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Louis Néel: His multifaceted seminal work in magnetism

Louis Néel : son œuvre fondatrice à multiples facettes en magnétisme

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ABSTRACT

Louis Néel was a world-renowned scientist who devoted the research part of his multi-faceted career to magnetism. Covering roughly the period 1930–1970, his work is explained for a non-specialized audience, with particular attention given to work published in the *Comptes rendus hebdomadaires des séances de l'Académie des sciences*.

RÉSUMÉ

Louis Néel fut un savant de réputation mondiale. De ses activités multiples, celle consacrée à la recherche concerne principalement le magnétisme. Elle s'étend de 1930 à 1970. Nous exposons ses travaux à l'intention des non-spécialistes, en insistant sur ceux publiés dans les *Comptes rendus hebdomadaires des séances de l'Académie des sciences*.

1. Introduction

The *Comptes rendus hebdomadaires des séances de l'Académie des sciences* (or "CRAS" for short), published by the French "Académie des sciences," has a long and rich history covering a period of about three and a half centuries. It started in 1666 with the creation of the "Académie royale des sciences," ancestor of the present Academy, in a time when printing, under royal control, was not widespread in France. Since then, some of the works of scientists, among the most famous, have been published, discussed or/and extended in this journal. Here, we shall concentrate on the second part of the 20th century with, in particular, the works on magnetism of the 1970 Nobel laureate, Louis Néel. With a total of about 200 publications, mostly written in French, but with a strong international impact, Néel's contributions to the CRAS were frequent: almost 40 papers before 1950 – around the middle of his scientific career – and 30 afterwards. Here, we have included the seven papers that Néel published during the Occupation in an ephemeral journal, *Cahiers de physique*, presumably created under the auspices of the Academy (section 4).

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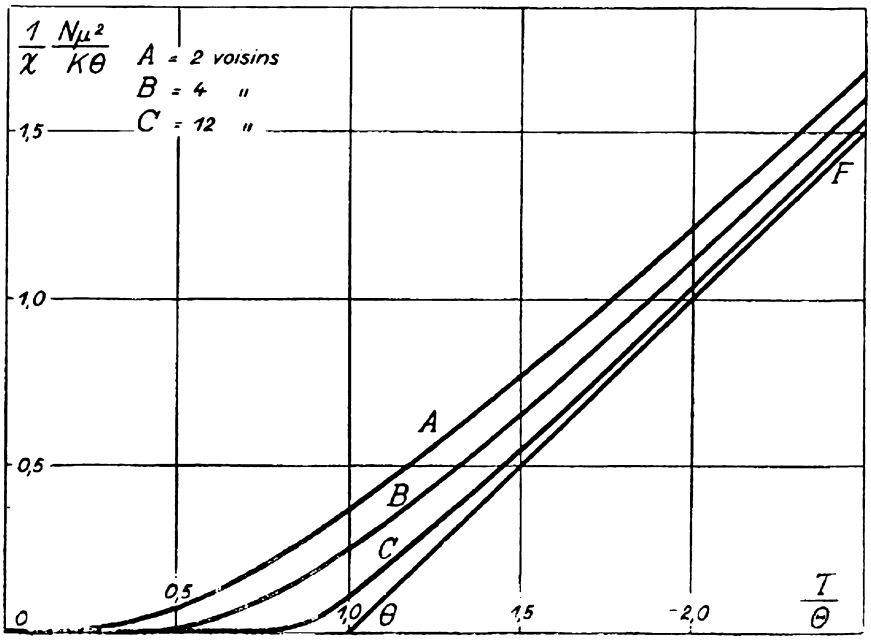

Fig. 1. Néel's plot of the reciprocal susceptibility of a ferromagnet, calculated with 2, 4, and 12 neighbours (curves A, B, and C). These curves show a positive curvature subsequent to the local character of ferromagnetic fluctuations. The last curve is linear and intercepts the temperature axis at the temperature θ (see text). It corresponds to the mean-field limit with an infinite number of neighbours. This curve is also the high-temperature asymptote of A, B, and C [6,9].

In November 1928, after graduating at the "École normale supérieure" in Paris, he started a thesis in Strasbourg under the supervision of Pierre Weiss (1865–1940) a contemporary of Pierre Curie and Paul Langevin (1859–1906 and 1872–1946). Everyone knows the "Curie law" or the "Langevin function" in paramagnetism, and the "Curie–Weiss" law in ferromagnetism. Weiss was also renowned for his works on ferromagnetism, especially for having introduced the basic notions of "ferromagnetic domains" [1] and "molecular fields" [2], both named after him.

2. The "local molecular field" and the discovery of antiferromagnetism

The subject of Néel's thesis simply consisted in the determination of the Curie constant of iron. After rapidly completing this work, he started working on three other subjects, trying to understand some questions that the scientific community felt they should be taken seriously. These subjects were: (i) the explanation of the so-called "two Curie temperatures," defined by deviations from linearity of the Curie–Weiss law: the Curie temperature on the ferromagnetic side and the Curie–Weiss temperature on the paramagnetic side, (ii) the non-linear reciprocal paramagnetic susceptibility measured in ferromagnetic alloys with uniform disorder, (iii) the interpretation of a phenomenon, called at that time "constant paramagnetism," observed at low temperature in several materials. In less than two years, Néel solved those problems thanks to careful experiments completing the existing ones, and, more importantly, to their interpretations, which were based on a completely new idea, viz., the replacement of the Weiss molecular field (which was what we now call a "mean field") by a "local" molecular field distributed among the different neighbours, paving the way for fluctuations, of thermal or/and spatial origin. Interestingly, Néel had this idea after having heard of the short-range character of the quantum exchange interactions discovered by Heisenberg [3].

Regarding his first own subject, Néel experimented [4–8], and then interpreted [6,9–11], the "two Curie temperatures," first by showing that the apparent Curie constant that appears in the Curie–Weiss law can be modified by thermal expansion [12] – thus being different from the Curie constant of an isolated moment – and then by evaluating the paramagnetic susceptibility χ of a ferromagnet for different numbers of neighbours in the case of the "Lenz hypothesis" – an early Ising model without justification and formulated for the sole purpose of simplifying calculations. Fig. 1, extracted from Néel's thesis [6], represents the thermal dependence of the reciprocal susceptibility in the presence of fluctuations as $1/\chi(p,T) = (kT/N\mu^2)\exp[-(p+1)\theta/2T]$ for $T < \theta$ and $1/\chi(p,T) = (kT/N\mu^2)[T/\theta - 1 + \theta/2pT + \ldots]$ for $T > \theta$, where θ is the Curie–Weiss temperature obtained either from high temperature extrapolation of $1/\chi(p,T)$ or from the $p \to \infty$ curve. Here, N is the number of spins μ, each one being coupled with its $2p$ neighbours, up or down, due to thermal fluctuations. When p is finite, $1/\chi(T)$ is non-linear, with a positive curvature, in a fashion similar to the current non-mean-field case (Fig. 2, bottom and middle).

