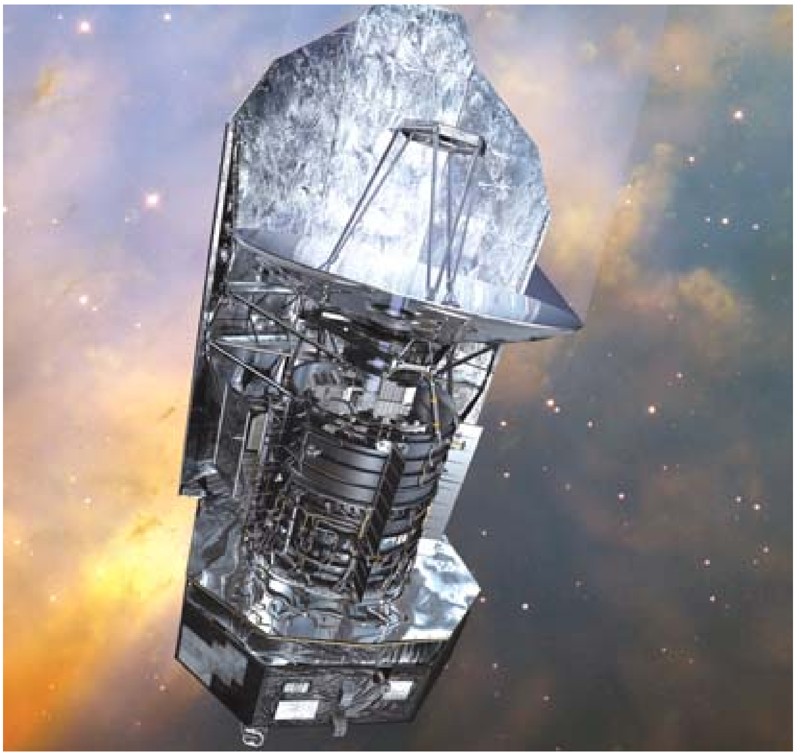
Adiabatic demagnetisation refrigeration for subkelvin cooling in space

The CEA is developing an ultra-low-temperature refrigeration system capable of operating in microgravity environments, and of meeting the reliability and robustness demands specific to spaceborne applications. The demand in this sector is now for satellite sensors that operate at increasingly low temperatures.

Satellite designers are now asking for 'refrigerators' capable of providing the ultra-low temperatures required to prevent detectors being affected by interfering radiation. The latest generation of sensors require temperatures of around 300 **millikelvins** (**mK**) (0.3 degrees above absolute zero, i.e. -272.85°C) in order to achieve the level of precision required for their mission. The **European Space Agency** (**ESA**) is finalising preparation of the Herschel space observatory. Scheduled for launch in 2008, the Herschel's key objectives are to collect data on the universe with the aim of investigating the genesis of stars and galaxies. The Herschel space observatory comprises three instruments: PACS, SPIRE and HIFI. The CEA has now delivered the refrigerators for the first two instruments, which are the crowning achievement to a committed research and development effort that has extended over a decade. These refrigerators are able to continuously produce 260 mK for a period of over 50 hours (Box).
And that is not all! The ESA is currently setting the framework for its forthcoming missions: in terms of refrigeration, the demand is strongly focused on the delivery of systems designed to operate at ultra-low temperatures. The latest generations of **superconducting** sensors exploit the properties of superconductivity and an operating temperature of around 50 mK. The CEA, in anticipation of this demand, has already begun working on the development of a system capable of producing these 50 mK in a space environment.

Seventy-five years of research in magnetic refrigeration

There are very few technologies capable of producing temperatures below 100 millikelvins (0.1 degrees above absolute zero). The *Pomeranchuk cell* produces cold by pressurising solid **helium**-3 (a rare helium **isotope**). David M. Lee, Douglas D. Osheroff and Robert C. Richardson used this cell for their research into the properties of helium-3, which won them the 1996 Nobel Prize in Physics. Other examples include *dilution refrigerators* which produce cold by diluting helium-3 in helium-4 (the most naturally abundant helium isotope), which could otherwise be described as the evaporation of the helium-3 isotope in a helium-4 'vacuum'. This is the most versatile solution, usually

