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# Magnetic Susceptibility Instructions

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# Abstract

The magnetic susceptibility is a material property, which describes the reaction of a substance to an external magnetic field. The main goal of the experiment is to check the quantitative agreement with reality of the Curie-Weiss law, which can be done quite easily with the so-called Gouy method: this method consists in measuring with a sensitive balance the downforce exerted on a sample by an inhomogeneous magnetic field as a function of some parameters. Even though the magnetic susceptibility is well described by some simple formulae – based on measurable macroscopic properties and well-known constants – this property can be properly explained only with a statistical, quantum-mechanical approach. In this tutorial we will also give a theoretical introduction to this topic, in order to get the feeling of the depth needed for properly understanding it.

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# 2 Assignment

Measure the vertical force, K, acting on a pulverized Dysprosium Oxide  $(Dy_2O_3)$  sample placed in an inhomogeneous magnetic field:

- 1. as a function of the height of the sample in the magnet, K(z). For the subsequent measurements you have to set the height so, that the force is maximized.
- 2. as a function of the strength of the magnetic field, thus of the current flowing through the electromagnet: K(I). These measurements must be performed at approximately T = 290 K and T = 800 K.
- 3. as a function of the temperature, K(T), with a fixed current chosen between 6 and 10 A, in the temperature interval 290 K  $\leq T \leq$  900 K, both with increasing and decreasing temperature and steps  $\Delta T \approx 40$  K. Enquire the deviations of the results from the Curie-Weiss law by means of the complete Van Vleck's theory (see section 3.2.5).

In addition, you have to:

- 1. Compare all the results with literature values.
- 2. Measure the magnetic field as a function of the current flowing through the electromagnet, and check whether the dependence is linear. The Gaussmeter is to be found in the laboratory store. Describe the method you used for performing the measurements.

The evaluation and analysis of the measured values and the determination of the susceptibility will be explained in chapter 7.

# 3 Theory

## 3.1 Phenomenological model

### **3.1.1** The magnetisation $\vec{M}$

The magnetic induction,  $\vec{B}$ , (also called magnetic flux density) is proportional to the magnetic field intensity  $\vec{H}$ . The proportionality constant is given by the product of the vacuum and material permeability,  $\mu_0$  and  $\mu$  respectively, such that

$$\vec{B} = \mu \mu_0 \vec{H},\tag{1}$$

where the vacuum permeability is given by  $\mu_0 = 4\pi \cdot 10^{-7} \frac{V s}{A m}$ .

The magnetic induction can also be decomposed into a vacuum contribution  $\vec{B}_0 = \mu_0 \vec{H}$ , and a material contribution  $\mu_0 \vec{M}$ , such that:

$$\vec{B} = \mu_0 \left( \vec{H} + \vec{M} \right) \tag{2}$$

The quantity  $\vec{M}$  is called magnetisation. In this notation (proposed by Mie)  $\vec{M}$  represents the component of the total magnetic field due to the material. Therefore  $\vec{M}$  has the same dimensionality of  $\vec{H}$ , and represents the sum of magnetic momenta of the particles per unit of volume. Another definition of  $\vec{M}$  was introduced by Pohl:  $\vec{B} = \mu_0 \vec{H} + \vec{M}$ . In this introduction we use Mie's definition (eqn. (2)).

We define the magnetic volume-susceptibility  $\varkappa$  as follows:

$$\vec{M} = \varkappa \vec{H} \tag{3}$$

Thus:

$$\vec{B} = \mu \mu_0 \vec{H} = \mu_0 (\vec{H} + \varkappa \vec{H}) = \mu_0 (1 + \varkappa) \vec{H}$$
$$\Rightarrow \mu = 1 + \varkappa \tag{4}$$

On a macroscopic level we can distinguish paramagnetic and diamagnetic substances with the sign of their susceptibilities:

$$\varkappa_{para} > 0$$
  
 $\varkappa_{dia} < 0$ 

Instead of the volume susceptibility  $\varkappa$  (which is dimensionless) we often use the molar susceptibility  $\chi_M$  (m<sup>3</sup>kMol<sup>-1</sup>) and the specific susceptibility  $\chi$ (m<sup>3</sup>kg<sup>-1</sup>).

#### 3.1.2 Remarks for completeness

**3.1.2.1 Primary quantities:** As argued by Sommerfeld,  $\vec{B}$  has to be treated as a "primary" quantity. Indeed, only  $\vec{B}$  can be measured by using the Lorentz force or the induction law. On the other hand,  $\vec{H}$  shows up in the Maxwell equation  $\vec{\nabla} \times \vec{H} = \vec{j} + \vec{D}$ , and therefore comes out necessarily in the presence of moving charges. Also  $\vec{M}$  is caused by the motion of charges, on an atomic level.

**3.1.2.2** Anisotropy of  $\varkappa$ : In the general case of an anisotropic crystal the susceptibility can only be represented through a symmetric tensor. Since a symmetric matrix represents a self-adjoined linear function, it can be orthonormally diagonalized, that is we can perform a main axis transform: we can choose an orthonormal basis, in which  $\varkappa$  is diagonal. From a physical point of view, this means that there are three orthogonal axes (the main axes) along whose  $\varkappa$  is scalar (but the scalars relative to different axes are in general not equal to each other). The anisotropy of the materials we use can potentially be a problem, since the orientation of the crystalline lattice is relevant, but not forcedly easy to determine. Nevertheless, in this experiment we only deal with polycrystalline or pulverized samples, thus we can effectively approximate the susceptibility of an anisotropic medium by averaging over the elements of the diagonalized tensor:

$$\varkappa = \frac{\varkappa_1 + \varkappa_2 + \varkappa_3}{3} \tag{5}$$

**3.1.2.3** The demagnetisation factor: If we produce an external, homogeneous field with strength  $\vec{H}_{ext}$ , this field will be disturbed by the introduction of a sample. This perturbation near and inside of the sample can be calculated with magnetostatics. For an arbitrary shape of the sample, the internal field  $\vec{H}_{in}$  is not only unequal to  $\vec{H}_{ext}$ , but it is different from place to place. Only for a sample with an ellipsoidal shape, amongst whose main axes one is parallel to  $\vec{H}_{ext}$ , we get an homogeneous  $\vec{H}_{in}$  inside of the sample:

$$\vec{H}_{in} = \frac{\vec{H}_{ext}}{1 + F\varkappa}$$
$$\vec{M} = \varkappa \vec{H}_{in}$$

