnetism

3.1 Classical Theory of Paramagnetism

Paramagnetism ($\chi > 0$) was first accurately explained by Langevin in the 19th century based on simple statistical mechanics developed by Boltzmann. Langevin first assumed that the paramagnet consisted of an ensemble of non-interacting magnetic moments which would align to an external field according to minimization of the dipole energy

$$E = -\mu \cdot H. \quad (5)$$

The average magnetic moment of the ensemble is given by the average moment along the field direction times the number of moments:

$$M = N\mu \langle \cos \theta \rangle. \quad (6)$$

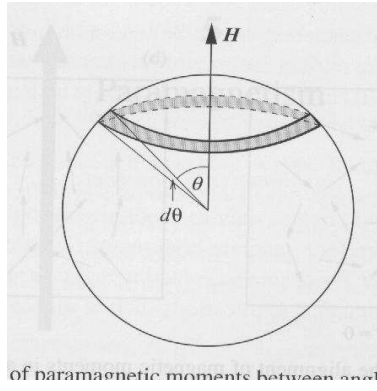
According to Boltzmann's statistical mechanics, the distribution of moment orientations is determined by the probability of the system being in a state of energy E proportional to the Boltzmann factor $e^{-\frac{E}{k_B T}}$. Therefore, since $E = -\mu \cdot H$, the probability of finding a moment at an angle θ to the field H is given by

$$P(\theta) = \frac{e^{\frac{\mu H \cos \theta}{k_B T}} \sin \theta d\theta}{\int e^{\frac{\mu H \cos \theta}{k_B T}} \sin \theta d\theta} \quad (7)$$

where the denominator is a normalization factor calculated by summing the probabilities of all states. The $\sin \theta$ accounts for the reduction of phase space close to the field axis.

We can use this expression to calculate the average value of $\cos \theta$ of moments interacting with the field by using the weighted average of the probabilities of all states of different values for θ :

$$\langle \cos \theta \rangle = \frac{\int_0^\pi e^{\frac{\mu H \cos \theta}{k_B T}} \cos \theta \sin \theta d\theta}{\int_0^\pi e^{\frac{\mu H \cos \theta}{k_B T}} \sin \theta d\theta} \quad (8)$$



Using the substitution $x = \cos\theta$ and $\alpha = \frac{\mu H}{k_B T}$, this reduces to

$$\langle \cos\theta \rangle = \frac{\int_{-1}^1 e^{\alpha x} x dx}{\int_{-1}^1 e^{\alpha x} dx} \quad (9)$$

The numerator integral can be conveniently solved by using differentiation under the integral:

$$\int_{-1}^1 \frac{d}{d\alpha} e^{\alpha x} dx = \frac{d}{d\alpha} \int_{-1}^1 e^{\alpha x} dx = \frac{d}{d\alpha} \left(\frac{e^\alpha}{\alpha} \right) = \frac{1}{\alpha} (e^\alpha + e^{-\alpha}) - \frac{1}{\alpha^2} (e^\alpha - e^{-\alpha}) \quad (10)$$

and the denominator is simple:

$$\int_{-1}^1 e^{\alpha x} dx = \frac{1}{\alpha} (e^\alpha - e^{-\alpha}) \quad (11)$$

So we get

$$\langle \cos\theta \rangle = \frac{\frac{1}{\alpha} (e^\alpha + e^{-\alpha}) - \frac{1}{\alpha^2} (e^\alpha - e^{-\alpha})}{\frac{1}{\alpha} (e^\alpha - e^{-\alpha})} = \coth(\alpha) - \frac{1}{\alpha} \quad (12)$$

This is known as the Langevin function, $L(\alpha)$, where $\alpha = \frac{\mu H}{k_B T}$. When $\alpha \ll 1$, we can expand this function in powers of α as

$$L(\alpha) = \alpha/3 - \alpha^3/45 + \dots \quad (13)$$

Therefore, under conditions of small field H or high temperature T , we can approximate

$$L(\alpha) \approx \alpha/3 \quad (14)$$

which gives us

$$M = \mu N \frac{\alpha}{3} = \mu N \frac{\mu H}{3k_B T} = \left(N \frac{\mu^2}{3k_B T} \right) H \quad (15)$$

Therefore, since $M = \chi H$,

$$\chi = N \frac{\mu^2}{3k_B T} = \frac{C}{T} \quad (16)$$

This inverse dependence on temperature is known as the ‘‘Curie Law’’ after Pierre Curie, who did his doctoral thesis work on magnetism before isolating polonium and radium with his wife Marie. This latter work won them both the Nobel prize in 1903.