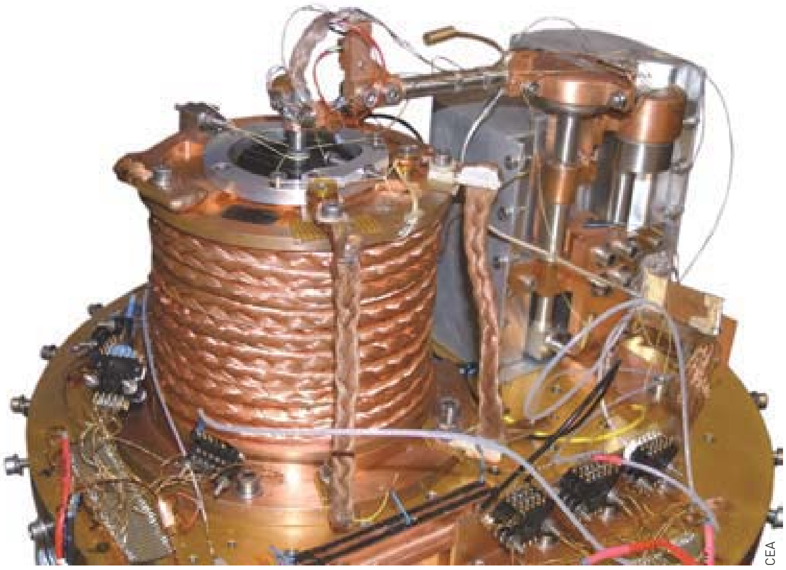

ESA

The Herschel satellite is scheduled to be placed in orbit in late 2008 by an Ariane 5 launcher at the same time as the Planck satellite.

The adsorption refrigerator

An **adsorption** refrigerator is based on a simple operating principle: a liquid's temperature depends on its vapour pressure[1]. The liquid can be cooled by lowering this pressure. By pumping liquid **helium** (4.2 **kelvins**), the temperature can be lowered to around 700 millikelvins (mK) (0.7°C above absolute zero). By pumping helium-3, liquid under 3.2 K, the temperature can be lowered to around 200 mK (0.2°C above absolute zero). The CEA has developed a pumping principle that makes use of a particular property of solids, i.e. their ability to trap gas molecules at their surface. Activated carbon has a particularly large specific surface area (covering about 1,000 m²/g), which makes it a highly effective helium trap at 4.2 K and, therefore, a first-rate pump with the added advantage of operating at a cold-fixed temperature, and of being exceptionally compact and efficient.

(1) Vapour pressure: pressure exerted by the vapour phase above a liquid or solid phase of a substance when these two phases are at equilibrium. It thus represents the maximum value of the pressure of this substance required for it to release vapour and is directly proportional to its temperature.



One of the laboratory's latest refrigerator prototypes, capable of producing 48 mK.

the preferred choice of researchers seeking to produce ultra-low temperatures in the laboratory. *Magnetic refrigeration* is one of just a handful of technologies capable of cooling down to a few millikelvins.

The pioneering experiments were carried out in 1933 by two independent teams, Giauque and MacDougall and Haas, Wiersma and Kramers. These experiments were the first to achieve ultra-low temperatures, thus paving the way to a new area of research in physical sciences. This technology was later edged out in the laboratory by the advent of dilution refrigerators, capable of producing continuous cold and maintaining high-scale cold power, even at the very lowest temperatures. In their standard form, however, dilution refrigerators are not suited to **microgravity** environments. This is because the dilution effect integral to the helium-3/helium-4 demixing(1) interface, which is crucial to refrigerator operation, only works under gravity. Researchers at the **CNRS** have therefore been working to develop a system that exploits capillary forces. One of these refrigerators equips the Planck satellite, which is scheduled for launch at the same time as the Herschel space observatory. Researchers, however, have already turned their attention to developing the next generation of dilution refrigerators as current models will not meet the requirements of future European space missions.

Space missions impose fairly drastic constraints, particularly in terms of reliability, footprint, weight, mechanical strength and power consumption. There is renewed interest in magnetic refrigeration technology since there is every likelihood that future space missions will require operating temperatures below 100 millikelvins.