The demagnetisation factor F is a number; it is equal e.g. 1/3 for spherical samples. For the samples we use in our experiment we have  $F \varkappa \ll 1$ , thus

$$\dot{H}_{in} \approx \dot{H}_{ext}$$

So, starting from now, we can avoid using the vector sign on  $\vec{M}$  and  $\vec{H}$ , since  $\vec{M}$  is always parallel to  $\vec{H}_{ext}$  (we reduced the problem to an one-dimensional one). Can you think of a situation in which the demagnetisation factor is relevant?

#### 3.1.3 Thermodynamic state functions of magnets

Electric and magnetic phenomena are in general not independent from thermal and mechanical properties of matter (secondary effects). Paramagnets are amongst the simplest systems, to which we can apply the methods of statistical thermodynamics, since – as long as the interactions between magnetic moments are negligible – they are composed by a set of independent systems, that are connected by a thermal bath (the crystal lattice).

The internal energy per unit volume  $U^\prime$  of a magnetic system can be represented as follows:

$$\mathrm{d}U' = T \cdot \mathrm{d}S - p \cdot \mathrm{d}V + H \cdot \mathrm{d}B \tag{6}$$

where:

$$H \cdot \mathrm{d}B = H \cdot \mathrm{d}H + H \cdot \mathrm{d}M \tag{7}$$

Analogously to the procedure in 3.1.1, we can separate  $H \cdot dB$  into two components. Only the second part  $H \cdot dM$  depends on the material. Subtracting the "vacuum part"  $H \cdot dH$  of the magnetic energy from U' would mean taking into account for our system only the physical body, and not the magnetic field in vacuum. Such a separation is not permitted in more complicated cases, though. The characteristic internal energy of the system is thus

$$U = U' - \frac{\mu_0}{2}H^2$$

And so

$$dU = T \cdot dS - p \cdot dV + \mu_0 H \cdot dM \tag{8}$$

This internal energy U = U(S, V, M) is then defined through the variables entropy, volume and magnetisation.

It is often convenient to make a Legendre transform from the variables (S, M) to the conjugated variables (T, H). This way we obtain a new state function, which is called (Helmholtz') thermodynamic potential  $\Phi$ :

$$\Phi = U - TS - \mu_0 HM = \Phi(T, V, H)$$
  
$$d\Phi = -S \cdot dT - p \cdot dV + \mu_0 M \cdot dH$$
(9)

From this we gain the important relation:

$$M = -\frac{1}{\mu_0} \left. \frac{\partial \Phi}{\partial H} \right|_{T,V} \tag{10}$$

and, by using equation (3):

$$\varkappa = \frac{M}{H} = -\frac{1}{\mu_0 H} \left. \frac{\partial \Phi}{\partial H} \right|_{T,V} \tag{11}$$

If we know the thermodynamic potential  $\Phi$  of the system (e.g. from a statistical model), we can also calculate:

statistical model), we can also concern. Entropy:  $S = -\frac{\partial \Phi}{\partial T}\Big|_{V,H}$ Specific heat capacity:  $c_{V,H} = T\frac{\partial S}{\partial T}\Big|_{V,H}$ Pressure:  $p = -\frac{\partial \Phi}{\partial V}\Big|_{T,H}$ 

## 3.2 Model-based part

#### 3.2.1 Partition function and energy-eigenstates

We want to calculate the thermodynamic potential  $\Phi$  with statistical mechanics based on an atomic model.

First of all, we treat a system composed of n identical particles, which do not interact with each other. We assume the volume of the system to be constantly V = 1. Statistical mechanics predict the potential  $\Phi$  of such a system with the Boltzmann distribution:

$$\Phi = kT \log \left( Z^n \right) = nkT \log Z \tag{12}$$

$$Z = \sum_{i} \exp\left(-\frac{E_i}{kT}\right) \tag{13}$$

Z is the *partition function* for the 1-particle eigenstates relative to the energy eigenvalues  $E_i$ , which are obtained by solving the Schrödinger equation for a representative particle.

Then the *susceptibility* of the system follows from (10), (11), (12), and (13):

$$\varkappa = M/H$$
$$M = -\frac{1}{\mu_0} \cdot k \cdot T \cdot n \cdot \frac{\partial \log Z}{\partial H}$$
(14)

If we know the energy-eigenvalues  $E_i$  as functions of H (Zeeman effect), we can calculate  $\varkappa$  with (13) and (14). This will be discussed in 3.2.2, 3.2.3, and 3.2.4.

Before that, we would like to generalize equation (14) for a system, that contains different types of particles, which have the number of particles per unit volume  $n_a$ ,  $n_b$ ,  $n_c$ , etc., and let us assume again no interaction between the particles. In this case we can prove, that

$$\Phi = kT \log \left( Z_a^{n_a} Z_b^{n_b} \dots \right) = n_a kT \log Z_a + n_b kT \log Z_b + \dots$$
(15)

The partition functions  $Z_a$ ,  $Z_b$ , ... can be obtained as in the case of equation (12) from the energy-eigenvalues  $(E_i)_a$ ,  $(E_i)_b$ , ... of the corresponding 1-particle Schrödinger equations. The susceptibility is then composed additively by the contributions of the various types of particles:

$$\varkappa = \varkappa_a + \varkappa_b + \ldots = -\frac{1}{\mu_0} \cdot \frac{k \cdot T}{H} \cdot \left( n_a \cdot \frac{\partial \log Z_a}{\partial H} + n_b \cdot \frac{\partial \log Z_b}{\partial H} + \ldots \right)$$
(16)

In this calculation, one must keep in mind the following remarks:

- 1. The comfortable additivity used in (16), (14), and (12) is clearly based on the assumption that the interaction between the particles is negligible (1-particle approximation).
- 2. In the literature there often appears a partition function  $Z_{system}$  for the eigenvalues  $E_i$  of a system, which is composed by many particles. The Schrödinger's method has the advantage to be applicable both to the Boltzmann-statistic and to the Fermi- or the Böse-statistic. We will not analyse this any deeper, just be careful not to confuse  $Z_{system}$  and Z.