We must note that, here, the vanishing of the reciprocal susceptibility and therefore the emergence of ferromagnetism does not take place at the finite temperature T_c at it should be, but at the absolute zero of temperature, except when $p \to \infty$

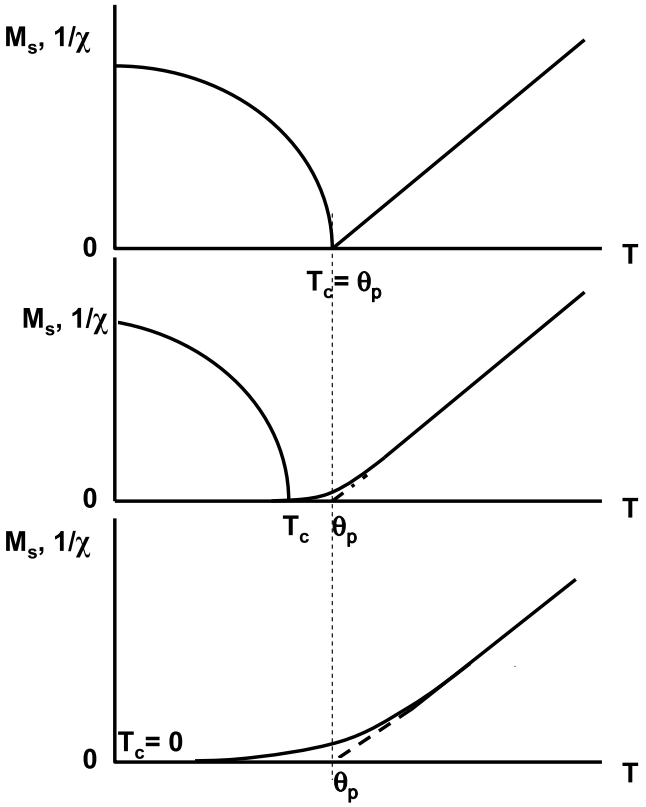

Fig. 2. Temperature variations of the reciprocal paramagnetic susceptibility $1/\chi$ and of the spontaneous magnetization M_s of a ferromagnet schematized for three different cases. In all cases, ferromagnetism takes place at the Curie temperature T_c when the susceptibility becomes infinite ($1/\chi = 0$), whereas high-temperature extrapolations of the linear variations of the $1/\chi$ curves give the paramagnetic Curie temperature θ_p. Top: mean-field case – without fluctuations – where $T_c = \theta_p$. Middle: non mean-field case where T_c is reduced by fluctuations leading to $T_c < \theta_p$. Bottom: Néel case ([6,9] and Fig. 1), where the Curie temperature is reduced to $T_c = 0$ due to overestimated fluctuations (see text).

(mean-field case, see also Fig. 2, bottom, when $T \to 0$). Néel pointed out this problem, which did not seem to bother him as he was mainly interested in paramagnetic fluctuations. The origin of this lack of ferromagnetism in this approximation is easy to understand: the "central spins" coupled with their $2p$ first neighbours constitute an ensemble of $N/(2p+1)$ independent blocks of $2p+1$ coupled spins subjected to strong thermal fluctuations, which is similar to a superparamagnet with a blocking temperature sitting below its Curie temperature. Note that superparamagnetism (section 5.6) was identified only about twenty years later, five years after P.R. Weiss avoided that pitfall by applying the well-known Bethe–Peierls method to ferromagnetic exchange interactions: one atom is chosen as the central atom and its interaction with the first shell of nearest neighbour atoms is calculated exactly, the rest of the system being self-consistently replaced by an internal molecular field acting only upon this first shell [14–16].

In Néel's approach beyond the mean-field theory, when the temperature decreases, the ferromagnetic correlations become stronger and stronger, although not more and more spatially extended. It is conceptually different from the Fisher–Widom–Kadanoff scale-invariance theory [17], where the size of self-similar ferromagnetic blocks diverges at T_c. Nevertheless, a step could be made in direction of this theory if (i) one assimilates the size of "Néel's spin-blocks" – proportional to $(2p+1)$ – to the FWK correlation length $\xi(T)$ at a given temperature and (ii) one imagines that, when the temperature decreases (Fig. 1), $\xi(T) \propto (2p+1)$ progressively jumps from one curve associated with a given p-value to the next one with a larger p-value until $p \to \infty$ ($A \to B \to \cdots \to \theta$). In such a case, "Néel spin blocks" become larger and larger, until they diverge at a finite temperature θ, as in the FWK model. For that reason, we might believe that Néel's approach, at a time when at the mean field was considered uncontroversial, could be taken as a first step towards the FWK model.

Incidentally, on the occasion of this study, in which both notions of interactions and distances were important, Néel started working on the variations of the interactions between 3d elements with their distance [13] referring to the Slater plot, which became later the Slater–Néel plot (section 3).

Regarding his second subject of research, after a first unsuccessful attempt where he simply added a constant susceptibility term to the paramagnetic one [18], Néel succeeded in solving it thanks to a generalization of the Weiss molecular field $H_m = n(M_A + M_B)$ created by the magnetic moments M_A and M_B belonging to two different magnetic sites. By extending the Weiss molecular field coefficient n according to the nature of the magnetic bonds, he obtained the new expressions

$H_{mA} = n_{AA}M_A + 2n_{AB}M_B$ and $H_{mB} = n_{BB}M_B + 2n_{AB}M_A$ [13]. This allowed him to understand the measured hyperbolic reciprocal paramagnetic susceptibility of alloys [6] and to explain several works including his own on Ni–Co, Fe–Co, Fe–Ni, Mn, and Cr alloys diluted in Au, Ag, or Cu [13,19,20]. As he pointed out, at that time the linear variation of the Curie–Weiss law was so institutionalized that his work went almost unnoticed and the curvatures observed experimentally in the reciprocal susceptibility of alloys were rather interpreted in terms of a succession of Curie–Weiss segments interlocking with one another.

With his third subject, Néel completed the most important part of his thesis. First, he experimentally confirmed the existence of a puzzling low-temperature "constant paramagnetism" in his diluted Mn and Cr systems [20], already observed in several systems. Then, with a desire to better understand this phenomenon, he looked at different systems such as S [21] and Ni, for which he made a model of Stoner paramagnetism with a simple rectangular band [22,129]. He also showed experimentally that the number of electrons participating in the magnetism of Ni remains unchanged when crossing the Curie temperature [18]. At that time and maybe in part with the same aim, Landau developed his theory of quantum diamagnetism, which involved a temperature-independent susceptibility [23].

All those unfocused experimental and theoretical results confirmed Néel in his idea of a "constant paramagnetism" originating from some unknown couplings between magnetic ions. To this end, he constructed a model based on another idea, still based on his first hypothesis of a local molecular field: the molecular field originating from some magnetic moments can be negative, *i.e.* with a direction antiparallel to the one of the other magnetic moments. This iconoclastic viewpoint led to the expression (1), also calculated on the basis of the Lenz hypothesis with a field H parallel to the magnetic moments direction [6,20],

$$\bar{\mu} = \frac{N\mu^2 H}{kT} e^{-\theta/T} \tag{1}$$

where $\bar{\mu}$ is the mean magnetization, N the total number of spins μ, $\theta = pw/k > 0$ the paramagnetic temperature – equal to the product of the anti-parallel coupling energy w between two magnetic moments with p the number of neighbours – and k is the Boltzmann constant (note that the sign of w depends on the one chosen for the coupling energy, which differs according to Néel's publications). This expression was based on an exact calculation for anti-parallel pairs of spin, with no coupling between them (*i.e.* limited to $p = 1$ with the notations of the previous section). Such a situation, corresponding to a "fictitious substance" as Néel wrote in his thesis, nevertheless reproduces the well-known minimum at temperature θ of the reciprocal susceptibility in a field parallel to the antiferromagnetism direction, and the linear variation of the high-temperature reciprocal susceptibility extrapolated down to the negative temperature $-\theta$, as shown in Fig. 3. The corresponding expression obtained from a high-temperature expansion of (1) is given just after expression (3). Nevertheless, Néel did not really understand his result immediately. As he wrote in his thesis dissertation, referring to Fig. 3, "there is no example of such a variation". Clearly, he was talking of the low-temperature side. In fact, most experiments were performed on polycrystalline samples and the term calculated with a field parallel to the spins was probably hidden by the contribution of the spins perpendicular to the field, leading to the nearly temperature-independent susceptibility curves observed. Then, admitting that the problem was due to restrictions of his Lenz hypothesis, Néel completed this calculation by another one, where the magnetic moments occupy all the space directions; this led to the desired outcome (Fig. 4) of a temperature-independent susceptibility at low temperature [6,20]:

$$\bar{\mu} = \frac{1}{3}\frac{\mu^2 H}{w}\left[1 - \frac{2w}{kT}\frac{1}{e^{2w/kT} - 1}\right] \tag{2}$$

Note that, in contrast to (1), where the number magnetic moment, N, intervenes, this expression is normalized.