Magnetic refrigeration technology does not rely on gravity and is only dependent on the supply of electrical power. This gives such systems the added advantage of not having any moving parts, thus eliminating any risk of wear that could undermine reliable operation. Magnetic refrigeration is therefore a solution well-geared to missions in space, despite two major drawbacks which will be discussed later on.

The principle of adiabatic demagnetisation

Cooling generally involves reducing the **entropy** of a system. Magnetic refrigeration is no exception, and relies on exploiting changes in magnetic entropy.

Take the example of a **paramagnetic** material. This material consists in a set of **magnetic moments** that can be likened to tiny magnets the size of an atom or a **molecule**. Like magnets, magnetic moments are characterised by a vector quantity and an intensity factor. Magnetic moments interact with each other within a material just like ordinary magnets. Paramagnetism is characterised by the weakness of this interaction. Molecular agitation (**thermal agitation**) prevails, with the result that the magnetic moments take up random orientations thus creating disorder, i.e. entropy. This entropy is related to the number of possible configurations the moments can take (defined by **quantum** physics).

Now let's place the paramagnetic material in an external **magnetic field** (generated by a **coil** for example). The material's magnetic moments will align themselves in the direction of the magnetic field, just as a magnetic compass aligns itself with the Earth's magnetic field. The weakness of the moment interactions actually makes this easier. The stronger the field, the stronger the alignment of the magnetic moments. This has the effect of reducing the amount of disorder in the system and, therefore, its entropy. This is called an **exothermic** transformation, i.e. the *magnetocaloric effect*. In practice, the material is connected to an isothermal bath which evacuates the heat generated during magnetisation.

Now let's fit the material with heat insulation and reduce the magnetic field. The magnetic moments will return to their random orientation, a transition that requires a certain input of energy. Now that the system has been insulated, the magnetic moments will extract thermal energy, triggering a drop in the material's temperature; this is the principle underlying **adiabatic** demagnetisation (Figure 1).

The technology

In technical terms, a demagnetisation stage comprises a coolant, a paramagnetic salt, and a means of produ-

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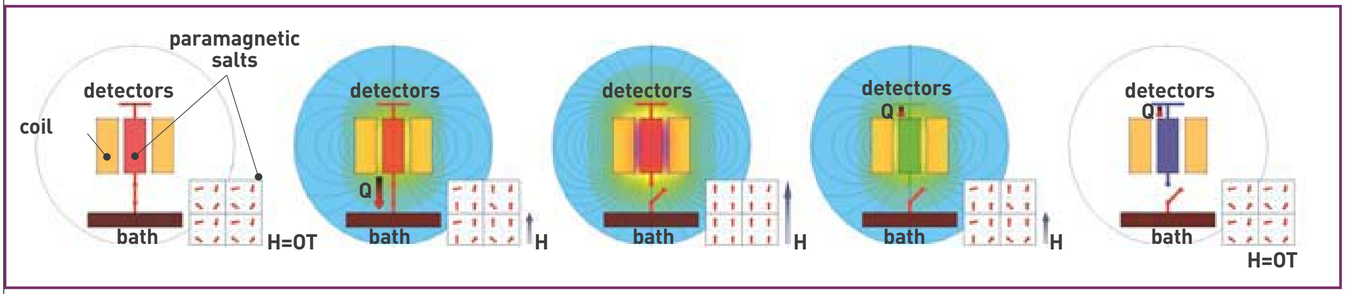
(1) Demixing: phenomenon involving the micro-separation of phases with different compositions in an alloy when one of the mixture's solutes exceeds its solubility threshold.

Figure 1.
The principle of adiabatic demagnetisation.



cing a variable magnetic field. The choice of coolant depends on the operating temperature range planned for the stage. Between temperatures of 4 and 1 kelvins, there are **garnets**, whose magnetism derives from **rare earth** elements such as gadolinium or dysprosium, which can be purchased as **single crystals**. Below 1 kelvin, the situation gets a little more complicated. There are several possible coolants to choose from, most of which are alums (double sulphates) whose magnetism derives from iron, chromium or cerium **ions**. Their usefulness stems from the fact that they contain water. The water molecules act as 'cages' that insulate the magnetic ions from each other, making it possible to obtain very weak interactions and produce coolants able to reach exceptionally low temperatures of just a few millikelvins (Figure 2). These water molecules, however, are also the source of alum's main drawbacks: the alum has to be sealed to prevent water loss and any consequent damage. Another drawback is their very low melting point (39°C for iron alum).

