Louis Néel: forty years of magnetism

Michel Prévot, David Dunlop

LOUIS NEEL: FORTY YEARS OF MAGNETISM

Michel Prévot[1] et David Dunlop[2]

[1] Laboratoire de Géophysique, Tectonique et Sédimentologie, CNRS et Université de Montpellier 2, 34095 Montpellier Cedex 05,France. Adresse courriel : prevot@dstu.univ-montp2.fr
[2] Geophysics, Department of Physics, University of Toronto, Toronto, Canada M5S 1A7. Email address : dunlop@physics.utoronto.ca

It was in 1928, at the Institut de Physique de Strasbourg directed by Pierre Weiss, that Louis Néel undertook his first investigations of magnetism. At that time, theoretical magnetism was dominated by Weiss' (1907) molecular field theory. According to this theory, below their Curie temperatures, ferromagnetic materials can be considered as paramagnets whose magnetic moments are strongly coupled by a fictitious magnetic field proportional to the spontaneous magnetization of the substance. Above the Curie point, the substance becomes paramagnetic, the inverse of the susceptibility thus being constrained to obey the linear Curie-Weiss law. The molecular field is enormous, of the order of several hundred Tesla. Why then is it easy to change the magnetism, or even demagnetize, a piece of ordinary iron using quite small magnetic fields? To explain this puzzling observation, Weiss hypothesized that magnetic matter is subdivided into elementary domains. The directions of magnetization within domains and the boundaries between them could be changed by weak or moderate fields.

Many questions remained unanswered, however. A large number of metals have a positive susceptibility that is independent of temperature – «paramagnétisme constant». In the ferrites, of which magnetite is the type example, inverse susceptibility varies with temperature in hyperbolic fashion. Above all, even for archetypal ferromagnets like iron and nickel, the variation of inverse susceptibility with temperature only becomes linear about a hundred degrees above the Curie point. Extrapolating this linear segment gives a second Curie point, called the paramagnetic Curie point, ten or so degrees higher than the first. Néel himself, while a student at Strasbourg, measured and pointed out some of these experimental discrepancies, which had been glossed over by previous researchers in their desire to «verify» Weiss' theory. Néel's independence of mind and strength of character were thus in evidence from the very beginning of his career.

In his thesis work, Néel first focused his attention on the enigma of the two Curie points. Weiss had supposed that the molecular field was uniform. In 1928, just as Néel was beginning his work, Heisenberg demonstrated that coupling between magnetic moments was the result of exchange interaction between electrons, and that exchange coupling would decrease exponentially with increasing distance between the atoms. Néel therefore decided to replace the uniform molecular field of Weiss by a local molecular field, defined at the atomic scale and varying in time and space (Néel, 1957). But rather than taking a quantum mechanical approach, he preserved the simple approximation of classical molecular field theory.

The article which he published in 1932, drawn from his thesis of the same year, demonstrated the rich possibilities of this approach (Néel, 1932). In it, he showed that thermal agitation produces temporal fluctuations of the local molecular field and that in these fluctuations lies the explanation of the two Curie points. The spatial variations of the molecular field, on the other hand, are the cause of the hyperbolic variation of inverse susceptibility above the Curie point in ferrite solid solutions like magnetite. Still in the same article, he studied the case of negative interactions, showing that thermal fluctuations lead to a temperature-independent paramagnetism at low temperatures.

A few years later, he carried out an extensive study of the variation of exchange interactions with interatomic distance (Néel, 1936a) and returned to a more rigorous theoretical treatment of substances with negative interactions (Néel, 1936b). In this latter note, little more than a page long, was born the model of antiferromagnetism (or «paramagnétisme constant» as it was then known) as two interleaved ferromagnetic sublattices, the spontaneous magnetization of each obeying the equations of the molecular field approximation. Néel thus demonstrated, in a more general way than in his previous work, that antiferromagnetic ordering generates a constant paramagnetism from absolute zero to a temperature, now called the Néel temperature, that plays the same role as the Curie temperature in ferromagnetic ordering. This brief paper marks the discovery of the fourth magnetic state of matter, antiferrromagnetism (as named by Bitter in 1939).