3.2 Quantum Theory of Paramagnetism

Of course, we know that the magnetic moment is due to quantum variables like spin subject to angular momentum quantization, and not classical moments which can have continuous values.

In the simplest case, we have a spin-1/2 particle which has only up and down states. The integral determining the average value of the microscopic moments can then be replaced by a sum over two terms corresponding to these states. Each term is the value of the magnetic moment in the state times the thermal occupation probability:

$$M = N \langle \mu \rangle = N \frac{\mu e^{\frac{\mu H}{k_B T}} + (-\mu) e^{-\frac{\mu H}{k_B T}}}{e^{\frac{\mu H}{k_B T}} + e^{-\frac{\mu H}{k_B T}}} \quad (17)$$

$$M = N \mu \frac{e^{\frac{\mu H}{k_B T}} - e^{-\frac{\mu H}{k_B T}}}{e^{\frac{\mu H}{k_B T}} + e^{-\frac{\mu H}{k_B T}}} = N \mu \tanh \frac{\mu H}{k_B T} = N \mu \tanh \alpha. \quad (18)$$

At high temperature or in low field, we can expand to first order: $\tanh \alpha \approx \alpha$ giving

$$M = N \mu \alpha = \left(N \frac{\mu^2}{k_B T} \right) H. \quad (19)$$

Once again, we see that

$$\chi = \frac{M}{H} = N \frac{\mu^2}{k_B T} = \frac{C}{T} \quad (20)$$

(Curie's Law). When orbital angular momentum is considered, $J > 1/2$. In this case, there are more terms in the sums above. We can simplify the problem of a general value of J if we note that Eq. 17 can be written

$$M = N k_B T \frac{d(\ln Z)}{dH} \quad (21)$$

if

$$Z = \sum_{-j}^j e^{-\eta m_J}. \quad (22)$$

where $\eta = g \mu_B H / k_B T$. This is because $\frac{d}{dx} \ln Z = \frac{1}{Z} \frac{dZ}{dx}$ and the derivative brings down a factor of $g \mu_B m_J / k_B T$ before every term. Z is traditionally called the *partition function* in statistical mechanics.

If J is an integer, then we can write

$$Z = \sum_{-j}^j e^{-\eta m_J} = \sum_0^j e^{-\eta m_J} + \sum_0^j e^{\eta m_J} - 1 \quad (23)$$

Note the sign change in the argument of the exponent and the subtraction of 1 to make up for counting the $m_J = 0$ term in both sums. These are each finite geometric series which sums to

$$1 + x + x^2 + \dots + x^N = \frac{1 - x^{N+1}}{1 - x} \quad (24)$$

so we have

$$Z = \frac{1 - e^{-\eta(j+1)}}{1 - e^{-\eta}} + \frac{1 - e^{\eta(j+1)}}{1 - e^{\eta}} - 1. \quad (25)$$

Forming a common denominator and collecting like terms gives

$$Z = \frac{e^{\eta j}(1 - e^{\eta}) + e^{-\eta j}(1 - e^{-\eta})}{(1 - e^{\eta}) + (1 - e^{-\eta})} \quad (26)$$

which can be reduced to

$$Z = \frac{\sinh \eta(j+1)}{\sinh \eta/2}. \quad (27)$$

This procedure can be repeated with half-integer angular momentum ($J=3/2, 5/2, \text{etc.}$) if we start with

$$Z = \sum_{-j}^j e^{-\eta m_J} = e^{-\eta/2}(1 + e^{-\eta} + \dots + e^{-\eta(j-1/2)}) + e^{\eta/2}(1 + e^{\eta} + \dots + e^{\eta(j-1/2)}). \quad (28)$$

This leads to

$$Z = e^{-\eta/2} \frac{1 - e^{-\eta(j+1/2)}}{1 - e^{-\eta}} + e^{\eta/2} \frac{1 - e^{\eta(j+1/2)}}{1 - e^{\eta}} \quad (29)$$

and after some manipulation the result (Eq. 27) is the same.