Superconducting coils are the technology currently used to generate magnetic fields in **cryogenic** environments. If the field is to be produced at the intensity required for successful demagnetisation, engineers either have to increase the number of current loops, thus adding to the weight of the coil, or engineer a higher current input. Space missions are constrained by limited mass and power budgets meaning that engineers have to find a compromise between mass and current input. The problem here is that at higher operating temperatures the demagnetization stage will require stronger magnetic fields, making it increasingly difficult to find a compromise.

As part of its forthcoming space observatory project called Xeus(2), the European Space Agency recently issued a call for bidders to fund the development of a coolant solution able to continuously produce 50 mK from a baseline temperature of 2.5 K and with a total mass below 5 kg. The most efficient cascade-system adiabatic demagnetisation refrigerator currently available, which was developed by **NASA**'s Goddard Space Flight Centre in Greenbelt (Maryland), lies just above this weight limit. Indeed, engineers are finding it is virtually impossible to meet the specifications using a process based solely on adiabatic demagnetization.

Two adiabatic demagnetization stages

The CEA took up the ESA's call by proposing to develop the two adiabatic demagnetisation stages (one to serve mainly as a 50-mK heat reservoir) coupled with an **adsorption** refrigerator, similar to those already developed for the Herschel space observatory (Figure 3). This refrigerator is able to pre-cool the demagnetisation stage to a temperature of around 350 millikelvins, yet is also extremely compact and lightweight. The 'active' part of the Herschel refrigerators only weighs about 300 g and is built to withstand the vibrations of the Ariane 5 rocket launcher. Thus, the total refrigerator system only weighs in at just 1.8 kg. Pre-cooling the two demagnetisation stages makes it possible to significantly cut back on magnetic field strength requirements, thus keeping mass and coil current values within acceptable limits.

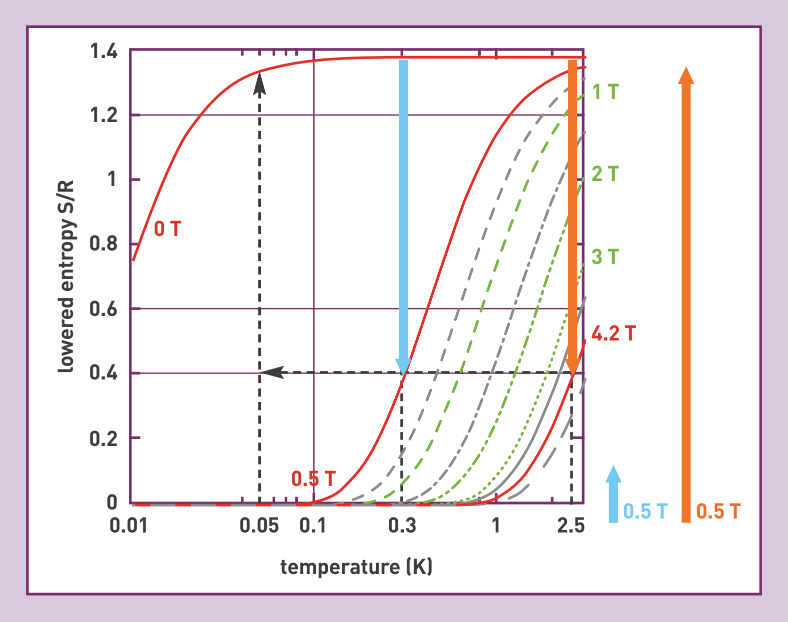


Figure 2.
Theoretical magnetic entropy curves for chromium-potassium alum. The values of the magnetic field shown by the arrows are the fields required to lower the entropy of a given quantity (for a given efficiency at 50 mK) between 2.5 K and 300 mK.

