11 Magnetism and low temperatures

MAGNETIC INTERACTIONS are of considerable interest throughout much of physics; they are of particular importance in the study of matter at low temperature and provide also the means for attaining extremely low temperatures. Before leaving the subject of systems in thermal equilibrium, we shall therefore devote some attention to the application of thermodynamic ideas to these topics.

The study of a macroscopic system at very low temperatures provides an opportunity for investigating this system when it is in its ground state and in the quantum states which lie very close to it. The number of such states accessible to the system, or its corresponding entropy, is then quite small. The system exhibits, therefore, much less randomness, or a much greater degree of order, than it would at higher temperatures. The low temperature situation is thus characterized by a fundamental simplicity* and by the possibility that some systems may exhibit in striking fashion a high degree of order on a macroscopic scale. An example of such order is provided by a system of spins all of which, at sufficiently low temperatures, become aligned parallel to each other, thus giving rise to ferromagnetism. A more spectacular example is provided by liquid helium, which remains a liquid down to absolute zero (provided that its pressure is not increased above 25 atmospheres). Below a critical temperature of 2.18°K (the so-called "lambda point") this liquid becomes "superfluid"; it then exhibits completely frictionless flow and can pass through extremely small holes with the greatest of ease. Another set of spectacular examples is provided by many metals (e.g., lead or tin) which become "superconducting" below characteristic sharply defined critical temperatures. The conduction electrons in these metals then exhibit completely frictionless flow with the result that the metals become perfect conductors of electricity (with strictly zero dc electrical resistivity) and manifest striking magnetic properties. We refer the interested reader to the references at the

* There is at least simplicity in principle, since the task of understanding the nature of the ground state of a many-particle system may, at times, be far from trivial.

end of this chapter for more detailed discussions of these remarkable properties. The foregoing comments should, however, be sufficient to indicate why the field of low temperature physics has become a well-developed active field of current research.

It is worth inquiring just how close to its ground state a macroscopic system can be brought in practice, i.e., to how low an absolute temperature it can be cooled. The technique is to insulate the system at low temperatures from its room temperature surroundings by enclosing it in a "dewar." (This is a glass or metal vessel which provides thermal insulation by a double-walled construction; a vacuum maintained between these walls minimizes heat conduction by residual gases and proper polishing of the walls minimizes heat influx due to radiation.)* Helium is the gas which liquefies at the lowest temperature, at 4.2°K under atmospheric pressure. The temperature of the liquid can be readily reduced further to about 1°K simply by pumping away the vapor over the liquid and thus reducing its pressure to the lowest practically feasible value.† Thus it is quite easy by modern techniques to bring any substance to 1°K simply by immersing it in a heat bath consisting of liquid helium. By using liquid He^3, the liquid consisting entirely of the rare isotope He^3 (normally constituting less than 1 part in 10^6 of ordinary helium, which consists almost entirely of He^4), one can apply similar methods to attain fairly readily temperatures down to 0.3°K. Appreciably greater effort and different techniques are necessary in order to work at still lower temperatures. By using a method (to be discussed in Sec. 11·2) which involves the performance of magnetic work by a thermally isolated system of spins, it is feasible to attain temperatures as low as 0.01°K or even 0.001°K. Extensions of this method have even made it possible to reach 10^{-6}°K!

After these general remarks about low-temperature physics and some of its connections with magnetism, we are ready to turn to a specific discussion of magnetic systems. Any subject involving electromagnetism raises immediately the question of choice of units. Since we are discussing problems in physics rather than in electrical engineering, we shall use the units which are currently in most common use in the physics journals of all countries, namely Gaussian cgs units. We recall that in these units all electrical quantities (such as current and voltage) are measured in electrostatic units, while magnetic quantities (such as magnetic field or magnetization) are measured in gauss.

11 · 1 *Magnetic work*

We consider a system of volume V in an externally applied field $\mathbf{H}_0$. The system might, for example, be a sample consisting of a magnetic solid. In

* An ordinary thermos bottle is a familiar example of a dewar.

† The principle of the method should be familiar to any hiker ambitious enough to have cooked out of doors. The boiling point of water on a mountain top is reduced below that at sea level because of the reduced atmospheric pressure.