#### 3.2.2 Electrons and ions in rigid bodies

**3.2.2.1 Magnetic moment:** Let us consider again the question posed in 3.2.1: why do the energy eigenvalues  $E_i$  of a particle depend on H? The answer is: because the particle has a magnetic moment, and the value of  $E_i$  depends on its orientation:

$$E_i = E_{i0} + (\vec{\mu} + q\vec{H}) \cdot \vec{H} = E_{i0} + \mu_z H + qH^2$$
(17)

where  $E_{i0}$  is the value of  $E_i$  for H = 0. By  $\mu_z$  we mean the z-component of the magnetic moment  $\vec{\mu}$  of the particle; we assumed that  $\vec{H} \parallel \hat{z}$ . The term  $q\vec{H}$  represents a magnetic moment, which disappears for H = 0.

Every particle with a proper angular momentum J - e.g. a neutron, an electron, a nucleus or a non-filled electron orbital – also exhibits a magnetic moment  $\vec{\mu}$ . From quantum mechanics it follows that  $\vec{J}^2$  and  $J_z$  – and thus also  $\vec{\mu}^2$  and  $\mu_z$  – are quantized quantities.

The term  $\mu_z H$  in equation (17) leads, on one hand, to the Langevin paramagnetism (which is going to be treated later on), and on the other hand to the splitting of the energy levels in the Zeeman effect. Since the magnetic moment of nuclei is about 10<sup>3</sup> times smaller than that of electrons, we can neglect the nuclear magnetism while treating the static susceptibility. As long as  $\vec{\mu} \neq 0$ , one can further show that the induced magnetic moment  $q\vec{H}$ is mostly negligible with respect to  $\vec{\mu}$ . For particles with  $\vec{\mu} = 0$  – for example the filled electron orbitals – we anyway have to consider the quadratic term  $qH^2$  in equation (17). In the case of a free atom or ion, q is composed of two parts  $q = q_{L,L} + q_{v,V}$ .

From the term  $q_{L.L.}H^2$  follows a diamagnetic contribution to the susceptibility – the so-called Larmor-Langevin diamagnetism  $\varkappa_{L.L.}$  – which is always present for bound electrons (also in the case of filled orbitals). In a precise treatment of paramagnets we thus have to take into account  $\varkappa_{L.L.}$  as well.

Apart from  $q_{L.L.}H^2$ , an additional term  $q_{v.V.}$  comes up, which leads to the so-called van Vleck paramagnetism. This is based on the fact, that in the case of bound electrons the eigenstates for  $H \neq 0$  are slightly different from the ones for H = 0, and so the charge distribution is slightly modified by the application of the magnetic field.

As a first approximation, in our experiment we are going to neglect all the effects due to  $qH^2$ . Investigations about such effects can, for example, provide us with some information about chemical bonds in organic molecules, which are mostly diamagnetic. The effects induced by H are also important for some ions of the rare earth elements. The van Vleck model will be given in 3.2.5 for completeness.

**3.2.2.2 Insulator and conductor:** As an example of an insulator we treat a salt, that contains diamagnetic ions, like e.g.  $CuSO_4 \cdot 5H_2O$ . The  $SO_4$  ions and the crystal water are composed of filled electron orbitals, whose contribution to the susceptibility is negligible if compared to that of  $Cu^{2+}$  ions. The electronic configuration of  $Cu^{2+}$  is (Ar)  $3d^q$ . It exhibits an incomplete 3d orbital, which causes Langevin paramagnetism – even though it is deformed by the introduction into the crystal.

As an example of a metal we treat one with filled internal orbitals, as e.g. Cu. The configuration of Cu is  $(Ar) 3d^{10}4s^1$ . In a first approximation we assume that in the Cu-metal the 4s electrons are free, so that in this

system exist two types of particle: the filled orbitals and the free electrons. The latter supply two contributions to the susceptibility, namely the Pauli spin paramagnetism  $\varkappa_P$  and the Landau diamagnetism  $\varkappa_L$ . More detailed informations about this item can be found in the literature, we consider ourselves satisfied with the following two remarks:

- 1. The eigenstates of the free electrons with  $H \neq 0$  are different from the ones with H = 0 because of the cyclotron motion. Correspondingly, the quantum condition (Landau quantisation) responsible for the Landau diamagnetism for  $H \neq 0$  is different.
- 2. For the calculation of  $\varkappa_P$  and  $\varkappa_L$  for a degenerate electron gas we need the Fermi statistics. In this case  $\varkappa_P$  and  $\varkappa_L$  are – in a first approximation – temperature-independent and so little, that the contribution given to the susceptibility by the inner, closed orbitals cannot be neglected any longer (equation (16)).

**3.2.2.3** The atomic model by Russel-Sounders: From quantum mechanics we know that an electron, i, in an orbital can be described by the quantum numbers  $n_i$ ,  $l_i$ ,  $m_i$ ,  $s_i$ , and  $m_{s,i}$  with the following properties:

- The square of the angular momentum,  $L^2$ , of an electron moving in the spherically-symmetric potential of the nucleus can only assume the discrete values  $\hbar^2 l_i(l_i + 1)$ , where  $l_i \in \{0, 1, 2, ..., n_i - 1\}$ . The zcomponent  $L_z$  of the angular momentum can only be given by  $L_z = \hbar m$ , with  $m \in \{-l, -l + 1, ..., l - 1, l\}$ .
- The square of the electron-spin,  $S^2$ , can only take the value  $\hbar^2 s_i(s_i+1)$ , where  $s_i = 1/2$ , and its z-component is always given by  $S_z = \hbar m_s$ , with  $m_s = \pm 1/2$ .