The Second World War brought this era in Néel's career to an end. The Faculté des Sciences of Strasbourg University relocated in 1939 to Clermont-Ferrand. Néel went instead to the Institut Polytechnique de Grenoble, and in Grenoble he would spend the rest of his career. In 1946, just after the end of the war, the Centre National de la Recherche Scientifique created within the University of Grenoble the «Laboratoire d'Electrostatique et de Physique du Métal» under Néel's direction.

On arriving in Grenoble, Néel set to work on an interpretation of the Rayleigh laws of magnetization, discovered in 1887 but never explained theoretically. A simple approach allowed him to explain these laws for a substance containing multiple domains and to suggest a physical basis for the Preisach-Néel diagram (Néel, 1942, 1943). He then became interested in the role of the «dispersion field», the internal demagnetizing field due to local deviations in the direction of the spontaneous magnetization or to the effect of inclusions. He showed that non-magnetic inclusions of sufficient size give rise to new, arrowhead-shaped domains, now called «Néel spikes» (Néel, 1944a). The same year, in an article he considered one of his most original (Néel, 1991), he was able to show that different modes of magnetization can be recognized according to the number of magnetic phases present (Néel, 1944b). This approach allowed him to make the first theoretical predictions of the shapes and sizes of elementary domains.

Néel next established the foundations of a new general theory of coercive force (Néel, 1946) based on the existence of fluctuations in the direction or intensity of the spontaneous magnetization within domains. The fluctuations in direction are caused by elastic perturbations of the lattice, while the intensity fluctuations result from chemical inhomogeneities, inclusions or cavities. He showed that the dispersion field energy resulting from these fluctuations plays a fundamental role in domain wall propagation. Numerical values predicted by this theory agree well with measured coercive forces of many alloys. In another paper (Néel, 1948b), he showed that dispersion fields likewise play an important role in the approach to magnetization saturation.

Néel was always interested in the applications of magnetism, notably applications in earth science. During an extended visit in 1931 to the Institute and Observatory of Earth Physics in Clermont-Ferrand, where he was considering becoming director, he learned of the observations by Brunhes around 1900 of lavas at the summit of the Puy de Dôme (Massif Central) whose remanent magnetization is reversed to that of the Earth's present magnetic field. He considered this phenomenon of reversed magnetization tremendously exciting («passionnant»: Néel, 1991) and envisaged a major study involving a substantial investment in people and resources. Unfortunately it proved difficult to justify the necessary investment, and eventually he decided to abandon geophysics and return to Strasbourg.

Nevertheless, the explosive development of rock magnetism and paleomagnetism after the war, in France most notably the work of Emile Thellier, could hardly fail to attract his attention. In 1947, Néel established that iron in grains smaller than 32 nm in size is single-domain and has a high coercive force (Néel, 1947). It was in a geophysics journal, Annales de Géophysique, that two years later he published his fundamental study of the magnetism of single-domain grains (Néel, 1949), endeavouring in particular to explain quantitatively the experimental results of Thellier's group. Studying the effect of thermal fluctuations, he discovered the phenomenon of superparamagnetism and calculated the lower limit of magnetic stability of single-domain iron to be 16 nm.

It is the considerable energy due to thermal fluctuations that explains how the weak magnetic field of the Earth is able to leave an imprint in rocks on grains whose coercive forces can be as much as 1000 times larger. Néel proposed the idea of relaxation time, now so fundamental in geology, and calculated that a mere doubling of the size of the grains would multiply the relaxation time by the enormous factor of 10^{10}. He studied in detail the properties of thermoremanent magnetization, total and partial, of isothermal remanence, and of viscous remanence, as well as the effect of treatment by heating in zero field and by alternating fields (the latter had not yet been introduced as a practical technique at the time). And because his theory was eminently testable by simple experiments, Néel proposed to rock magnetists the ultimate objective of determining the distribution of sizes and microscopic coercive forces of the magnetic grains in rocks. This challenge was taken up two decades later in the thesis research of one of us (Dunlop, 1968).