A high-temperature expansion of (2) leads to what is known as the Curie–Weiss law for antiferromagnets (Fig. 3):

$$\bar{\mu} = \frac{1}{3}\frac{\mu^2 H}{kT + w} \tag{3}$$

(in Néel's thesis, this expression contains a printing error by a factor of 3). Not surprisingly, (3) is very similar to the high-temperature expansion of (1), $\bar{\mu} = \frac{N\mu^2 H}{k(T+\theta)}$ for a field parallel to the magnetic moments. Equating the paramagnetic temperature of these two expressions gives $\theta = w/k$ and the same intercept with the T-axis at $-\theta = -w/k$. In expressions (1) and (2) and Figs. 3 and 4, a temperature linearity breaking takes place at the positive temperature θ with the onset of a minimum for (1) and of flatness with (2). Néel called this temperature the "degeneration temperature," even if, sometimes, he used the term "Curie temperature". Néel showed qualitatively that the general shape of these curves remains unchanged if more distant neighbours distributed on successive concentric spheres are taken into account in what we would call a mean-field approach. Later, he calculated the temperature-independent transverse susceptibility taking into account all the nearest-neighbour interactions [18].

At this point, it is clear that Néel's negative molecular field creates an anti-parallel arrangement of neighbouring magnetic moments, which was a local effect in his idea. While his expressions, given above, were equivalent to those that we currently use, the same was not really true for his physical interpretation. After he finished his thesis, in 1932, the above expressions

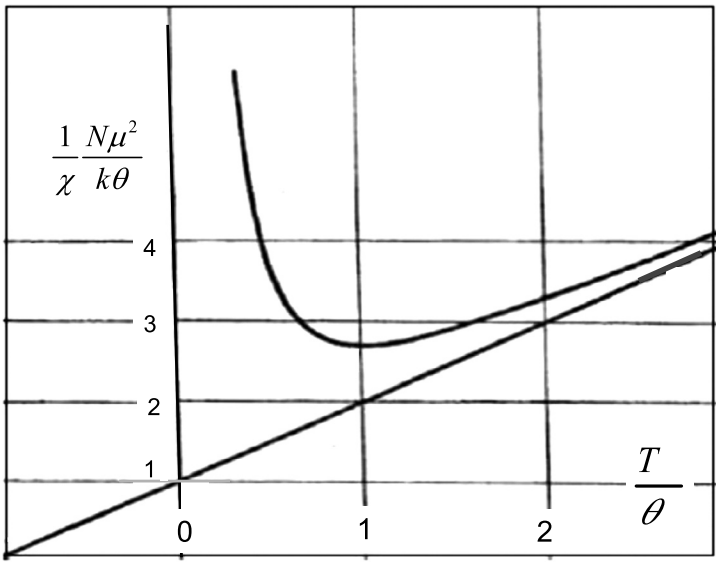

Fig. 3. Reciprocal susceptibility calculated with a "negative molecular field" for an applied field parallel to the antiferromagnetic direction [6,20].

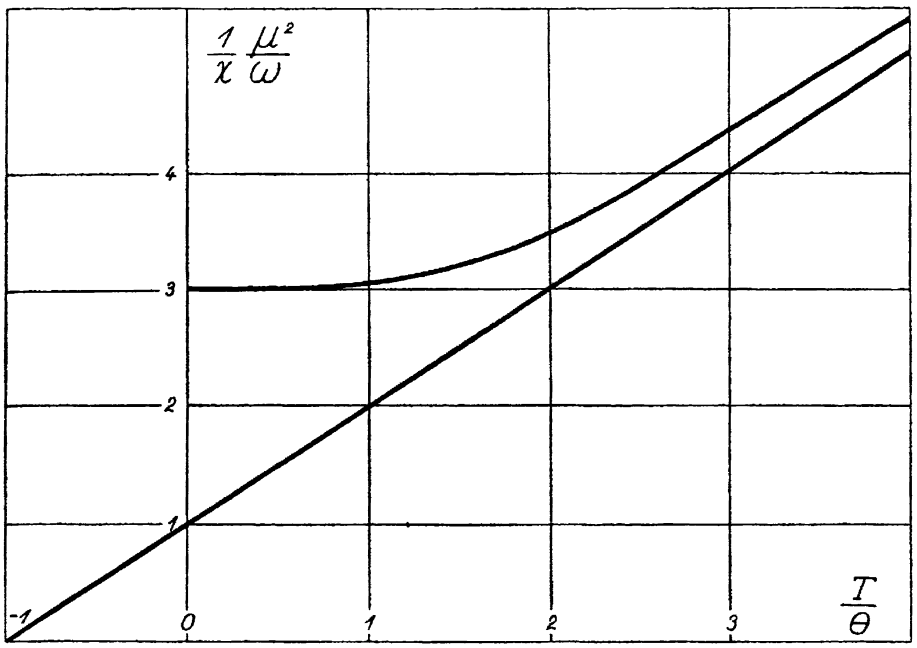

Fig. 4. Reciprocal susceptibility calculated for a polycrystal with the use of a "negative molecular field" [6,24].

represented for him a kind of paramagnetism with antiferromagnetic short-range order, consistent with his main idea of local fluctuations of the molecular field and also with his initial hypothesis limited to anti-parallelism of spin-pairs ($p = 1$).

Néel defended his thesis on 11 March 1932 and published it the same year [6]. With both detailed experiments and original theories, it included the resolution of the problem originally posed by Weiss, plus the applications of his ideas of a local molecular field to three different situations: (i) the explanation of the so-called "two Curie temperatures" [5,9,10,7,8], (ii) the explanation of the hyperbolic paramagnetism of ferromagnetic alloys [4,13,19], and (iii) the interpretation of the low-temperature "constant paramagnetism" in terms of what was six years later, in 1938, called "antiferromagnetism" by Bitter. His thesis [6] summarized several publications, which had appeared in the CRAS just before [5,21,7] or just after [9,12,13,11]. Note that in [11], he even took a step towards quantum mechanics by using quantized spins.