Now we have to evaluate Eq. 21 using Eq. 27. We transform back from $\eta = g\mu_B H/k_B T$ using

$$dH = \frac{k_B T}{g\mu_B} d\eta \quad (30)$$

and perform the derivative:

$$M = Nk_B T \frac{1}{\frac{\sinh(\eta(j+1/2))}{\sinh(\eta/2)}} \frac{(j+1/2) \cosh \eta(j+1/2) \sinh \eta/2 - 1/2 \cosh \eta/2 \sinh \eta(j+1/2)}{\sinh^2 \eta/2} \quad (31)$$

which can be conveniently simplified:

$$M = \mu N [(j+1/2) \coth \eta(j+1/2) - 1/2 \coth \eta/2] = \mu N B_J(\eta) \quad (32)$$

where $B_J(\eta)$ is the ‘‘Brillouin function’’ of order J .

The Brillouin function can of course be expanded in powers of α :

$$B_J(\alpha) = \frac{J+1}{3J} \alpha - \frac{[(J+1)^2 + J^2](J+1)}{90J^3} \alpha^3 \quad (33)$$

as you can see, when $J=1/2$, these two terms are the same as the expansion of $\tanh \alpha$, as expected. Furthermore, when $\alpha \rightarrow \infty$, these terms are the same as the expansion of the Langevin function $L(\alpha)$.

At low field or high temperature, we can keep the first term to give

$$\chi_J = N \frac{g^2 J(J+1) \mu_B^2}{3k_B T} \quad (34)$$

3.3 Band Electron Paramagnetism

A magnetic field will split the states of spin up and spin down by the Zeeman energy, $\pm\mu H$. The resulting magnetic moment will given by the uncompensated spins:

$$M = \frac{\mu_B}{2} (N_{\uparrow} - N_{\downarrow}) \quad (35)$$

In the zero-temperature approximation, we can integrate over the density of states to determine analytical expressions for the numbers of spin up and down electrons:

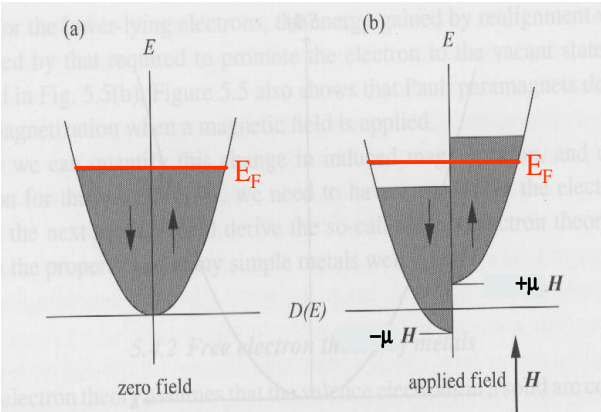
$$M = \frac{\mu_B}{2} \left[- \int_{\mu_B H}^{E_F} D(E - \mu_B H) dE + \int_{-\mu_B H}^{E_F} D(E + \mu_B H) dE \right] \quad (36)$$

which can be transformed into

$$M = \frac{\mu_B}{2} \left[- \int_0^{E_F - \mu_B H} D(E) dE + \int_0^{E_F + \mu_B H} D(E) dE \right] \quad (37)$$

$$M = \frac{\mu_B}{2} \left[\int_{E_F - \mu_B H}^0 D(E) dE + \int_0^{E_F + \mu_B H} D(E) dE \right] \quad (38)$$

$$M = \frac{\mu_B}{2} \int_{E_F - \mu_B H}^{E_F + \mu_B H} D(E) dE \quad (39)$$



7. (a) Density of states in a free electron gas with no applied field. The

If the field is small, the density of states is virtually constant over the integration region. Therefore, we can approximate this integral as

$$M = \mu_B D(E_F) [E_F + \mu_B H - (E_F + \mu_B H)] = 2\mu_B^2 H D(E_F). \quad (40)$$

This gives a magnetic susceptibility

$$\chi = \frac{M}{H} = \mu_B^2 D(E_F) \quad (41)$$

which is called “Pauli Paramagnetism”, after Wolfgang Pauli who won the Nobel Prize, for formulating his famous Exclusion Principle, in 1945.