The quantum-mechanical connection between the magnetic moment and the mechanical angular momentum is

$$|\vec{\mu}_l| = \sqrt{l(l+1)}\mu_B,$$

where  $\mu_B = 9.2740154 \cdot 10^{-24}$  A m<sup>2</sup> is the Bohr magneton. Correspondingly, for the magnetic spin-moment:

$$|\vec{\mu}_s| = \sqrt{s(s+1)}\mu_B.$$

If the atomic shell is composed by more than one electron, the quantum number l of the resulting angular momentum  $\vec{l}$  – for the example of two electrons with angular momenta  $l_1 \ge l_2$  – can take the values:

$$l \in \{l_1 + l_2, l_1 + l_2 - 1, \dots, l_1 - l_2\}.$$

We write  $\vec{L} = \sum \vec{l_i}$  and, respectively, for the resulting mechanical spin momentum  $\vec{S} = \sum \vec{s_i}$ . The total angular momentum  $\vec{J}$  of the electron shell of the atom is the vectorial sum of the resulting orbital momentum  $\vec{L}$  and of the spin-momentum  $\vec{S}$ :

$$\vec{J} = \vec{L} + \vec{S}$$
 (Russel-Saunders coupling) (18)

The quantum number of the total angular momentum  $\vec{J}$  can take the following values:

$$j \in \{l+s, l+s-1, \dots, l-s\} \quad \text{if } l > s \\ j \in \{s+l, s+l-1, \dots, s-l\} \quad \text{if } s > l$$

The contributions of the magnetic moment vectors  $\vec{\mu}_l$  and  $\vec{\mu}_s$  are, respectively:

$$|\vec{\mu}_l| = \sqrt{l(l+1)}\mu_B$$
 and  $|\vec{\mu}_s| = \sqrt{s(s+1)}\mu_B$ 

In general the relation between the total angular momentum and the total magnetic moment is

$$\frac{\vec{\mu}_j}{\mu_B} = g_j \frac{J}{\hbar},$$

or, expressed with the eigenvalue of  $\vec{J}$ ,

$$|\vec{\mu}_j| = g_j \sqrt{j(j+1)} \mu_B,$$

where the so-called Landé factor  $g_j$  is given by:

$$g_j = 1 + \frac{j(j+1) + s(s+1) - l(l+1)}{2j(j+1)}.$$

Inside of a single multiplet we can write:

$$\vec{L} + 2\vec{S} = g\vec{J}$$

The energy difference between two different spin states is in general large and in general is decisively in the ground state which then determines the magnetic properties of the material. For this ground state the quantum numbers l and s can be predicted through the Hund's rules:

- 1. The value of s for the ground state is equal to the maximal s, which is compatible with the exclusion principle.
- 2. The value of l for the ground state is equal to the maximal l, which is agreeable with condition 1 and with the exclusion principle.

#### 3.2.3 Langevin's theory of paramagnetism (1905)

As we discussed in 3.2.2.2 – while treating paramagnetic salts – in a fist approximation we only need to consider the susceptibility-contribution of the incomplete orbitals.

Let  $\vec{J} = \vec{L} + \vec{S}$  be the total angular momentum (according to equation (18)), then the quantum condition for  $\vec{J}^2$  is given by  $\vec{J}^2 = \hbar^2 j(j+1)$ , where j represents the internal quantum number, which can assume only integer or half-integer values. From the Schrödinger equation we can derive the energy eigenvalues:

$$E(j_z) \approx E_0 + \mu_0 m_j g_j \mu_B H, \tag{19}$$

where  $m_j$  is the magnetic quantum number, such that  $J_z$  can only assume the values  $J_z = \hbar m_j$ , where  $m_j \in \{-j, \ldots, j\}$ , analogously to  $m_l$  and  $m_s$ .

From eqn. (13) follows

$$Z = e^{-\frac{E_0}{kT}} \cdot \sum_{m_j = -j}^{+j} e^{-\mu_0 m_j g_j \mu_B \frac{H}{kT}}$$
$$= e^{-\frac{E_0}{kT}} \cdot \frac{\sinh\left((j+1/2)x\right)}{\sinh\left(x/2\right)},$$
(20)

where

$$x \coloneqq \mu_0 \cdot jg_j \mu_B \frac{H}{kT} \tag{21}$$

From equation (14) and by performing some calculus we gain an expression for the magnetisation:

$$M = M_S \cdot B_j(x), \tag{22}$$

where  $M_S := n \cdot j g_j \mu_B$  is the magnetisation-value in the saturation regime, that is, when the magnetic moments of all particles are aligned by a strong external magnetic field (in our case we assume it to be in the z-direction, without the loss of generality).  $B_j(x)$  is the so-called Brillouin-function:

$$B_j(x) \coloneqq \frac{2j+1}{2j} \coth\left(\frac{2j+1}{2j} \cdot x\right) - \frac{1}{2j} \coth\left(\frac{x}{2j}\right)$$
(23)

In the following we want to list some particular cases for (22) and (23):

1. j = 1/2:  $B_{1/2}(x) = \tanh x$  (24)

2.  $j \to \infty$ , (and  $\mu_B \to 0$ , such that x stays finite):

$$B_{\infty}(x) = \coth x - \frac{1}{x} = L(x), \qquad (25)$$

where L(x) is called Langevin-function.

3.  $x \to \infty$   $(H \to \infty \text{ or } T \to 0)$ :

$$B_j \to 1 \quad , \quad M \to M_S \tag{26}$$

In order to approach the paramagnetic saturation, we thus need a very strong magnetic field and a very low temperature.