Four years later, in 1936, Néel clarified his physical interpretation of "constant paramagnetism". In realistic papers [24, 18] he clearly mentioned the long-range character of an antiferromagnetic order (Fig. 5) disappearing with a specific heat anomaly at the temperature θ – his former "degeneration temperature" which became, in 1938, after a suggestion of Gorter,

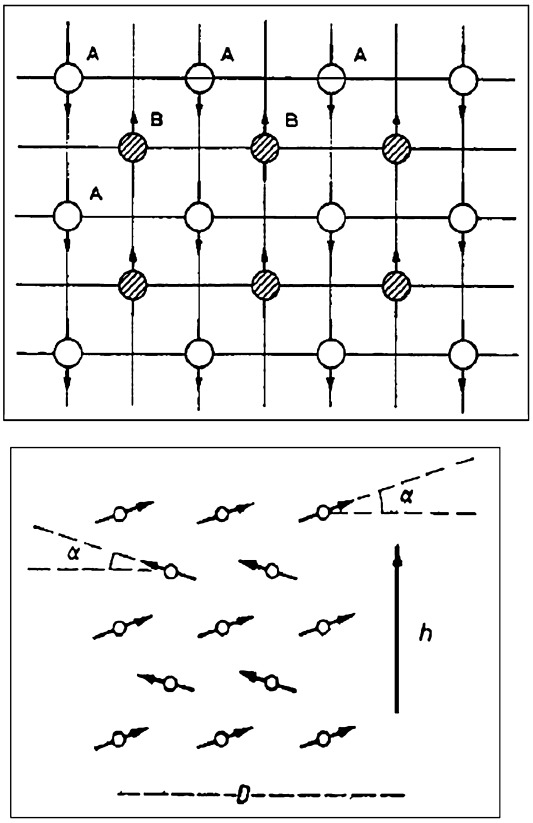

Fig. 5. Top: spin assembly of minimum potential energy for a negative molecular field at low temperature and its deformation under the application of a perpendicular magnetic field (bottom) [24,18].

the "Néel temperature θ_N". Interestingly, in the introduction of this paper Néel refers to his thesis by writing: "on several occasions, I have shown that a substance with negative atomic moment and molecular field shows at low temperature a temperature-independent susceptibility. But since those demonstrations have been made in cases that are too specific including the excessive role played by fluctuations, I think it is useful to repeat the question in a more general and rigorous way." [25]

Not surprisingly, he also replaced the term "constant paramagnetism" used since 1930 by "constant susceptibility". Incidentally, he also noted that this clarification gave him the opportunity to respond to his first detractors Gorter and Landau, who claimed, in 1932, that a correct theory of quantum dynamics could not give a temperature-independent susceptibility. This was not wrong but, clearly, understanding the application of quantum mechanics at the macroscopic scale was not yet imminent (see, e.g., [26,135]). In fact, one year after Néel's thesis, in 1933, and in spite of his severe objections – which were never published – Landau published a paper [27] in which he derived from his well-known expansion of the free energy, to 4th order in magnetization, the same type of expressions as those published by Néel the year before [6,20]. However, in addition, he calculated the specific heat anomaly associated with the onset of a long-range antiparallel order at a temperature that he called the "Curie temperature," corresponding precisely to Néel's "degeneration temperature". Landau's ideas were correct with regard to the onset of a long-range magnetic order, but were at the same time misleading, lumping together ferromagnetism and antiferromagnetism in the same bag. To conclude with this Néel–Landau controversy, one may say that the first and most important step was taken by Néel with his iconoclastic idea of a negative molecular field and his derivation of the right expressions for the antiferromagnetic susceptibility above and below his "degeneration temperature". This was so even if, contrary to Landau, he realized only in 1936 that his 1932 step forward opened the way for a completely new class of magnetic systems – beyond ferromagnets – much more important than he realized.

Two years later, in 1938 *i.e.* six years after Néel's thesis, Bizette, Squire, and Tsai from Bellevue found that the magnetic properties of MnO fitted perfectly with Néel's predictions for this system. This first validation of Néel's antiferromagnetism was also published in the CRAS [28]. The above-mentioned "antiferromagnetism" and "Néel temperature" denominations of Bitter and Gorter were subsequent to this publication. More than 10 years later, in 1949, just after neutron diffraction became possible in Oak Ridge, Shull and Smart showed that the arrangement of the atomic moments of MnO was precisely the one predicted by Néel... to the dismay of Anderson who, although being a physicist of genius, did not yet believe in Néel's antiferromagnetism.

The behaviour of antiferromagnets at large fields was unclear until Néel's prediction in 1936 [18] of the rotation of the "antiferromagnetism direction" – *i.e.* of the magnetic moments direction. When the field, applied along this direction, becomes comparable to the negative molecular field, the two anti-parallel moments rotate simultaneously by $\sim 90°$, making between them an angle of slightly less than 180°, *i.e.* with a small component along the field direction, as in Fig. 5. This transition was observed on $CuCl_2$ by Gorter et al. almost 20 years later by magnetic resonance [29]. Then Néel interpreted another type of field-induced transition observed in the in-plane "antiferromagnetic" system $MnAu_2$, which he assimilated to the so-called "metamagnetic transition" [30]. This transition differs from the above-mentioned "rotation of the antiferromagnetism direction" because it deals with systems having a large anisotropy, planar in the case of $MnAu_2$, which prevents the simultaneous rotation of the two moments – it is just the antiparallel one that reverses. In fact, $MnAu_2$ was not really an antiferromagnet, but a "helimagnet". Three years after Néel's paper, it was shown [31] that $MnAu_2$ was the first example of a new class of systems predicted by Yoshimori [133], in which a competition between the first and second neighbours interactions leads to a spiral magnetic structure.

3. Slater–Néel plot

When, in 1936, he clarified his physical interpretation of antiferromagnetism, Néel also published a work [18,32], that he had started two/three years before with Fe–Ni, $Fe_\gamma Co$, Ni–Co [13]: the plot of the coupling energy between 3d-elements in compounds and alloys *vs* their distance. In order to take into account the size of 3d atoms, Slater [33] and Stoner [34] previously used, for the "distance," the ratio of the 3d atoms separation to their diameters. Néel's improvements simply came from a more physical choice taking, instead, the non-normalized shortest distance separating the nearest points from the two 3d atoms – where electrons move. With a much greater set of available experimental data than his predecessors, he obtained a more comprehensive plot, now called the Slater–Néel plot. This plot enables to answer a large number of questions related to some general aspects of magnetism, such as the quantitative description of the volume anomaly or the magnetostriction in 3d ferromagnetic metals, their alloys and solid solutions.

4. End of the Pierre Weiss laboratory, the war period and Néel's multiple activities

In May 1939, about three months before the war broke out, the 4th International Meeting of Magnetism was organized in Strasbourg with, among the best world specialists, Becker and Döring, Mott, Simon, Stoner and Sucksmith, C.J. Gorter, Casimir and Kramers, Barnett and van Vleck. As written by Néel in his book *Un siècle de physique*, "In the daunting atmosphere of an imminent war, it marked the end of the laboratory activity that Pierre Weiss had founded and directed at the University of Strasbourg. The situation was very worrying. In his opening address, did not Terracher, the rector, say that this university 'maintained the pre-excellence of the cult of the spirit between the Maginot line and the Siegfried line'?" [35].