3.4 Density of States

What is the density of states at the Fermi Energy $D(E_F)$? Let’s take a closer look into the free electron gas. The solution to the Schrödinger Equation in 3-dimensions in the absence of a potential is

$$E = \frac{\hbar^2}{2m} (k_x^2 + k_y^2 + k_z^2) \quad (42)$$

In the zero-temperature approximation, these states are filled, according to Pauli’s Exclusion Principle, from the lowest energy ($E = 0$, where $k_x = k_y = k_z = 0$), increasing in energy until all electrons in the system are accounted for. The highest filled state defines the Fermi Energy E_F . The zero-temperature approximation is generally valid for free-electron metals since $E_F \gg k_B T$.

In k-space, the Fermi surface (the boundary between filled and unfilled states) defines a sphere

$$k_x^2 + k_y^2 + k_z^2 = \frac{2mE_F}{\hbar^2} \quad (43)$$

of radius $\sqrt{\frac{2mE_F}{\hbar^2}}$. Summing up all the filled states inside the sphere will tell us the real-space density:

$$\frac{N}{V} = n = 2 \int \int \int_{\text{sphere}} \frac{d^3k}{(2\pi)^3} = 2 \int_0^{k_F} 4\pi k^2 \frac{dk}{(2\pi)^3} \quad (44)$$

But we want to express the density as an integral over energy, not over states in k-space. Therefore, we want to write

$$n = 2 \int_0^{E_F} D(E) dE. \quad (45)$$

To do this, we must equate the integrands

$$4\pi k^2 \frac{dk}{(2\pi)^3} = D(E) dE. \quad (46)$$

with the relation

$$E = \frac{\hbar^2}{2m} k^2 \quad (47)$$

Specifically, we see that

$$k = \sqrt{\frac{2mE}{\hbar^2}} \quad (48)$$

and

$$dk = \frac{m}{\hbar^2 k} = \sqrt{\frac{m}{2\hbar^2}} E^{-1/2} \quad (49)$$

result in

$$4\pi k^2 \frac{dk}{(2\pi)^3} = \frac{1}{2\pi^2} \left(\frac{2m}{\hbar^2}\right)^{3/2} E^{1/2} dE. \quad (50)$$

If we calculate the density according to Eq. 44, we get

$$n = 2 \int_0^{k_F} 4\pi k^2 \frac{dk}{(2\pi)^3} = \frac{k_F^3}{3\pi^2} \quad (51)$$

Since the Fermi Energy is defined as

$$E_F = \frac{\hbar^2}{2m} k_F^2, \quad (52)$$

we can express this as

$$n = \frac{k_F^3}{3\pi^2} = \frac{2}{3} \left[\frac{m\sqrt{2mE_F}}{\hbar^3 \pi^2} \right] E_F. \quad (53)$$

Note that the term in brackets is exactly the density of states at the Fermi Energy. Therefore,

$$D(E_F) = \frac{3}{2} \frac{n}{E_F} \quad (54)$$

and the Pauli paramagnetism takes the form

$$\chi = \frac{3\mu_B^2}{2E_F} \quad (55)$$

Landau derived a diamagnetic contribution which is one-third this value. Since diamagnetism is a negative susceptibility, the total paramagnetic response for the free-electron gas is

$$\chi = \frac{\mu_B^2}{E_F}. \quad (56)$$

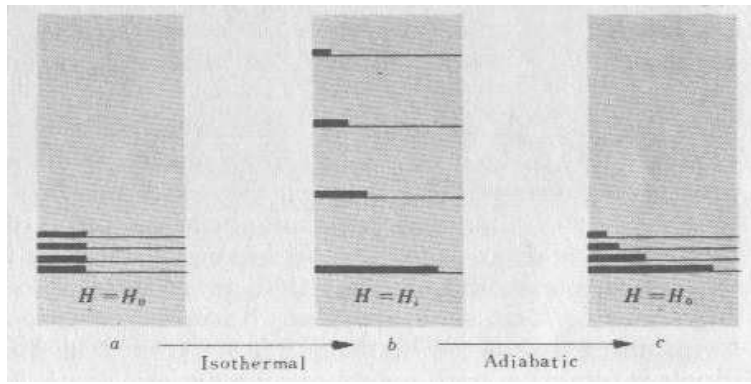
4 An Application of Paramagnetism: Adiabatic Demagnetization