4.  $x \ll 1$ :

$$B_j = \frac{j+1}{j} \cdot \frac{x}{3} \tag{27}$$

That is, for  $j \to \infty$ :

$$B_{\infty} = \frac{x}{3},\tag{28}$$

and for j = 1/2:

$$B_{1/2} = x \tag{29}$$

From eqn. (22) follows (for this special case) a very important result, which is valid for not too big fields and not too low temperatures:

### The Curie law:

$$M = \mu_0 n \cdot \frac{p^2 \mu_B^2}{3kT} \cdot H = \frac{C}{T} \cdot H \tag{30}$$

$$\varkappa = \mu_0 n \cdot \frac{p^2 \mu_B^2}{3kT} = \frac{C}{T} \tag{31}$$

where:

$$C \coloneqq \mu_0 n \cdot \frac{p^2 \mu_B^2}{3k} \tag{32}$$

$$p \coloneqq g_j \sqrt{j(j+1)} \tag{33}$$

Equation (33) shows a good agreement with experiments for the salts of the rare earth elements, but not for the salts of transition metals like Mn or Fe. In the latter case, the experimental results are way better described by the relation

$$p = 2\sqrt{S(S+1)},\tag{34}$$

where S represents the total spin quantum number of the ion. This phenomenon was motivated on a theoretical level by Bethe.

The z-component  $L_z$  of the angular momentum in this term is not a constant of motion anymore – because of the influence of the dominating electrostatic fields in the crystal – and its expectation value  $\langle L_z \rangle$  disappears. This effect is called "extinction of  $\vec{L}_z$ ".

#### 3.2.4 Weiss' theory of ferromagnetism

So far we have been neglecting the interaction between paramagnetic atoms. This is however not allowed, if we want to describe the phenomena of arrangement coming up in para-, ferro-, and antiferromagnetism. In a rigorous theory we should fist of all take into account every existing interaction; from the resulting partition function we would be able to determine the magnetic quantities. Unfortunately, this many-particles problem can only be solved in very simple cases. We will not get any deeper into this item, we recommend to the interested readers to take a look at the specialized literature.

**3.2.4.1 Weiss' internal field:** In 1907, P. Weiss found a solution to the problem posed above in better agreement with experimental results. He applied the statistics of the individual particles (as we did before), and introduced the interactions with nearby atoms through a Weiss' internal field  $H_E = \lambda M$ . With positive values of the field-factor  $\lambda$  we can explain paramagnetism, and – as was later shown by Néel – with the introduction of sub-lattices with different Weiss' fields we can derive both antiferro- and ferromagnetism.

Equations (22) and (25) can be re-written for a ferromagnet as follows:

$$M = M_S B_{\infty}(x)$$
 with  $x = \mu_0 \cdot \frac{H + \lambda M}{kT}$  (35)

Only the magnetisation, and not the susceptibility, is defined for a ferromagnet. In the framework of the classical theory we use the Langevin-function  $B_{\infty}(x)$ , but it is possible to replace it with any Brillouin-function  $B_j(x)$ , where  $x = \mu_0 j g_j \mu_B (H + \lambda M)/(kT)$ .

The whole theory of ferromagnetism is contained in the implicit equation (35), which provides the field- and the temperature-dependence of the magnetisation.

**3.2.4.2 Curie-Weiss law:** Above the ferromagnetic transition point (the Curie temperature  $\theta$ ) the magnetisation is small, that is  $x \ll 1$ . If in eqn.

(30) we substitute H through the sum  $H + H_E$  of the external magnetic field, we get:

$$M = \mu_0 n \cdot \frac{p^2 \mu_B^2}{kT} (H + H_E) = \frac{C}{T} (H + H_E), \qquad (35^1)$$

and with

$$H_E = \lambda M \tag{36}$$

follows

$$M = \frac{C}{T - \theta} \cdot H \tag{37}$$

$$\varkappa = \frac{C}{T - \theta},\tag{38}$$

with the Curie temperature  $\theta = C \cdot \lambda$ , and the Curie constant (as in eqn. (32))  $C = \mu_0 n \cdot \frac{p^2 \mu_B^2}{3k}$ .

**Pro memoria:** The assumptions needed for the last formulae to be valid are:

$$x = \mu_0 \cdot \frac{j g_j \mu_B}{kT} (H + H_E) \ll 1$$
 and  $T > \theta$ .

Instead of the volume unit we can normalize the quantities also over 1 kMol. They are then given by

$$\chi_M = \frac{C_M}{T - \theta},\tag{39}$$

where

$$C_M = \mu_0 N \cdot \frac{p^2 \mu_B^2}{3k},\tag{40}$$

where N is the number of magnetic atoms per kMol.

#### **3.2.4.3** Remarks:

1. With the experimental values of  $\chi_M$  a linear fit for  $1/\chi_M$  as a function of T can be made. From the measurements in the temperature domain  $T > \theta$  we can extrapolate the "paramagnetic Curie temperature  $\theta$ ". Be aware that the latter does not forcedly coincide with the ferromagnetic transition point  $T_C$ . In the ferromagnetic region arises "spontaneous magnetisation"  $M_S$ . For  $T \to 0$  it amounts to  $M_S(T \to 0) = ng_i j\mu_i$ .

- 2. For a negative field factor  $\lambda$  the Curie-temperature  $\theta$  is also negative, but the transition from para- to antiferromagnetic occurs at a positive temperature  $T_N$  – the so-called Néel point. The  $\chi_M(T)$ -curve exhibits a maximum at  $T_N$ . According to a fairly rough approximation made by Néel,  $T_N$  is located at about  $-\theta$ . Experimentally, it has been found that  $-\theta/T_N \in [1.5, 5]$ . In the literature, a derivation for the curie constant can be found to arrive at an expression similar to (32).
- 3. The experimental values for  $\theta$  are way bigger than what would correspond to a dipole-dipole interaction. According to Heisenberg, between nearby paramagnetic atoms comes a quantum mechanical exchange interaction, which is a consequence of Pauli's exclusion principle.

# 3.2.5 Van Vleck's quantum mechanical theory of paramagnetism (1932)

In many ions of the rare earth elements, the quadratic term (quadratic in H) of the expansion (17) plays a relevant role. The Larmor-Langevin diamagnetic susceptibility is not going to be discussed here.