On 1 September 1939, the German troops invaded Poland and, on the same day, the French government decided the general mobilization, although the war was only officially declared two days later, on 3 September. During the following months, Néel's life was completely disrupted, with titanic works and adventurous periods. The official decision was to evacuate the University of Strasbourg as soon as on 2 September. The place of retreat was Clermont-Ferrand. Néel was already in Clermont on 1 September. Eight days later, he was in Paris to organize the scientific mobilization. The following month, as he was convinced that the unique and heavy scientific equipment of the University of Strasbourg was essential for the war effort, he and Charles Sadron organized a bold transfer by night of three train carriages of equipment to Meudon, permitting in particular the creation, with Lallemand and Soleillet, of a photoelectric laboratory for the production and detection of infrared radiation. But, already at the end of November 1939, Néel was thinking of putting his scientific knowledge and ability to the service of the French navy. He developed a dredging process for magnetic mines, which later allowed him to see with his own eyes the "nice work by his friend Becker," who designed those mines! This enabled him to understand the inner mechanism of these magnetic mines and to find a defence: his famous process of "neutralization" [36] based on the demagnetization of ships hulls by the compensation of the two terms of the Rayleigh law $M = aH + bH^2$ [37] – the reversible term that was at the origin of mine attraction and the irreversible term compensating the former after a treatment consisting in magnetizing the hulls of the ships. This work, performed in the "Centre d'études de la marine de Toulon," led to the treatment of 640 vessels in Brest, Cherbourg, Dunkirk, Le Havre, and Toulon. Several hundred human lives were saved, which could never have been achieved by other means. His collaboration with the Navy, which ended on 6 August 1940, allowed him to get on with his own Grenoble laboratory, an extension of the Toulon laboratory: the "Laboratoire du magnétisme du navire," where different magnetic problems associated with newly constructed ships in France and neighbouring countries were studied from 1949 until 2006 (*i.e.* until 30 years after his retirement).

In May 1940, the German army crushed the French forces, and in June of the same year an armistice was signed which split the French territory into two zones, one of which was occupied by the German army while the other was not and was sometimes called "the free zone" for short. Moreover, Strasbourg and the surrounding regions were annexed to Germany.

Néel decided by the end of 1940 to settle in the "free zone" [38]. Following Esclangon's wise advice, he opted for Grenoble. Together with Forrer and Weil, his assistants from Strasbourg, he was supposed to create a laboratory with what was at hand. The following five years, from the end of 1940 to 1945, were for Néel years of maturation during which, in particular, he realized how applications are likely to energize basic research. Indeed – as we shall see below – many of his later discoveries, often published in the CRAS, germinated during and just after this war period.

As the *Comptes rendus hebdomadaires des séances de l'Académie des sciences* remained in Paris [38], an alternative journal called *Cahiers de physique* was founded in the non-occupied zone by Georges Guadet, editor of the *Revue d'optique*. In his book [35], Néel writes: "This paper appeared in 1944 in the *Cahiers de physique*, an ephemeral periodical founded in the free zone by G. Guadet, editor of the *Revue d'optique*. When peace returned, it did not survive long..." [39]. In the records of the weekly sessions of the French Academy of Sciences of that period, one finds the following announcement:

> M. Charles Fabry, actuellement en zone non occupée, fait hommage à l'Académie, par l'organe de M. Gaston Fayet, du premier cahier de la nouvelle publication, *Cahiers de Physique*, qui vient d'être fondée sous sa direction pour permettre aux physiciens et astrophysiciens de faire plus facilement paraître leurs travaux.

This suggests that this new scientific journal was favourably considered by the "Académie des sciences" through Charles Fabry (1867–1945), academician, famous for his interferometer, close colleague of G. Guadet. We therefore shall consider the *Cahiers de physique* as an emanation of the CRAS.

Between 1942 and 1943, Néel gave a first interpretation of the old experimental Rayleigh law [40–42], with some extensions later [43]. To this end he simplified the effects of the intermingled domain wall distributions of a polycrystalline magnet at low fields, by constructing a model based on an ensemble of interacting single-domain ferromagnetic particles with a broad distribution of rectangular hysteresis loops displaced by random-field interactions. About 10 years later, he extended this approach [44] in order to interpret and generalize the famous Preisach diagram [45] for sintered ferromagnetic powders, permanent magnets, and so on; this is nowadays known as the Néel–Preisach diagram [46] (see section 5.4). Clearly, these works prepared him for his future development of nanomagnetism (see section 5.6).

At the present stage of this article, one can easily overlook the veracity of Nozières's remark that [47] "The work of a Nobel Prize that marked 20th century science cannot be presented." And so, after this first period that won him the first part of his Nobel Prize – the second one being for the discovery and the study of "ferrimagnetism" (see section 5.5) – we shall limit this presentation to his most important works (see section 5.5).

5. A new start in Grenoble with new directions in magnetism

When, in 1941, he started a new career in Grenoble, Néel immediately took magnetism to new and often applied directions. This turn seems to be due, at least in part, to his experience in ship magnetism when, as mentioned above, in 1940 he saved hundreds of sailor lives by a clever use [36] of the Rayleigh law [37], followed, during the occupation time, by his first interpretation of this fifty-year-old law [40–42] (see section 4).

5.1. A new type of permanent magnet

If Néel already understood by the end of the 1930s that a single-domain "particle" – or "nanoparticle" as we say nowadays – constitutes a small magnet, a concept that he used in his interpretation of the Rayleigh law [40–42], the corresponding publications were delayed until the end of the war [48,49]. Based on this concept, Néel's first work on real nanoparticles was the elaboration with Weil in 1941, of a new type of permanent magnet based on an original concept consisting in powdering and sintering a ferromagnet in order to obtain a solid sample made of ultra-fine and often elongated particles with sizes as close as possible as their "single-domain size," *i.e.* smaller than their domain wall thickness. Néel and Weil patented this work twice [50] – in 1942 and 1951 – showing how to make reasonably good permanent magnets with – even soft – materials such as Fe, the single ferromagnetic material available at that time. This process was exploited between 1945 and 1952 at the steelworks of Ugine near Grenoble run by René Perrin, providing very useful magnets for daily activities such as the use of bicycles.

The academic counterpart of these works showing the angular dependence of the coercive field of a single nanoparticle of ellipsoidal shape was published in 1948 by Stoner and Wohlfarth [51]. Much later, when I communicated to Néel a paper giving the first experimental evidence of this famous "Stoner–Wohlfarth model" on a 25-nm single Co nanoparticle, he showed a great interest in it and then added that Aubry, Weil, and himself had already used this "so-called Stoner–Wohlfarth model since 1940". This could have been another celebrated CRAS paper, but Néel had already extended his works to other more complex and interesting directions: the coercivity mechanisms of the most efficient – multiphase – permanent magnets of the time, the AlNiCo magnets.

5.2. Sample imperfections, the "dispersion field" and the coercive field theory

In order to understand multi-phase permanent magnets such as AlNiCo, Néel rapidly developed his next fruitful idea: the notion of a "dispersion field". Contrary to his "local molecular field" on which he based his pre-war discoveries, the "dispersion field" does not derive from exchange interactions, but from dipolar interactions and more particularly from those coming from magnetic free poles sitting at samples imperfections – surface roughness, crystallites, internal strains, holes or magnetic inclusions [52]. The notion of a dispersion field may be considered as an extension to real samples of the old and academic "demagnetizing field" created by magnetic free poles distributed on second-order surfaces – planes, spheres, ellipsoids, etc. – of a no-defect ferromagnetic sample. Néel showed how such defects and their associated dispersion fields

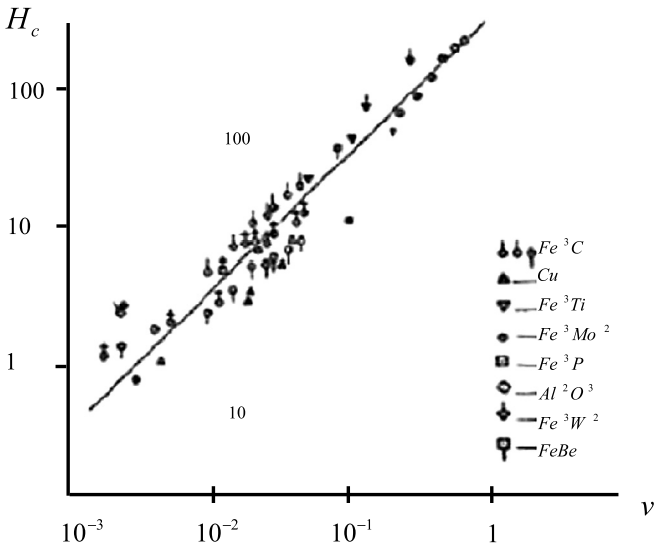

Fig. 6. Comparison of the measured and calculated coercive fields H_c *vs* the non-magnetic volume v in different Fe-based multiphase magnets, in logarithmic scales [53]. The data were clumped together by Kersten (1943).