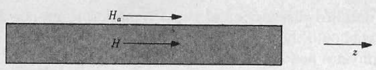


***Fig. 11·1·1** A long cylindrical sample in the presence of an externally applied magnetic field H_a. Here $H = H_a$ and $\bar{M}_0 = \chi H$.*

order to avoid uninstructive complications and problems of detail which are predominantly in the realm of electromagnetic theory, we shall focus attention on a physically simple situation. We assume that the externally applied field $\boldsymbol{H}_a$, even when it varies in space, is substantially uniform over the volume of the relatively small sample. We further assume that the sample is in the shape of a cylinder which is very long compared to its cross-sectional dimensions, and that it is always kept oriented parallel to the direction of $\boldsymbol{H}_a$. Then the mean magnetic moment per unit volume $\bar{\boldsymbol{M}}_0 = \bar{\boldsymbol{M}}/V$ is essentially uniform throughout the sample and parallel to $\boldsymbol{H}_a$. (These properties would also be true for any ellipsoidal sample.) In addition, if $\boldsymbol{H}$ denotes the magnetic field inside the sample, $\boldsymbol{H} = \boldsymbol{H}_a$ by virtue of the boundary condition that tangential components of $\boldsymbol{H}$ must be continuous. We also recall that quite generally the magnetic induction $\boldsymbol{B}$ is related to $\boldsymbol{H}$ by the relation

$$\boldsymbol{B} = \boldsymbol{H} + 4\pi\bar{\boldsymbol{M}}_0 \qquad (11\cdot1\cdot1)$$

Outside the sample where $\bar{\boldsymbol{M}}_0 = 0$, $\boldsymbol{B} = \boldsymbol{H}_a$. The magnetic susceptibility χ per unit volume of the sample is defined by the ratio $\chi \equiv \bar{M}_0/H$ so that (11·1·1) can also be written

$$\boldsymbol{B} = \mu'\boldsymbol{H} = (1 + 4\pi\chi)\boldsymbol{H} \qquad (11\cdot1\cdot2)$$

where μ' is called the magnetic permeability of the sample.

The starting point for applying macroscopic arguments of statistical thermodynamics to such a magnetic system is again the fundamental relation (3·9·6)

$$đQ = T\,dS = d\bar{E} + đW \qquad (11\cdot1\cdot3)$$

valid for any quasi-static process. Here the system is, in general, characterized by two external parameters, the volume V and the applied magnetic field $\boldsymbol{H}_a$. Hence the total work $đW$ done by the system includes not only the mechanical work $\bar{p}\,dV$ done by the pressure in a volume change dV but also the magnetic work $đW^{(m)}$ associated with changes in $\boldsymbol{H}_a$. We proceed to derive an expression for this magnetic work.

To keep the geometry simple by making the problem one dimensional, we suppose that the applied magnetic field H_a points in the z direction and that the cylindrical sample is always oriented parallel to this direction. Then the magnetic field H inside the sample (and its magnetic moment $\bar{M}$) also points in the z direction and $H = H_a$. Suppose then that the sample is in a particular state r, where its total magnetic moment is M_r, and that the external magnetic field $H_a = H$ at the position of the sample is changed slowly by a small amount. The work done in this process cannot depend on just how the field is changed.

Let us therefore imagine that the magnitude of the applied field is not quite uniform in space, but that it vanishes at infinity and varies gradually so as to attain the value H_a in the region of experimental interest. The magnetic field then exerts on the sample a force having a component $F_x = M_r(\partial H/\partial x)$ in the x direction (see Fig. 11·1·2). The magnetic field at the position of the sample can now be changed by moving the sample slowly from a position x where $H = H(x)$ to a neighboring position $x + dx$ where $H = H(x + dx)$. In this process one must exert on the sample a force $-F_x$ in the x direction and must do *on* the sample an amount of work $đ\mathcal{W}_r^{(m)}$ which goes to increase the energy of the sample in this state by an amount dE_r. Thus

$$đ\mathcal{W}_r^{(m)} = dE_r = (-F_x)\,dx = \left(-M_r\frac{\partial H}{\partial x}\right)dx$$

or*
$$đ\mathcal{W}_r^{(m)} = dE_r = -M_r\,dH \qquad (11\cdot1\cdot4)$$

Thus
$$M_r = -\frac{\partial E_r}{\partial H} \qquad (11\cdot1\cdot5)$$

i.e., the magnetic moment is the "generalized force" conjugate to the magnetic field regarded as an external parameter. Taking the statistical average of (11·1·4) over an equilibrium statistical ensemble of similar systems, one then obtains for the macroscopic magnetic work $dW^{(m)}$ done *by* the sample when the field in which it is located changes by an amount dH the result

$$dW^{(m)} = -đ\mathcal{W}^{(m)} = \bar{M}\,dH \qquad (11\cdot1\cdot6)$$

* Note that this expression justifies the familiar result $E_r = -M_rH$ for the energy of a magnetic moment of *fixed* size in an external field H.