In the section 3.2.3 (about Langevin's theory of paramagnetism) we assumed that the ions are in the ground state, so that the moments induced by H start giving a contribution to the magnetic susceptibility at not too high temperatures.

The susceptibility of a set composed by n atoms with a magnetic moment  $m = -\partial E/\partial H$  is given (according to (14)) by

$$\varkappa = n \cdot \frac{\langle m \rangle_T}{H} = -\frac{n}{H} \cdot \frac{\sum \frac{\partial E}{\partial H} e^{\frac{E}{kT}}}{e^{\frac{E}{kT}}}.$$
(41)

The energy of every level is representable as a power series in H in the following form:

$$E_{j,m}(H) = E_0^{(j)} + W_1^{(j,m)} \cdot H + W_2^{(j,m)} \cdot H^2 + \dots$$
(42)

The index j refers to the multiplet j, and m denotes the levels of the same multiplet, which is degenerate in the absence of a field. Let again  $\vec{H} = (0, 0, H)^T$ , so that the coefficients  $W_i^{(j,m)}$  can be written as follows:

$$W_{1}^{(j,m)} = -\mu_{0}\mu_{B} \langle j, m | L_{z} + 2S_{z} | j, m \rangle$$

$$W_{2}^{(j,m)} = -\mu_{0}\mu_{B} \sum_{j' \neq j} \sum_{m'} \frac{|\langle j, m | L_{z} + 2S_{z} | j', m' \rangle|^{2}}{E_{j'} - E_{j}}$$
(43)

Assumption: The magnetisation is proportional to the magnetic field, so that no saturation occurs; that is, the temperature is big enough for guaranteeing, that

$$W_1^{(j,m)} \cdot H + W_2^{(j,m)} \cdot H^2 \ll kT.$$

So we are allowed to expand the exponential in (41) and keep only the linear terms (in H). So we get the following expression for the susceptibility:

$$\varkappa = -\frac{n}{H} \cdot \frac{\sum_{j,m} \left( W_1^{(j,m)} + 2W_2^{(j,m)} \cdot H \right) \left( 1 - \frac{W_1^{(j,m)} \cdot H}{kT} \right) e^{-\frac{E_0^{(j)}}{kT}}}{\sum_{j,m} \left( 1 - \frac{W_1^{(j,m)} \cdot H}{kT} \right) e^{-\frac{E_0^{(j)}}{kT}}}$$
(44)

By symmetry of the problem (space isotropy)  $\sum_{m} W_1^{(j,m)}$  vanishes for any j – think about it! The susceptibility is finally given by

$$\varkappa = -\frac{n}{kT} \cdot \frac{\sum_{j,m} \left( W_1^{(j,m)^2} - 2kTW_2^{(j,m)} \right) e^{-\frac{E_0^{(j)}}{kT}}}{\sum_j e^{-\frac{E_0^{(j)}}{kT}} \cdot \omega_j},$$
(45)

where  $\omega_j$  is the multiplicity of the level j. This is the general formula by Van Vleck, which is valid as long as H is small enough.

#### Particular cases:

**3.2.5.1 Large multiplet-splitting compared to** kT: Only the groundmultiplet is appreciably occupied, thus we recover the Langevin-formula (31)

$$\varkappa = \mu_0 \cdot \frac{ng_j^2 \mu_B^2 j(j+1)}{3kT} + n\alpha.$$
(46)

The presence of the second term  $n\alpha$  in (46) is connected to the Zeeman effect of second order. It is a constant contribution to the susceptibility, which is independent from the temperature, and has the following value:

$$n\alpha = \mu_0 \frac{n\mu_B^2}{6(2j+1)} \left( \frac{F(j+1)}{E_{j+1} - E_j} - \frac{F(j)}{E_j - E_{j-1}} \right)$$
(47)

with the abbreviation

$$F(j) = \frac{1}{j} \left[ (s+l+1)^2 - j^2 \right] \cdot \left[ j^2 - (s-1)^2 \right]$$
(47<sup>1</sup>)

In general, this second term is small and is thus neglected. Nevertheless, it can show up, as we will see in 3.2.5.4.

**Remark:** In opposition to the classical case, the magnetic moment of the atom does not exist in the beginning; in order to make it appear we need to apply a magnetic field, with which we measure it. This is a consequence of the realistic character of quantum mechanics: the existence of a physical quantity only has meaning in an environment that this quantity can be measured in.

**3.2.5.2 High magnetic field:** If the magnetic field is high enough and the temperature is low enough, we can observe saturation effects. The expansion (44) is not valid anymore, and a more precise approximation yields for the magnetisation the value of equation (22):

$$M = ng_j j\mu_B B_j(x) \quad \text{with} \quad x = \mu_0 g_j j\mu_B \frac{H}{kT}$$
(22)

**3.2.5.3 Small multiplet-splitting with respect to** kT: Given the splitting between the levels is small compared to kT, then the exponential function in formula (45) can be substituted by 1. On the other hand, the small splitting energy implies that the coupling between orbital and spin moment is small in comparison with their coupling with the magnetic field. Like in the case of the Paschen-Back effect, both moments in the field are quantized separately (first approximation!): every magnetic level is determined by  $m_l$  and  $m_s$ . Thus

$$W^{(l)}(m_l, m_s) = -\mu_0 \mu_B(m_l + m_s),$$

and the susceptibility is given by

$$\varkappa = \mu_0 \cdot \frac{N\mu_B^2}{3kT} \cdot \left[l(l+1) + 4s(s+1)\right].$$
(48)

This susceptibility is the one given by a mix of orbital and spin magnetic moment. In this case we talk about Van Vleck's paramagnetism of low frequencies.