create cooperative multi-valley energy barriers and contribute to the coercive field through different mechanisms of domain wall nucleation or/and pinning. This led in particular to his general coercive field theory of multi-phase systems [52–55,44]. Fig. 6 shows the variation of the coercive field measured on several Fe-based magnets with non-magnetic inclusions and fitted to the corresponding Néel expression [53]. Much later, this subject was adapted to the next generations of permanent magnets such as the ones based on rare earths ($SmCo_5$, $SmCo_{5-x}Cu_x$, NdFeB, etc.). The basic properties of rare earths were studied in Néel's laboratory from the early 1950s, whereas the applied aspects of their alloys have been studied for a few decades only. In these systems, with much narrower domain walls – due to much larger anisotropies – larger coercivities are associated with smaller defects, down to atomic or grain boundary sizes.

Before ending this section, one should note that Néel also considered the contributions of inter-grain interactions to the $1/H$ and $1/H^2$ laws of approach to saturation and their possible modifications by non-magnetic inclusions or/and cavities [56,55,130]. He also studied other aspects of this problem with, *e.g.*, the effect, on those laws, of modifications of the magnetocrystalline anisotropy by the creation of structural anisotropies [57,58,131] (section 5.4).

5.3. Magnetic domains and domain walls

Weiss magnetic domains [1] and Bloch domain walls [59] have often been mentioned in previous sections, and this is because they play a key role in macroscopic magnetism. Here, we shall discuss Néel's theoretical evidence for their topological structures and properties, such as the ways they can be arranged in crystals with different symmetries, their angular configuration, thickness and energy (generally limited to uniaxial and cubic). Concerning Bloch walls, Néel first showed that when the magnetization rotates from a domain up to a domain down, the magnetization must stay in the plane of the wall in order to avoid the strong energy cost due to in-wall dipolar interactions. He then calculated the characteristics of 180° and 90° Bloch walls taking also into account magnetostriction [60]. When, more than ten years later, he applied those ideas to thin films with in-plane magnetization – which is often the case, dipolar shape anisotropy being generally significant – he understood that below a critical film thickness, depending on its actual perpendicular anisotropy, the magnetic moments must rotate in the plane of the film causing inevitable intra-wall dipolar interactions. His calculations led, as for the Bloch walls, to the analytical determination of the angular configuration, thickness and energy of those domain walls. Not surprisingly, in addition to exchange and anisotropy, these quantities also depend on dipolar energy. This new type of domain wall, replacing Bloch walls in 2D magnetic films, was called a Néel wall [61].

With regard to the magnetic domains, Néel first tried to understand the numerous experiments performed since the thirties, and so he defined what he called the "modes" [62], each of them being specified by a very large number of Weiss domains with parallel magnetizations. For example, he demonstrated that a thick ferromagnetic film with perpendicular anisotropy subdivides into alternated up and down domains perpendicular to the film with, at their extremities, small in-plane domains enabling the magnetic flux going from one perpendicular domain to the next one, to close partially inside the film. The corresponding figure, with two main Néel modes (four if the closure domains are taken into account) appears in all magnetism books.

Interestingly, Landau had obtained similar results a few years before, of which Néel was clearly not aware, the publications in French and Russian being probably not translated in real time. Besides, Néel considered several other situations: for example, those connected with the presence of non-magnetic inclusions with the idea of understanding domain-wall

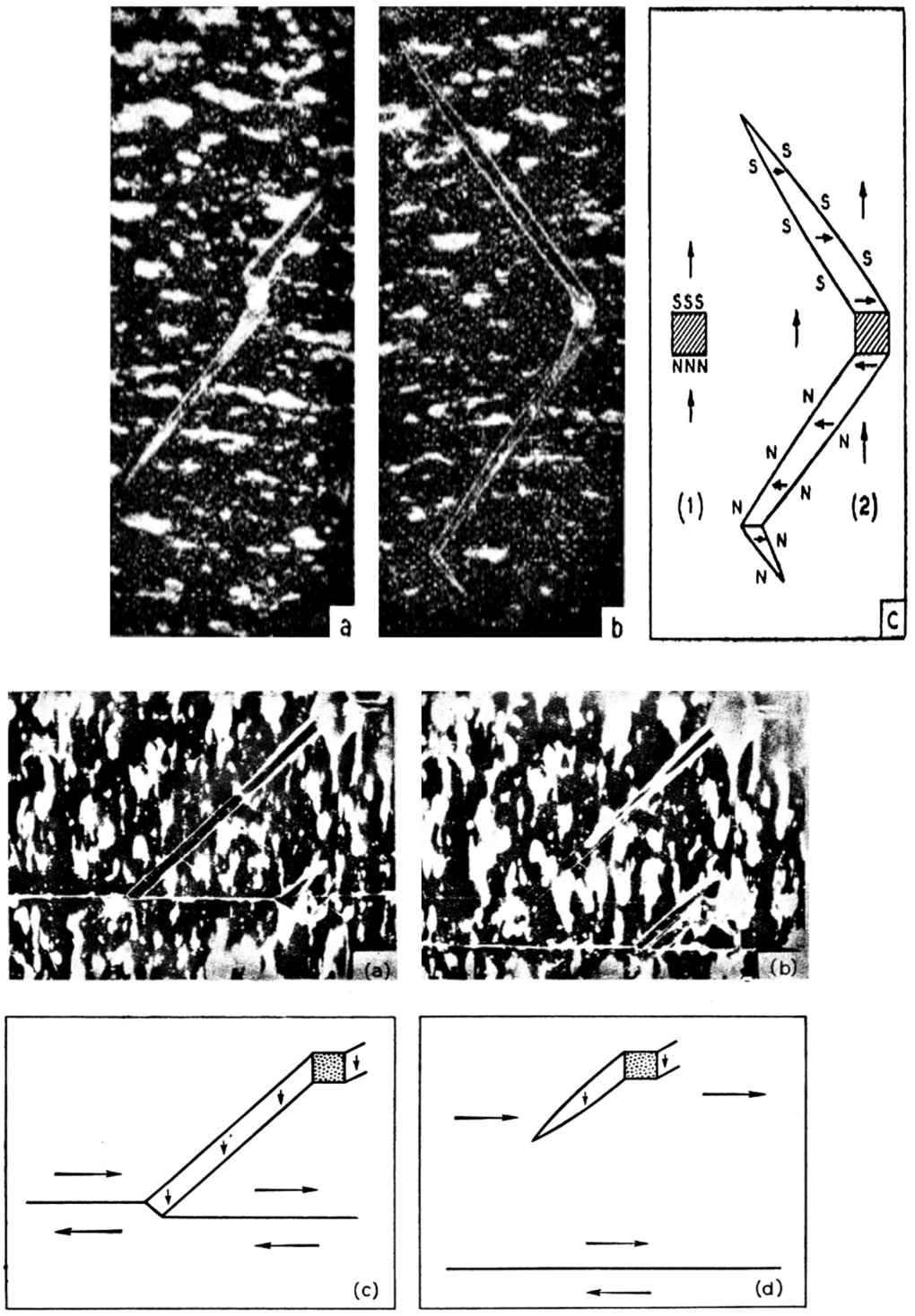

Fig. 7. Top: "Néel's spikes" observed on the surface of a ferromagnetic single crystal around holes and Néel's prediction. Bottom, pinning and depinning of a domain wall: observations and predictions [52].

pinning and coercivity. A well-known example, which also appears in most books on magnetism, is the one of the so-called "Néel's spikes" (Fig. 7), a giant artistic representation of which adorns the façade of the central building of the Louis Néel Institute in Grenoble.