Engineers finally settled on a chromium-potassium alum (CPA) coolant that combines high performance with relative robustness (melting point: 89°C). This is a **polycrystalline** alum. The thermal conductivity of the complete system is limited by the weak conduction at **grain boundaries**. This means engineers have to create a 'thermal bus' in order to thermalise the crystals. The CEA-developed thermal bus comprises 260 copper wires with diameter 0.25 mm and length 80 mm, for a total **conductance** of 0.3 µW/mK at 50 mK. The paramagnetic salts are recrystallised directly onto this copper wire network in order to minimise thermal contact resistances. When the salts recrystallise they exploit the weak solubility of CPA in water: the thermal bus is immersed in a saturated solution at 50°C. Cooling the bus to 0°C while maintaining the temperature of the solution ensures the salts recrystallise on the wires. The next step is the tricky task of sealing the coolant without exceeding its melting point.

The coolant is then attached to its coil core using Kevlar®

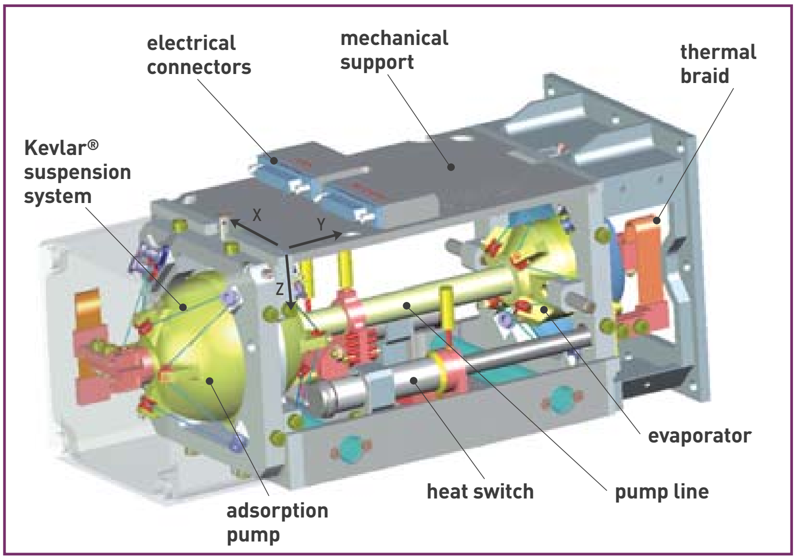


Figure 3.
3D view of the adsorption refrigerator equipping the Herschel satellite. The model earmarked to pre-cool the adiabatic demagnetisation stage will be of a similar design.

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http://www.sciops.esa.int/index.php?project=XEUS

Heat tests on the back-up cryocooler for the Herschel satellite at the Low Temperatures Department at the CEA Grenoble centre.

F. Vigouroux/CEA

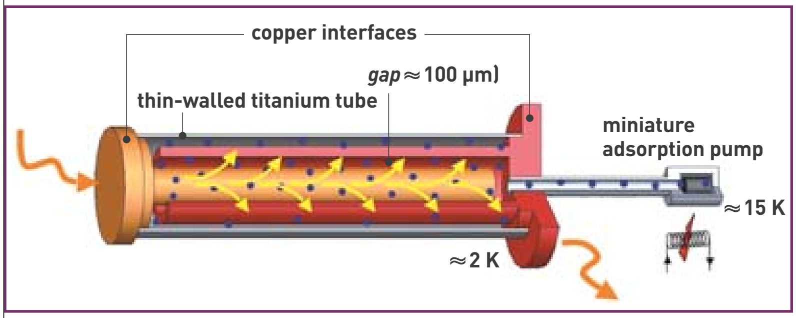


Figure 4.
How the heat switch works. When the miniature adsorption pump is cold, the chamber is emptied meaning that heat can only pass via the thin-walled tube. When the mini-pump is heated, the tube fills with helium, thus enabling the conduction of heat between the copper interfaces through the gas contained in the gap.

fibres that provide the dual advantage of great mechanical strength and low thermal conductivity. This means the salt can be attached sufficiently firmly to resist the vibrational forces of the launcher while providing the coil with thermal insulation. Researchers at the laboratory carried out an in-depth study on Kevlar fibres, which had previously been included in the design of the Herschel cryocoolers. Kevlar is one of the rare materials that expands on cooling. A creep(3) mechanism was observed during thermal cycling (succession of cooling and reheating phases) that reduced the tension of the attachment fibres. This could generate a risk of breakage during the rocket launch phase. The CEA has developed a Kevlar® tensioning process that makes it possible to counteract this effect and thereby ensure the fibres hold tight during the launch.