**3.2.5.4** Multiplet splitting comparable to kT: At room temperature we can observe this case especially in samarium- and europium-bindings. Basing on an even more general equation than (45), Van Vleck (1932) was able to calculate the susceptibility of these two three-valued ions. The total number of atoms n gets distributed in groups at given values of the quantum number j:  $\sum n_j = n$ . Further we assume, that

$$n_j = n(2j+2)e^{-\frac{E_0^{(j)}}{kT}}.$$

As a result we gain the following formula for the susceptibility:

$$\varkappa = \frac{\sum_{j=|l-s|}^{l+s} \left[\frac{\mu_0 g_j^2 \mu_B^2}{3kT} \cdot j(j+1) + \alpha_j\right] (2j+1) e^{-\frac{E_0^{(j)}}{kT}}}{\sum_{j=|l-s|}^{l+s} (2j+1) e^{-\frac{E_0^{(j)}}{kT}}}$$
(49)

By dint of the Goudsmit's formula for the spin-orbit coupling it is possible to express the spin-orbit constant through a screening constant, and then to derive some usable expressions for the magnetic susceptibility. This contribution to the susceptibility is temperature dependent, and corresponds to the second term in (46). We talk about Van Vleck's paramagnetism at "high frequency".

# 4 Experimental methods

From the numerous known methods for determining the susceptibility of a sample we can pick three amongst the most important principles:

# 4.1 Induction

Let the sample be in the homogeneous magnetic field of a pickup coil. If the sample is abruptly removed or the magnetic field changed, in the coil will be produced a voltage-kick, which will be proportional to M or, respectively, to  $\mu$ . To measure that we need very sensitive instruments, though. Gauging measurements with known substances are not necessary, in the case of a simple, well-known geometry of the sample.

# 4.2 Torque

This method is principally suited for analysing anisotropic substances, since it allows the measurement of the diagonal elements of the tensor  $\varkappa$  in the main axis system. The mono-crystal sample is hung in the homogeneous magnetic field so, that a main axis is vertical (hence parallel to the field). The torque acting on the sample will thus be proportional to the difference between the two horizontal main susceptibilities. Absolute measurements presuppose additionally the determination of  $(\varkappa_1 + \varkappa_2 + \varkappa_3)/3$  with a pulverized sample, for example with the Gouy method.

# 4.3 Force in an inhomogeneous field (Faraday)

This method (which is by far the most used) allows to make very precise relative determinations of  $\varkappa$ , since a force acting on a sample hung in an inhomogeneous field is proportional to the susceptibility. If the field gradient where the sample is placed is unknown, the proportionality constant has to be determined with a gauging measurement. In our experiment we are going to apply a variant of this principle – the Gouy method – which will be discussed in the next chapter.

# 5 Samples and measurement setup

# 5.1 The Gouy method



Figure 1: The layout of the Gouy method.

#### 5.1.1 Description

In this method the substance we want to examine has to be in the form of a long cylindrical pipe, and must be hung to a balance between the pole shoes of an electromagnet. The force exerted on the sample will be given by the gradient of the magnetic energy U (see eqn. (19)):

$$\vec{K} = \vec{\nabla}U = \mu_0 \vec{\nabla} \left( \int \mathrm{d}V \cdot \frac{1}{2} \left( \varkappa - \varkappa_{air} \right) H^2 \right) \approx \frac{\mu_0}{2} \vec{\nabla} \left( \int \mathrm{d}V \cdot \varkappa H^2 \right), \quad (50)$$

where:

 $\varkappa =$  Volume susceptibility of the sample  $\varkappa_{air} =$  Volume susceptibility of the air surrounding the sample (neglected) dV = Volume element of the sample H = Magnetic field intensity

#### 5 SAMPLES AND MEASUREMENT SETUP

Let us call A the constant cross section of our sample. According to equation (50), the vertical force (in z-direction) will thus be given by

$$K_{z} = \frac{\mu_{0}}{2} \frac{\partial}{\partial z} \left( \int_{z_{u}}^{z_{o}} \mathrm{d}z \cdot \varkappa H^{2} A \right) = \frac{\mu_{0}}{2} \varkappa A \left( H_{o}^{2} - H_{u}^{2} \right).$$
(51)

This force can be measured with the balance as in the arrangement shown in figure 1, where we can observe the weight variation of the sample when turning the electromagnet on.

We want to determine  $\chi_M$  with our weighting-method. From the definitions of  $\varkappa$  and  $\chi_M$  follows:

$$\varkappa = \chi_M \frac{\rho}{[\text{Molecular weight}]} \tag{52}$$

If we insert this into equation (51) and take into account, that for a prismatic sample the density  $\rho$  is given by

$$\rho = \frac{m}{V} = \frac{m}{A \cdot L},\tag{53}$$

where:

m = Mass of the sample

A = Cross section of the sample

L = Length of the sample

we then get:

$$K_{z} = \mu_{0} \frac{\rho}{[\text{Molecular weight}]} \cdot \chi_{M} \cdot \frac{1}{2} A \left(H_{o}^{2} - H_{u}^{2}\right)$$
$$= \mu_{0} \frac{m}{[\text{Molecular weight}] \cdot 2L} \cdot \chi_{M} \left(H_{o}^{2} - H_{u}^{2}\right)$$
(54)

or

$$K_z = a \cdot \chi_M$$
 with  $a = \mu_0 \frac{m}{2L \cdot [\text{Molecular weight}]} \left(H_o^2 - H_u^2\right).$  (55)

Since  $H_o$  and  $H_u$  are normally unknown, *a* must be determined with a gauging measurement. Nevertheless, in this experiment will be given a parameter, which will make this unnecessary.

#### 5.1.2 Remarks about our experiment

In our experiment the susceptibility can be determined starting both from experimental and from theoretical quantities, and thus the constant a – which depends strongly from the setup – can be calculated (see chapter 7).

#### 5 SAMPLES AND MEASUREMENT SETUP

The value of *a* is approximately constant in our temperature range. The temperature dependence of the susceptibility  $\chi_M = \chi_M(T)$  is thus given exclusively by the downforce  $K_z = K_z(T)$ :

$$\chi_M(T) = const \cdot K_z(T) \tag{56}$$

Formulae (54) and (55) were derived assuming the validity of equation (53), which is respected only for prismatic samples. It is anyway easy to convince ourselves, that it holds for a pulverized sample, if the powder is filled homogeneously in a quartz pipe. Furthermore equations (55) and (56) are independent of the cross section A of the sample, assuming that the magnetic field is constant across the whole cross section at any height z.