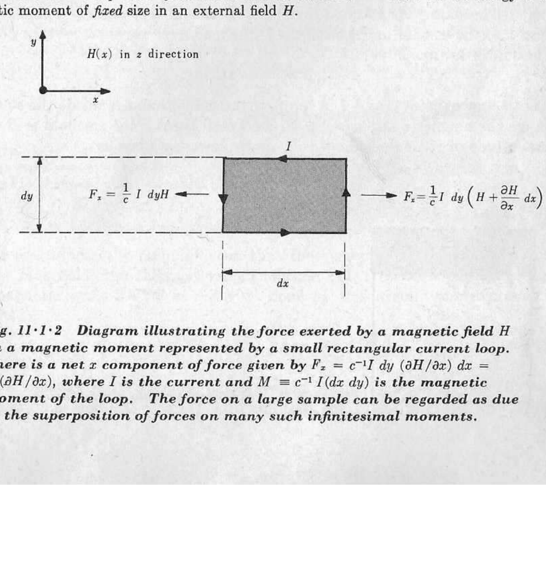


***Fig. 11·1·2** Diagram illustrating the force exerted by a magnetic field H on a magnetic moment represented by a small rectangular current loop. There is a net x component of force given by $F_x = c^{-1}I\,dy\,(\partial H/\partial x)\,dx = M(\partial H/\partial x)$, where I is the current and $M = c^{-1}I(dx\,dy)$ is the magnetic moment of the loop. The force on a large sample can be regarded as due to the superposition of forces on many such infinitesimal moments.*

where $\bar{M}$ is the total mean magnetic moment of the sample. Hence the fundamental thermodynamic relation (11·1·3) can be written

$$T\,dS = d\bar{E} + \bar{p}\,dV + \bar{M}\,dH \tag{11·1·7}$$

where the last two terms represent the total work done by the sample in a general infinitesimal quasi-static process.

The relation (11·1·7) can, as usual, be rewritten in a variety of other forms. For example, if it is desired to consider $\bar{M}$ rather than H as an independent variable, one can write $\bar{M}\,dH = d(\bar{M}H) - H\,d\bar{M}$, so that (11·1·7) becomes

$$T\,dS = d\bar{E}^* + \bar{p}\,dV - H\,d\bar{M} \tag{11·1·8}$$

where $\bar{E}^* \equiv \bar{E} + \bar{M}H$ is the analog of some kind of enthalpy. The thermodynamic consequences of (11·1·8) or (11·1·7) are, of course, equivalent; the essential content of these relations is that both $d\bar{E}$ and $d\bar{E}^*$ are exact differentials of well-defined quantities characteristic of the macrostate of the system.

Alternative point of view There is another way in which one can calculate the magnetic work. Imagine that the sample is placed inside a close-fitting solenoid whose length l and area A are then equal to those of the sample so that $lA = V$, the volume of the sample. The solenoid is supposed to consist of N turns of wire and to have negligible electrical resistance. It can be connected to a source of emf (e.g., a battery) as shown in Fig. 11·1·3. Work must be done by the source of emf on the system consisting of the coil and sample in order to produce the desired magnetic field. The reason is that, in trying to change the magnetic field inside the coil, a counter-emf $\mathcal{V}$ is induced across the coil. The source of emf must then provide an emf $\mathcal{V}$ to overcome this induced emf. If the current in the circuit is I, the magnetic work $\bar{d}\mathcal{W}'^{(m)}$ thus done by the source in time dt is

$$\bar{d}\mathcal{W}'^{(m)} = \mathcal{V}I\,dt \tag{11·1·9}$$

Let us now express $\mathcal{V}$ and I in terms of the fields B and H inside the solenoid. Since the magnetic flux passing through each turn of the solenoid is BA, the magnitude of the induced emf is given by Faraday's law as