With the latest prototype, consisting of a single demagnetisation stage, engineers have been able to achieve a performance of 32 mK. The brief now is to add a second stage at 50 mK, which will serve as an 'active' magnetic reservoir. This means that the next key priority will be to install a heat switch(4) capable of operating at this temperature. No multi-stage refrigerator can operate without this kind of device, as each stage has to be thermally coupled to and disengaged from its adjacent stage in turn. The footprint of the complete refrigerator system hinges on the performance of this heat switch. The laboratory has developed a solution that makes use of a 'gas-gap' heat switch which boasts excellent performance up to around 300 millikelvins (Figure 4). The team's next task will be to design a system capable of operating at 50 millikelvins; they plan to bring out a functional engineering prototype within the next couple of years.

Two novel applications in view

Xeus has the potential to carry two instruments which would require a continuous production of 300 and 50 millikelvins, respectively. The CEA is in a position to offer a robust and reliable solution for each instrument: an adsorption refrigerator with a proven track record, coupled with its magnetic refrigeration solution.

At the same time, **JAXA**, the Japanese Space Agency, is mounting another scientific mission, SPICA(5), which may include a European-built instrument, the ESI, in its payload. At its current stage of development, the ESI is posting the same continuous cryogenic requirement of 50 millikelvins. The CEA is liaising on this issue with the **Rutherford Appleton Laboratory** which is supervising the project's research component, with a special focus on the thermal design module.

With these two missions in mind, the CEA is currently seeking to leverage the strengths of its adiabatic adsorption/demagnetisation refrigerator system with the aim of equipping the satellites.

> Nicolas Luchier
Institute for Nanoscience and Cryogenics (Inac)
Physical Sciences Division
CEA Grenoble Centre

(3) Creep: change over time of the plastic deformation of a material subjected to a prolonged mechanical stress.

(4) A heat switch is a device capable of opening or closing a heat conduction process depending on whether it is set to its 'on' or 'off' value.

(5) http://www.ir.isas.jaxa.jp/SPICA/index.html

FOCUS A

The different types of magnetism

The origins of magnetism lie in the properties of electrons as explained by the laws of quantum physics. Part of an electron's magnetic properties (*spin magnetism*) results from its quantum-mechanical spin state, while another part results from the orbital motion of electrons around an atom's nucleus (*orbital magnetism*) and from the magnetism of the nucleus itself (nuclear magnetism). This is put to use, in particular, for nuclear magnetic resonance imaging in the medical field. Magnetism is therefore produced by electric charges in motion. The force acting on these charges, called the Lorentz force, demonstrates the presence of a magnetic field.

Electrons have an intrinsic magnetic dipole moment (the magnetic quantum state being the Bohr *magneton*), which can be pictured as an electron's rotational motion of spin around itself in one direction or another, oriented either upwards or downwards. The *spin quantum number* (one of the four numbers that 'quantifies' the properties of an electron) equals 1/2 (+ 1/2 or - 1/2). A pair of electrons can only occupy the same *orbital* if they have opposite magnetic dipole moments.

Each atom acts like a tiny magnet carrying an intrinsic magnetic dipole moment. A nucleus (the neutron and proton individually have a half-integer spin) will have a half-integer spin if it has an odd atomic mass number; zero spin if the atomic mass number and charge are even, and an integer spin if the atomic mass number is even and the charge odd.

On a larger scale, several magnetic moments can together form magnetic domains in which all these moments are aligned in the same direction. These spatial regions are separated by domain walls. When grouped together, these domains can themselves form a macroscopic-scale magnet (Figure E1).

The type of magnetism that comes into play is determined by how these elementary constituents are ordered, and is generally associated with three main categories of material: *ferromagnetic*, *paramagnetic* and *diamagnetic*.

Any material that is not diamagnetic is by definition paramagnetic provided that its magnetic susceptibility is positive. However, ferromagnetic materials have particularly high magnetic susceptibility and therefore form a separate category.