## 5.2 Composition of the equipment

The magnetic field is generated with a direct-current magnet with current stabilisation. The force has to be measured with a normal analytical balance.

An oven allows one to perform measurements in the interval between room temperature and 950 K. The measurements of the temperature are executed with a digital thermometer.

# 6 Measurements

## 6.1 Sample and mounting



Figure 2: The hanging of the sample.

The substance to be investigated is Dysprosium Oxide  $(Dy_2O_3)$  in pulverized form, and was placed in a carefully cleaned quartz pipe, which was then closed under vacuum by fusing its end. The samples cannot be heated beyond 650°C.

The sketch in figure 2 shows the hanging of the sample. With quartz bars of different lengths we can regulate the height of the sample in the apparatus. The precise placement can be done with a hook with a screw thread. The height must be adjusted so, that the force acting on the sample is maximal. **Take care**: The quartz glass pipes break very easily!

The part of  $K_z$  which arises from the quartz must be measured with an empty pipe placed at the same height. In a first approximation, this contribution to  $K_z$  is temperature-independent. Quartz (silicon dioxide) is diamagnetic. The measured values of  $K_z$  must be corrected appropriately.

## 6.2 The weighing scale

The analytical balance was originally set with a scale pan, whose weight has to be compensated with tare weight (figure 2). A user guide for the balance is to be found at the workplace.

Any changes to the weight hanging from the balance should be made exclusively in the "AUS" ("off") mode! The position "HALB" ("half") is then used for the adjustment of the gram- and tenth-of-gram measurement. The fine adjustments and measurement is then made in the "EIN" ("on") po-

sition.

One should observe the green measurement scale when unblocking the weight ("HALB" or "EIN" mode) for oscillations. If there are no smooth oscillations initially, then it is likely the sample has gotten caught on something. If this happens, re-block the balance and carefully move it until the sample is placed exactly in the middle of the oven.

The green milligram scale of the balance has a 200 mg scope. The same weight can thus be measured in two different adjustments of the hundredthgram-indicator. If the magnetic field is turned off, you must set the indicator to the highest of the two possible values, such that, after turning the magnet on, you will not have to change it (which would change the height of the sample).

**Important:** The sample must hang freely in the oven!

## 6.3 Oven and temperature measurement

The oven is movable. It must be slid on so, that the sample is in the middle, since the upper and lower part of the oven-tube are composed of badlyconducting nickel silver and only the central part guarantees a homogeneous heating of the sample. For the construction of the oven no ferromagnetic materials were used.

Under the oven is fixed an acrylic glass tube, in order to prevent the convective air currents to falsify the measurements. The weight of the sample with the magnetic field off has to be checked before every measurement because air convection and water condensation cause small deviations, despite the protection tubes.

The current through the oven – and thus the dissipated power and, consequently, the temperature – can be regulated within a variable transformer. Its external part is cooled with water.

Make sure that the temperature stays constant during the measurement of a value because we are interested in the dynamics during thermodynamic equilibrium.

## 6.4 The electromagnet

As a power supply we use a digital rectifier (whose instructions can be found at the workplace). The magnet is driven by an adjustable direct voltage source between 0 V and 150 V. The current in the magnet can be quasicontinuously regulated in the interval between 0 and 10 A; it is stabilized, in order to avoid fluctuations of H. The maximal permitted current is:

- Persistently: 5 A
- Only for short time: 10 A

The magnetic field strength is proportional to the current intensity. As a part of the assignment you have to check this: what could eventual corrections be due to?

### 6 MEASUREMENTS

The magnet is protected by water cooling. Built-in relays prevent it to work without cooling. The values for the tension and the current can be read on the instruments. The grouped potentiometers allow a good reproducibility of the measurements.

#### Important:

- When turning the device on, the voltage must be set to zero! The potentiometer on the left of the front panel must be completely turned to the anticlockwise direction. The voltage can be increased only when the device is on.
- In order to prevent damage to the rectifier, an ultra-fast fuse is built into the front panel. If this melts, do absolutely **not** replace it by a normal one!

# 7 Data evaluation and analysis

# 7.1 Verification of ferromagnetic contamination and downforce as a function of the current

For the values taken at 290 K evaluate the quotient

$$\alpha \coloneqq \frac{K_z(4 \text{ A})}{K_z(6 \text{ A}) \cdot K_z(8 \text{ A})},$$

and for the ones taken at 800 K evaluate:

$$\alpha' \coloneqq \frac{K'_z(4 \text{ A})}{K'_z(6 \text{ A}) \cdot K'_z(8 \text{ A})}.$$

In the case there are no ferromagnetic contaminations, we should get  $\alpha = \alpha'$ , while ferromagnetic substances show  $\alpha \neq \alpha'$  (why?).

Plot graphically  $K_z = K_z(I)$ ,  $K'_z = K'_z(I)$ , and also  $K_z = K_z(I^2)$ ,  $K'_z = K'_z(I^2)$ . Does the data fit your expectations?

# **7.2** Calculation of $\chi_M$ and a

By equations (39), (40), and (56) we have

$$a \cdot K_z = \chi_M = \mu_0 \cdot \frac{N \cdot p^2 \mu_B^2}{3k(T-\theta)} = \frac{C_M}{T-\theta}$$

where N is the number of magnetic atoms per kMol. Normally a is determined with a gauging measurement. We can extrapolate  $\theta$  from a linear fit of  $(1/K_z)(T)$ , and calculate  $C_M$ , where we assume for Dy<sub>2</sub>O<sub>3</sub>:

$$p_{exp.} = p_{theor.} = 10.6 \qquad (Why?)$$

Actually, experimental determinations for p may yield different values.

 $C_M$  and  $\theta$  allow the calculation of  $\chi_M$  and a at T = 290 K.