If a paramagnet in a magnetic field is insulated from its surroundings and the field is turned off, the paramagnet's temperature is reduced. This process, called adiabatic demagnetization, was first suggested by Peter Debye (Chemistry Nobel Prize, 1936) in 1926.

A heuristic explanation of this phenomenon can be seen by considering a quantum system at $T = T_i$ in $H = 0$. Without a field, there are many nearly degenerate states with $\Delta E \ll k_B T_i$. Therefore, since the relative occupation between successive states is given by the Boltzmann factor

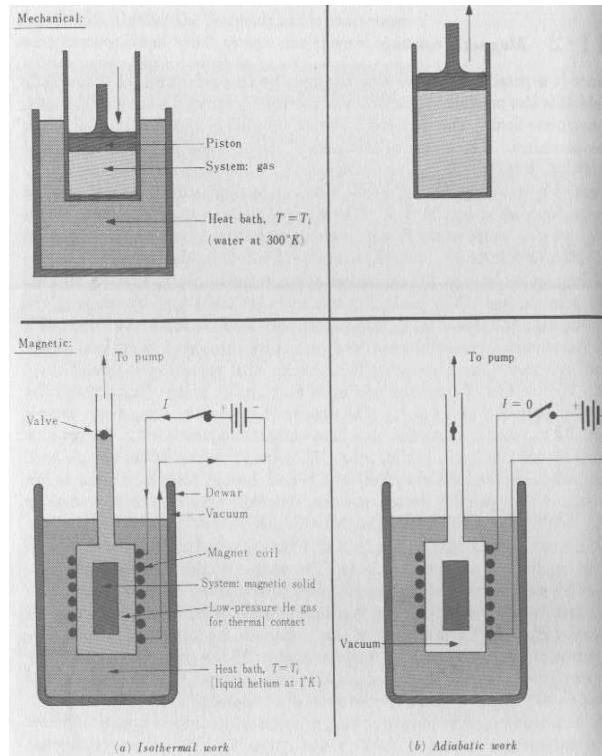
$$\frac{P(E + \Delta E)}{P(E)} = e^{\frac{-\Delta E}{k_B T_i}}, \quad (57)$$

the occupation probabilities are roughly equal. If a field is turned on, the levels will split due to the Zeeman interaction and ΔE will increase. If the paramagnet can exchange heat with a thermal bath also at $T = T_i$, then



the occupations change, and there are less electrons filling the states with higher energy, according to the Boltzmann factor.

If the system is then insulated and the field is reduced adiabatically, the levels return to their previous (nearly degenerate) condition. No heat can enter or exit the system, so the occupations remain constant. If ΔE has become smaller, so must T to maintain the constant Boltzmann factor. Therefore, the system has cooled. In some ways this is similar to cooling through adiabatic gas expansion.



During the adiabatic demagnetization, the entropy S remains constant. Since the entropy is a function of

$$\frac{\mu H}{k_B T} \quad (58)$$

We expect that $S_i = S_f$ implies

$$\frac{H_i}{T_i} = \frac{H_f}{T_f} \quad (59)$$

or

$$\frac{T_f}{T_i} = \frac{H_f}{H_i}. \quad (60)$$

Strictly speaking, this suggests that reducing the final field H_f to zero will result in a reduction of the temperature T_f to zero as well! However, H_f is never actually zero since there is a local field due to interactions between magnetic ions in the paramagnet. Dilution of the magnetic ions can reduce these interaction fields, but continued dilution will lead to reduction of the cooling power of the adiabatic demagnetization. In practice, pre-cooling with pumped liquid Helium to around 1K can result in final temperatures approaching 1mK. *Nuclear* demagnetization can achieve temperatures many orders of magnitude lower.