In 1951, Néel published the self-reversal hypothesis, that reversed magnetization in rocks might not always be due to reversals of the geomagnetic field (Néel, 1951b). For lavas, self-reversal of magnetization could occur during cooling, through magnetostatic interaction between two phases or by reversal of the spontaneous magnetization of a single phase. For sediments and possibly certain lavas, chemical alteration of the magnetic mineral in the course of time could lead to the same result. A matter of months later, Nagata and his colleagues discovered a real example of self-reversal, a lava from Mount Haruna (Japan) that acquired, in the laboratory, a thermoremanence reversed in direction to the field applied during cooling. Other (rare) examples are now known but the mechanism believed to govern the self-reversal is different from those proposed by Néel.

Néel's most influential paper in the English-speaking world was published (in English!) as a review of theoretical magnetism for rock magnetists and paleomagnetists in Advances in Physics (Néel, 1955). In it, he described the essentials of his theories of ferrimagnetism and of single-domain and multidomain grains. Although the only truly new aspect of the paper is his theory of thermoremanent magnetization of multidomain grains, the impact and lasting value of this paper cannot be overestimated. Like Kittel's review of about the same time, it is encyclopedic while being both succinct and clear. Not surprisingly, it is still frequently cited today.

Néel's (1949) theory ascribed magnetic viscosity in single-domain grains to thermal fluctuations. In massive material, Néel recognized two types of magnetic viscosity (Néel, 1951a). The first, like that of single-domain material, owes its origin to thermal fluctuations, in this case helping domain walls to cross barriers to their motion. Néel represented this effect by a «viscosity field» which increases as the logarithm of time. The intensity of viscous magnetization is then equal to the product of the viscosity field and the irreversible differential susceptibility of the material. The second type of viscosity, called diffusion after-effect (Néel, 1952), results from the redistribution of non-magnetic impurity atoms in the crystal lattice following modification by the applied field of the direction of the spontaneous magnetization. Unlike thermal fluctuation viscosity, it is manifested over a limited time interval following a field change.

In Néel's opinion, it was his work on the spinel ferrites which had the greatest impact and the most important practical applications. It was in 1948 that he published his fundamental memoir entitled «Les propriétés magnétiques des ferrites: ferrimagnétisme et antiferromagnétisme» (Néel, 1948a). In this paper, he generalized his theory of antiferromagnetic materials by supposing that the two magnetic sublattices A and B of ferrites have unequal moments, and introduced three molecular field coefficients representing interactions within the two sublattices (AA, BB) and between the sublattices (AB). He attributed these interactions to superexchange or indirect exchange between non-neighbouring magnetic atoms. His theory explained quantitatively the then-known properties of ferrites and predicted other novel behaviour, such as the possibility of unusual temperature variations of the spontaneous magnetization, which in certain cases can pass through zero and self-reverse in the course of cooling. Néel proposed also the existence of antiferromagnetic domains separated by walls analogous to ferromagnetic domain walls but requiring fields of 1 T or more to displace them.

In 1954, Néel suggested an explanation for the magnetic properties of rare earth garnets, which have considerable practical interest in high-frequency applications because they are good insulators. His proposal was a ferrimagnetic structure in which the ferric ions form a strongly interacting pair of sublattices, interacting weakly with a third sublattice containing the rare earth ions (Néel, 1954b). It was for «Fundamental research and discoveries in antiferromagnetism and ferrimagnetism which have important applications in solid-state physics» that the Nobel Prize in Physics was shared by Louis Néel in 1970.

In a departure from his earlier work, anisotropic coupling between neighbouring atoms was the subject of a groundbreaking article in 1954. Néel emphasized the existence in ferromagnetic bodies of a surface anisotropy energy which plays a potentially important role in ultrafine grains below 10 nm in size (Néel, 1954a). The importance of these effects has been highlighted in experimental research in recent years.

Néel also examined the conditions necessary for the appearance of directional order and its effects. These considerations were applied to the properties FeNi alloys with composition near 50:50, discovered in nature 15 years later as the mineral tetrataenite found in meteorites and lunar rocks. This substance possesses a magnetic superstructure of enormous anisotropy whose direction is determined by the ambient magnetic field (Néel et al., 1962). The presence of tetrataenite supports the hypothesis that meteorites containing this mineral originated in a large parent body whose cooling lasted millions of years, the time necessary to form the superstructure (Néel, 1991). Tetrataenite in meteorites also suggests the presence of a magnetic field whose direction was constant over this length of time, perhaps created by the parent body itself.