Another example of magnetic domain configuration treated by Néel is the one of a cubic system with easy axes of magnetization along the three quaternary directions. In an increasing magnetic field applied along one of the ternary directions,

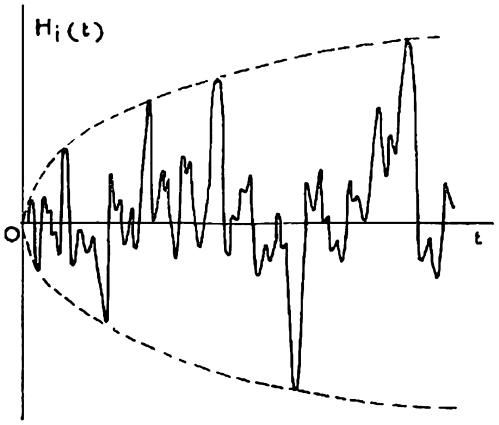

Fig. 8. Schematic time evolution of Néel's magnetic after-effect field $H_{\mathrm{m}}(t)$ [70].

the eight initial zero-field phases decrease to 6, 3, and 1 when all the magnetic moments are oriented along the applied field. As Néel pointed out, "I was able to interpret, in their smallest details, all the experimental results obtained over the previous twenty years by various observers." However, one must say that the last step, corresponding to the passage from three phases oriented along three different [63] directions, symmetrical with respect to the field parallel to the fourth one, was assumed to be continuous, but this is without accounting for the more recent 3D–3S Pott model predicting, in this case, a first-order phase transition in which these three phases abruptly jump along the field direction [64]; this was rapidly experimentally confirmed.

5.4. The magnetic after-effect theory

Important follow-ups of Néel's coercive field theory (section 5.2) were the first "magnetic after-effect" theories. Magnetic after-effect is a slow or relatively fast (in ac-experiments) time evolution of magnetization reversal after the modification of an initial condition (e.g., field vector or temperature). It has been studied theoretically for the first time by Néel, who essentially distinguished two different mechanisms: the "thermal after-effect" resulting from a fast-time variation of the field amplitude and the "diffusion after-effect" after a variation of the field direction.

The thermal magnetic after-effect had been studied experimentally in 1935 by Preisach, who rightly attributed it to thermal fluctuations [45]. Other experiments followed with, in particular, those of Néel and his co-workers Barbier and Lliboutry. They confirmed that the magnetization reversals in all types of existing magnets are time-dependent: at low fields (in the Rayleigh regime [43] or sometimes in soft materials) and at large fields (in permanent magnets or single-domain magnetic particles [65–68]). Néel showed that this effect results from thermally activation over nanometre-scale energy barriers, associated with domain-wall depinning or nucleation. Interestingly, these over-barrier processes are at the origin of the famous Barkhausen irreversible magnetization jumps discovered a long time earlier, thanks to a clever use of an amplifier/loudspeaker system [69]. Néel gave the first interpretation of those jumps, which could not be measured directly until the direct observations of large avalanche-prone quantum Barkhausen jumps between the end of the 1970s and the beginning of the 1990s. Based on his understanding of Barkhausen jumps, Néel described the thermal after-effect in terms of a local temperature and time-dependent mean field H_{m}, that he called the "magnetic after-effect field" [43,70]. In fact, this field was nothing else but his former – now time-dependent – dispersion field [52], a schematic time-representation of which is given Fig. 8. After having calculated the mean-square deviation $v\overline{H}_{\mathrm{m}}^2 = \frac{4\pi kT}{3}$ of this after-effect field, by equating the magnetic energy in the volume v with the thermal energy associated with the three thermal degrees of freedom, he assumed a Gauss distribution and obtained the following magnetization relaxation law [43,70]:

$$\frac{\theta}{t} \approx \sqrt{\frac{\overline{H}_{\mathrm{m}}^2}{2\pi h^2}} \exp\left(-h^2 \Big/ 2\overline{H}_{\mathrm{m}}^2\right) \tag{4}$$

where h is the after-effect field averaged within the measuring time interval t only. Here θ is the shortest time for which $H_{\mathrm{m}}(t)$ values can be considered as independent. Clearly, this expression which can be rewritten as $\theta/t \approx \sqrt{\frac{2kT}{3vh^2}} \exp\left(-3h^2 v \Big/ 8\pi kT\right)$ recalls the well-known Arrhenius one $\tau_0/t \approx \exp\left(-Kv(1-H/H_{\mathrm{A}})^{\alpha}/kT\right)$ for the reversal of the magnetization of a single nanoparticle of volume v and anisotropy constant K, the exponent α being ideally equal to 2. This law of relaxation for a single particle was also derived by Néel [68]. The comparison of these two laws suggests that h – given in cgs units – plays the role of an energy barrier reduced by the applied field. It is not surprising that the expression for h given in Néel's papers strongly differs from the single-particle energy barrier given above. This is because h does not

represent a single energy barrier, but a distribution of energy barriers averaged over a complex macroscopic system such as a permanent magnet.

Those Néel relaxation laws were applied to interpret the relaxation measurements performed on the different magnets of the time: Ni–Zn ferrite, iron, Ni, Fe–Co, steels, AlNiC, etc. [70] and, later on, on nanoparticles (section 5.6). Néel's description of magnetic irreversibility in terms of thermally activated energy barriers distributions has been a starting point for the study of disordered systems such as glasses, spin-glasses, random anisotropy systems, etc., and also for new aspects of the old Preisach–Néel model (see, *e.g.*, [71,72]). He also introduced what he called the "anhysteretic magnetization," which covers the non-conventional magnetic response of a ferromagnet to a slow oscillating field of initial amplitude larger than the coercive field and decreasing down to zero. For example, the initial reciprocal susceptibility – usually given by $1/\chi_i = 1/\chi_{i0} + N$, where $1/\chi_{i0}$ is the reciprocal susceptibility in the absence of dipolar interactions and N the demagnetizing field factor – is no longer valid, and Néel derived a new expression for it [73]. When this field is non-symmetric with respect to the field origin, the response – such as the logarithmic increase of the remanent magnetization – is different, and this phenomenon is called "reptation" [74,132].

Resulting from a fast change of the direction of the applied field, the second type of magnetic after-effect, called the "diffusion magnetic after-effect" (previously, the poorly named "reversible after-effect"), must be linked to symmetry, *i.e.* to magnetic anisotropy. In fact, in 1938 already, Snoek indicated that this phenomenon should result from field-induced atomic diffusion, especially in materials with vacancies. Néel supplemented this by showing how atomic diffusion may cause a progressive stabilization of a new magneto-crystalline symmetry, leading to a slow rotation of the spontaneous magnetization. While developing theoretical models [75–77], he also guided his Grenoble co-workers in the study and the creation of new types of "orientation superstructures" obtained through thermal or mechanical deformations taking into account magnetostriction [78], mechanical deformations [79], and electron [80] or neutron [81] irradiation.