1. Ferromagnetic materials are formed of tiny domains inside which atoms exhibiting parallel magnetisation tend to align themselves in the direction of an external magnetic field like elementary dipoles. In fact, the magnetic moments of each atom can align themselves spontaneously within these domains, even in the absence of an external magnetic field. Applying an external field triggers domain wall movement that tends to strengthen the applied field. If this field exceeds a certain value, the domain most closely oriented with the direction of the applied field will tend to grow at the expense of the other domains, eventually occupying the material's whole volume. If the field diminishes, the domain walls will move, but not symmetrically as the walls cannot fully reverse back to their original positions. This results in remanent magnetisation, which is an important feature of naturally occurring magnetite, or of magnets themselves.

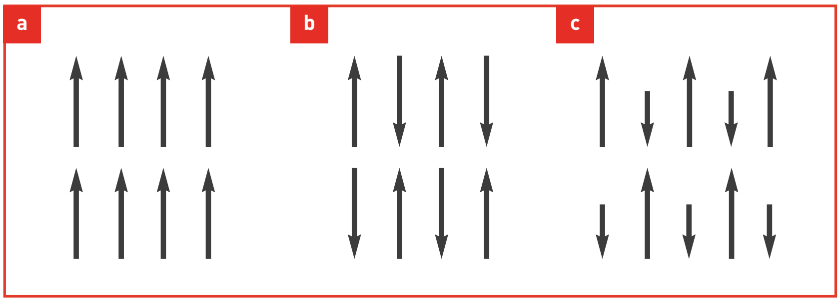


Figure E1.
Intrinsic magnetic dipole moments have parallel alignment in ferromagnetic materials (a), anti-parallel alignment but zero magnetisation in antiferromagnetic materials (b), and anti-parallel alignment with unequal moments in ferrimagnetic materials (c).

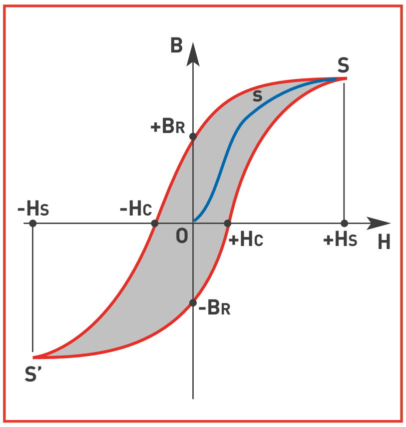


Figure E2.
The induction B of a magnetic material by a coil is not proportional to its magnetic excitation (*field H*). While the initial magnetisation forms an OsS-type curve, shown in blue in the figure, it reaches saturation at point s. Only a partial induction is retained if the field approaches zero; this remanent induction can only be cancelled out by reversing the magnetic field to a "coercive" field value. This hysteresis loop illustrates the losses due to "friction" between the magnetic domains shown on the area bounded by the magnetisation and demagnetisation curves.

The whole process forms a hysteresis loop, i.e. when the induced field is plotted against the applied field it traces out a *hysteresis curve* or *loop* where the surface area represents the amount of energy lost during the irreversible part of the process (Figure E2). In order to cancel out the induced field, a coercive field has to be applied: the materials used to make artificial permanent magnets have a high coercivity.

Ferromagnetic materials generally have a zero total magnetic moment as the domains are all oriented in different directions. This ferromagnetism disappears above a certain temperature, which is known as the Curie Temperature or Curie point.

The magnetic properties of a given material stem from the way the electrons in the metallic cores of a material or of a transition metal complex collectively couple their spins as this results in all their spin moments being aligned in the same direction.