$$\mathcal{V} = \frac{1}{c} N \frac{d}{dt}(AB) \tag{11·1·10}$$

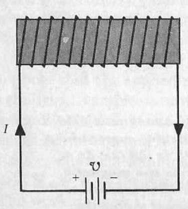


***Fig. 11·1·3** A magnetic sample placed inside a solenoid.*

where the constant c is the velocity of light (since we use Gaussian units) and where B is expressed in gauss and $\mathcal{V}$ in statvolts. Also, by Ampere's circuital theorem, H inside the solenoid satisfies the relation

$$Hl = \frac{4\pi}{c}(NI) \tag{11·1·11}$$

Hence (11·1·9) becomes

$$\bar{d}\mathcal{W}'^{(m)} = \left(\frac{NA}{c}\frac{dB}{dt}\right)\left(\frac{c}{4\pi}\frac{l}{N}H\right)dt = \frac{Al}{4\pi}H\,dB$$

or

$$\bar{d}\mathcal{W}'^{(m)} = \frac{V}{4\pi}H\,dB \tag{11·1·12}$$

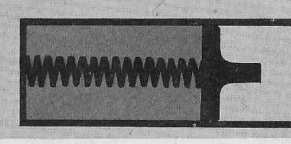
Using (11·1·1) and $\bar{M} = V\bar{M}_0$ this becomes

$$\bar{d}\mathcal{W}'^{(m)} = \frac{V}{4\pi}H(dH + 4\pi\,d\bar{M}_0) = d\left(\frac{VH^2}{8\pi}\right) + H\,d\bar{M} \tag{11·1·13}$$

This expression represents the work necessary to magnetize the sample *and* to establish the magnetic field; i.e., it is the work done *on* the system consisting of the sample *plus* the magnetic field. On the other hand, (11·1·6) represents the work done on the sample in some given magnetic field, i.e., it is the work done on the system consisting of the sample alone. It is, of course, equally legitimate to consider either the sample alone, or the sample plus electromagnetic field, as the system of interest.

The following analogy may serve to clarify the situation. Consider a gas and a spring contained in a cylinder, as shown in Fig. 11·1·4. It is possible to consider the gas as the system of interest. In this case the spring is part of the environment capable of doing work on this system. Alternatively, one can consider the gas plus the spring as the system of interest. The potential energy of the spring is then part of the internal energy of this system.

***Fig. 11·1·4** A gas and a spring contained within a cylinder closed by a movable piston.*



If one adopts the point of view that the system of interest consists of sample plus field, the thermodynamic relation (11·1·3) becomes, using for the magnetic work $dW'^{(m)} \equiv -\bar{d}\mathcal{W}'^{(m)}$ done *by* this system the expression (11·1·13),

$$T\,dS = d\left(\bar{E}' - \frac{VH^2}{8\pi}\right) + \bar{p}\,dV - H\,d\bar{M} \tag{11·1·14}$$

where $\bar{E}'$ denotes the mean energy of this system. Putting $\bar{E}^* \equiv \bar{E}' - VH^2/8\pi$, this relation is identical with (11·1·8) and thus equivalent to (11·1·7). This shows explicitly that the thermodynamic consequences of our discussion are the same irrespective of which system one chooses to consider.

****Remark*** It is instructive to exhibit in detail the equivalence of the expressions (11·1·6) and (11·1·13) for magnetic work. To show this explicitly, consider the situation illustrated in Fig. 11·1·3. Suppose one starts with $H = 0$ and an unmagnetized sample with $\bar{M} = 0$. What then is the work $\mathcal{W}$ which one must do to reach the final situation where the field is H_0 and the magnetic moment of the sample is $\bar{M}(H_0)$? By using the reasoning leading to (11·1·13), one gets

$$\mathcal{W} = \frac{VH_0^2}{8\pi} + \int_0^{H_0} H\, d\bar{M} \qquad (11\cdot1\cdot15)$$

Let us now look at the problem from a different point of view. Imagine that one starts with $H = 0$, and that the sample with $\bar{M} = 0$ is located at infinity. The final situation can then be brought about in the following steps:

1. Turn on the field H_0 inside the coil.
2. Bring the sample from infinity into the coil, magnetizing it in the process. This requires work for two reasons:
   a. Work must be done, for *fixed* current I_0 in the coil (i.e., for *fixed* H_0), to move the sample into the coil against the forces exerted on it by the field.
   b. Work must be done by the battery to keep the current I_0 constant even though an emf is induced in this coil by the moving magnetized sample which produces a changing flux through the coil.