Louis Néel was born in Lyon on November 22, 1904 and died in Meudon (Paris) on November 17, 2000, in his 96th year. When we wrote to invite him to participate in the symposium organized in his honour at EGS 2000, he expressed a lively interest but declined to attend in person because of his age and failing health. He was delighted to learn that we geophysicists continue to be absorbed by the fascination of the magnetism of rocks and pointed to tetrataenite as an important unsolved problem to which we should direct our attention. This lively mind is now stilled but his scientific legacy will endure. We list below a selection of his most important papers, referred to in this introduction.

References

L. Néel (1932). Influence des fluctuations du champ moléculaire sur les propriétés magnétiques des corps. *Ann. de Phys.,* 17, 5-105.

L. Néel (1936a). Propriétés magnétiques de l'état magnétique et énergie d'interaction entre atomes magnétiques. *Ann. de Phys.,* 5, 232-279.

L. Néel (1936b). Théorie du paramagnétisme constant; application au manganèse. *C. R. Acad. Sc.,* 203, 304-306.

L. Néel (1942). Théorie des lois d'aimantation de Lord Rayleigh. 1ère partie: les déplacements d'une paroi isolée. *Cah. Phys.,* 12, 1-20.

L. Néel (1943). Théorie des lois d'aimantation de Lord Rayleigh. 2ère partie: multiples domaines et champ coercitif. *Cah. Phys.,* 13, 18-30.

L. Néel (1944a). Effet des cavités et des inclusions sur le champ coercitif. *Cah. Phys.,* 25, 21-44.

L. Néel (1944b). Les lois de l'aimantation et de la subdivision en domaines élémentaires d'un monocristal de fer. *J. Phys. Rad.,* 5, 241-279.

L. Néel (1946). Base d'une nouvelle théorie générale du champ coercitif. *Ann. Univ. Grenoble,* 22, 299-343.

L. Néel (1947). Propriétés d'un ferromagnétique cubique en grains fins. *C. R. Acad. Sc.,* 224, 1488-1490.

L. Néel (1948a). Propriétés magnétiques des ferrites. Ferrimagnétisme et antiferromagnétisme. *Ann. Phys.,* 3, 137-198.

L. Néel (1948b). La loi d'approche en a/H et une nouvelle théorie de la dureté magnétique. *J. Phys. Rad.,* 9, 184-192.

L. Néel (1949). Théorie du traînage magnétique des ferromagnétiques en grains fins avec application aux terres

cuites, *Ann. Géophys.*, 5, 99-136.
L. Néel (1951a). Le traînage magnétique. *J. Phys. Rad.,* 12, 339-351.
L. Néel (1951b). L'inversion de l'aimantation permanente des roches. *Ann. Géophys.*, 7, 90-102.
L. Néel (1952). Théorie du traînage magnétique de diffusion. *J. Phys. Rad.,*13, 249-263.
L. Néel (1954a). Anisotropie magnétique superficielle et surstructures d'orientation *J. Phys. Rad.,* 15, 225-239.
L. Néel (1954b). Sur l'interprétation des propriétés magnétiques des ferrites de terres rares. *C. R. Acad. Sc.,* 239, 8-11.
L. Néel (1955). Some theoretical aspects of rock magnetism. *Phil. Mag. Suppl.,* 4, 191-243.
L. Néel (1957) . Le champ moléculaire de Weiss et le champ moléculaire local. Colloque National de Strasbourg, commémoratif de l'œuvre de Pierre Weiss, 8-10 juillet 1957, CNRS éd., 24 pp.
L. Néel (1991). Un siècle de physique. Ed. Odile Jacob, Paris.
L. Néel, J. Paulevé, D. Dautreppe et J. Laugier (1962). Etablissement d'une structure ordonnée FeNi par irradiation aux neutrons. *C. R. Acad. Sc.,* 254, 965-968.