Materials whose atoms are widely distributed throughout their crystal structure tend to better align these elementary magnets via a coupling effect. This category of materials, which is characterised by a very high positive magnetic

Stoiber Productions, München

A Transrapid train using magnetic levitation arriving at the Long Yang bus station in Shanghai (China). This German-built high-speed, monorail train was commissioned in 2004 to service the rail link to Pudong international airport.

susceptibility, includes iron, cobalt and nickel and their alloys, steels in particular, and some of their compounds, and, to a lesser extent, some rare earth metals and alloys with large crystal lattices, and certain combinations of elements that do not themselves belong to this category. In ferrimagnetic materials, the magnetic domains group into an anti-parallel alignment but retain a non-zero magnetic moment even in the absence of an external field. Examples include magnetite, ilmenite and iron oxides. Ferrimagnetism is a feature of materials containing two types of atoms that behave as tiny magnets with magnetic moments of unequal magnitude and anti-parallel alignment. Antiferromagnetism occurs when the sum of a material's parallel and anti-parallel moments is zero (e.g. chromium or haematite). In fact, when atoms are in a close configuration, the most stable magnetic arrangement is an anti-parallel alignment as each magnet balances out its neighbour so to speak (Figure E1).

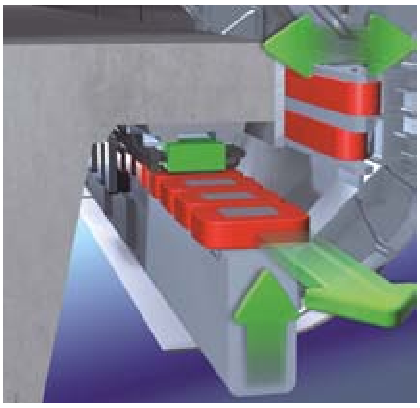
2. Paramagnetic materials behave in a similar way to ferromagnetic materials, although to a far lesser degree (they have a positive but very weak magnetic susceptibility of around 10^{-3}). Each atom in a paramagnetic material has a non-zero magnetic moment. In the presence of an external magnetic field, the magnetic moments align up, thus amplifying this field. However, this effect decreases as temperature rises since the thermal agitation disrupts the alignment of the elementary dipoles. Paramagnetic materials lose their magnetisation as soon as they are released from the magnetic field. Most metals, including alloys comprising ferromagnetic elements are paramagnetic, as are certain minerals such as pegmatite.

3. Diamagnetic materials exhibit a negative and an extremely weak magnetic susceptibility of around 10^{-5}. The magnetisation induced by a magnetic field acts in the opposite direction to this field and tends to head away from field lines towards areas of lower field strengths. A perfect diamagnetic material would offer maximum resistance to an external magnetic field and exhibit zero permeability. Metals such as silver, gold, copper, mercury or lead, plus quartz, graphite, the noble gases and the majority of organic compounds are all diamagnetic materials.

In fact, all materials exhibit diamagnetic properties to a greater or lesser extent, resulting from changes in the orbital motion of electrons around atoms in response to an external magnetic field, an effect that disappears once the external field is removed. As Michael Faraday showed all that time ago, all substances can be "magnetised" to a greater or lesser degree provided that they are placed within a sufficiently intense magnetic field.

Electromagnetism

It was the Danish physicist Hans Christian Ørsted, professor at the University of Copenhagen, who, in 1820, was first to discover the relationship between the hitherto separate fields of electricity and magnetism. Ørsted showed that a compass needle was deflected when an electric current passed through a wire, before Faraday had formulated the physical law that carries his name: the magnetic field produced is proportional to the intensity of the current. Magnetostatics is the study of static magnetic fields, i.e. fields which do not vary with time.



Close-up of the magnets used to guide and power the train.

Magnetic and electric fields together form the two components of electromagnetism. Electromagnetic waves can move freely through space, and also through most materials at pretty much every frequency band (radio waves, microwaves, infrared, visible light, ultraviolet light, X-rays and gamma rays). Electromagnetic fields therefore combine electric and magnetic force fields that may be natural (the Earth's magnetic field) or man-made (low frequencies such as electric power transmission lines and cables, or higher frequencies such as radio waves (including cell phones) or television.

Mathematically speaking, the basic laws of electromagnetism can be summarised in the four Maxwell equations (or Maxwell-Lorentz equations) which can be used to provide a coherent description of all electromagnetic phenomena from electrostatics and magnetostatics to electromagnetic wave propagation. James Clerk Maxwell set out these laws in 1873, thirty-two years before Albert Einstein incorporated the theory of electromagnetism in his *special theory of relativity*, which explained the incompatibilities with the laws of classical physics.