By (11·1·13) the work done in step (1) is simply

$$\mathcal{W} = \frac{VH_0^2}{8\pi} \qquad (11\cdot1\cdot16)$$

The work done *on* the sample in step (2*a*) is given by (11·1·6) so that

$$\mathcal{W} = -\int_0^{H_0} \bar{M}(H)\, dH \qquad (11\cdot1\cdot17)$$

Finally in step (2*b*), where $H = H_0$ is maintained constant, the work done by the battery is given by (11·1·12)

$$\mathcal{W} = \frac{V}{4\pi} H_0(B_f - B_i) \qquad (11\cdot1\cdot18)$$

where B_i is the initial and B_f the final value of the magnetic induction inside the coil. But when the sample is initially at infinity, $B_i = H_0$; when the sample is finally inside the coil, (11·1·1) yields $B_f = H_0 + 4\pi\bar{M}(H_0)/V$. Hence (11·1·18) becomes

$$\mathcal{W} = \frac{V}{4\pi} H_0 \frac{4\pi\bar{M}(H_0)}{V} = H_0\bar{M}(H_0) \qquad (11\cdot1\cdot19)$$

Adding the three works (11·1·16), (11·1·17) and (11·1·19), one gets then

$$\mathcal{W} = \frac{VH_0^2}{8\pi} - \int_0^{H_0} M(H)\, dH + M(H_0)H_0 \qquad (11\cdot1\cdot20)$$

Integration by parts shows that this is indeed identical to (11·1·15).

11 · 2 *Magnetic cooling*

Since it is possible to do work on a sample by changing the applied magnetic field, it is also possible to heat or cool a thermally insulated sample by changing a magnetic field. This provides a commonly used method to attain very low temperatures. The nature of this method can be made clearer by comparing it with a more familiar mechanical analogue. Suppose that it is desired to cool a gas by means of mechanical work. One can proceed in the manner illustrated in the top part of Fig. 11·2·1. The gas is initially in thermal contact with a heat bath at temperature T_i, e.g., with a water bath. One can now compress the gas to a volume V_i. In this process work is done on the gas, but it can give off heat to the bath and thus remains at the temperature T_i after equilibrium has been reached. The gas is then thermally insulated (e.g., by removing the water bath) and is allowed to expand quasi-statically to some final volume V_f. In this adiabatic process the gas does work at the expense of its internal energy and, as a result, its temperature falls to some final value T_f less than T_i.

The method of magnetic cooling is very similar and is illustrated in the bottom part of Fig. 11·2·1. The system of interest is a magnetic sample initially in thermal contact with a heat bath at temperature T_i. In practice this heat bath is liquid helium near 1°K, thermal contact of the sample with the bath being established by heat conduction through some helium gas at low pressure. One can now switch on a magnetic field until it attains some value H_i. In this process the sample becomes magnetized and work is done, but the sample can give off heat to the bath and thus remains at the temperature T_i after equilibrium has been reached. The sample is then thermally insulated (e.g., by pumping off the helium gas which provided the thermal contact with the bath) and the magnetic field is reduced quasi-statically to a final value H_f (usually $H_f = 0$).* As a result of this "adiabatic demagnetization" the temperature of the sample falls to some final value T_f less than T_i. In this way temperatures as low as 0.01°K can readily be attained. Indeed, temperatures close to 10^{-6}°K have been achieved by elaboration of this method.

Let us now analyze the method in greater detail in order to understand how the temperature reduction comes about. The first step is an isothermal process: here the system is kept at a constant temperature T_i while it is brought from some macrostate *a* to some other macrostate *b* by a change of external parameter. The second step is an adiabatic process: here the system is thermally isolated and is then brought quasi-statically from the macrostate *b* to a macrostate *c* by a change of external parameter. The entropy S of the system therefore remains constant in this last step. The whole method is then most conveniently illustrated in a diagram of entropy S versus temperature T. Such a diagram is shown schematically in Fig. 11·2·2 for a paramagnetic sample where the significant external parameter is the magnetic field H. For such a sample the entropy S becomes smaller when the individual atomic

* Internal equilibrium is usually attained rapidly enough that reducing the field to zero in a few seconds is sufficiently slow to be considered quasi-static.