5.5. Ferrimagnetism: a third class of magnetic systems. Superexchange interactions

It was also between the late 1940s and the 1960s that Néel made his second most important breakthrough after discovering antiferromagnetism: "ferrimagnetism". This story started when, in 1947, after reading a paper by Verwey and Heilmann on the structure of ferrites, he became interested in the so-called spinel ferrites of formula Fe_2O_3MO, where M is a 3d bivalent metal (*e.g.*, Fe^{2+}). The magnetic properties of those materials, with a spontaneous magnetization, were absolutely not understood, despite a very important set of experimental results accumulated over the years, such as those of Serres in Strasbourg [82]). The two main questions were (i) how the spontaneous magnetization of those "ferromagnets" could be so small (much weaker than the sum of their magnetic moments (10 to 15 μ_B per unit cell) and (ii) why the reciprocal susceptibility shows such an extraordinary hyperbolic shape with a negative curvature and a high-temperature asymptote extrapolating to a negative temperature.

Owing to his knowledge of antiferromagnetism, it was surely easy for Néel to find the solution: those ferrites are simply non-compensated antiferromagnets; he called them "ferrimagnets". For their interpretation, he developed the same local molecular field approach as for antiferromagnets with different, site-dependent, molecular field coefficients: one within the Fe site, one within the M site and one in between. Assuming the latter to be negative and much larger than the two others, the magnetic moments belonging to each site (Fe or M) had to polarize in an anti-parallel direction – later confirmed by neutron diffraction investigations, *e.g.*, [83] – giving a total magnetization equal to their difference $M_{Fe} - M_M$. As expected, this difference perfectly fitted in with the previous experimental results. Néel's discovery of ferrimagnetism in 1947 was published one year later in a long paper [84] giving the expression that completely characterizes this new type of magnetic system, and enabled Néel to make accurate predictions. The expression for the hyperbolic reciprocal susceptibility (5) and its graphical representation (Fig. 9) are given by:

$$\frac{1}{\chi} = \frac{T}{C} + \frac{1}{\chi_0} - \frac{\sigma}{T-\theta} \tag{5}$$

where $C = \mu^2$ is the Curie constant, whereas χ_0, σ and θ are functions of the relative numbers and of the sizes of the different types of magnetic moments, of their associated molecular field coefficients, and of their number of neighbours. The characteristic temperatures involved in Fig. 9 are $\theta_a = -C/\chi_0$ and θ_p is a more complex function of the different parameters, factorized by the Curie constant. These expressions are, of course, extensions to the Curie and Néel temperatures of ferromagnets and antiferromagnets. Néel systematically investigated all the possible situations (starting with different types of sublattice magnetizations with different thermal variations, different coupling strengths...) that led to the prediction of different types of ferrimagnetic behaviours with, in particular, the famous "compensation point" [84] (Fig. 10). This temperature, at which the magnetizations of the two sublattices exactly compensate themselves, as in an antiferromagnet, lead to a divergence of the coercive field – the hysteresis loop area being constant. It is a signature of ferrimagnetism. Néel then used his theory to interpret some of the existing results on spinel ferrites [85–87]. Note that none of them showed a compensation point, which is not surprising as the two sublattice magnetic elements are not different enough.

At nearly the same time, in 1953, a new series of compounds initially considered as "ferromagnets" was synthesized in Strasbourg [88]: the so-called rare-earth ferrites. Three years later, Bertaut and Forrat, in Grenoble, clarified their crystal

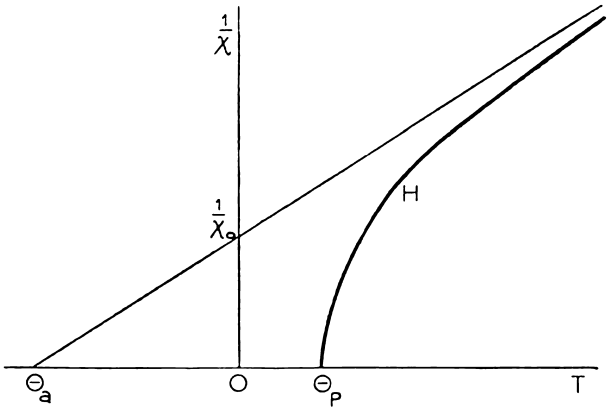

Fig. 9. Reciprocal paramagnetic susceptibility of ferrimagnetism showing the negative paramagnetic temperature θ_a and the magnetic ordering temperature θ_p [84].

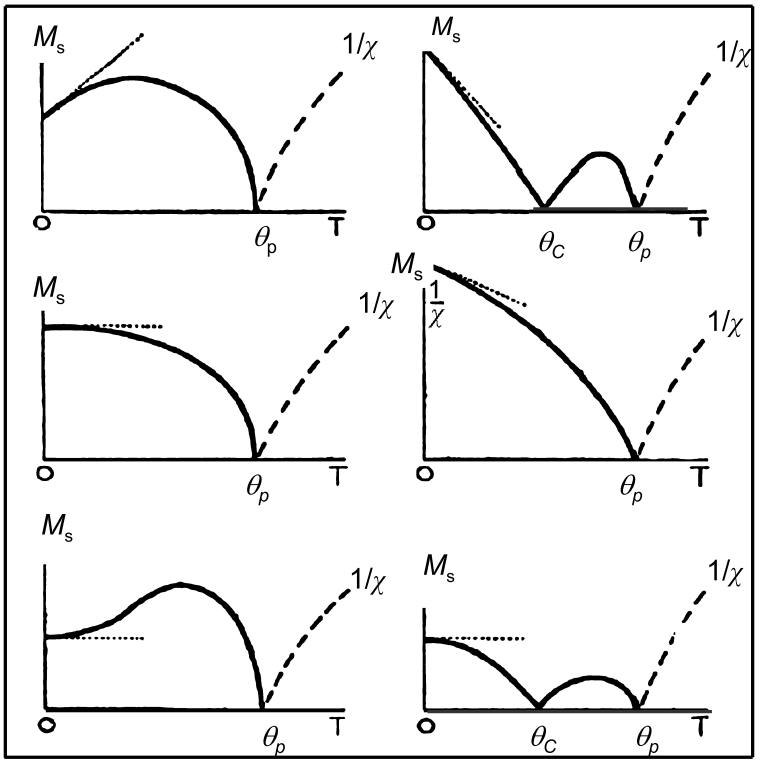

Fig. 10. Néel's prediction of main types of thermal variations of the spontaneous magnetisation and reciprocal susceptibility of a ferrimagnet [84,91].

structure and exact formula: $Fe_5M_3O_{12}$ (where M is a rare-earth 4f transition metal), showing that they belong to the well-known family of garnet precious stones [89]. Pauthenet started the study of their magnetic properties in 1954. Blum and he immediately discovered the first compensation point in the gadolinium garnet [90]. The same year, generalizing his theory of ferrimagnetism to three sublattices with six independent molecular fields, Néel gave a first qualitative interpretation of those results [91].

Ten years later, Pauthenet, Dreyfus, and Néel gave a detailed comparison between experiment and theory for all the heavy rare earths [92], obtaining the excellent agreement of Fig. 11. Several other papers attesting to the ferrimagnetic character of these systems were also published with Aléonard and Barbier, just after Pauthenet defended his thesis in 1957 under the supervision of Néel [93,94]. The fits of Fig. 11 confirmed that the compensation point decreases regularly from Gd to Yb, which is a consequence of the faster decrease in magnetization $M(T)$ when the total angular momentum J becomes smaller.

Another interesting aspect of Fig. 11 resides in the observation of very large ferrimagnetic ordering temperatures. Néel remarked that the large inter-anion interactions observed between different sublattices could not come from direct ex-

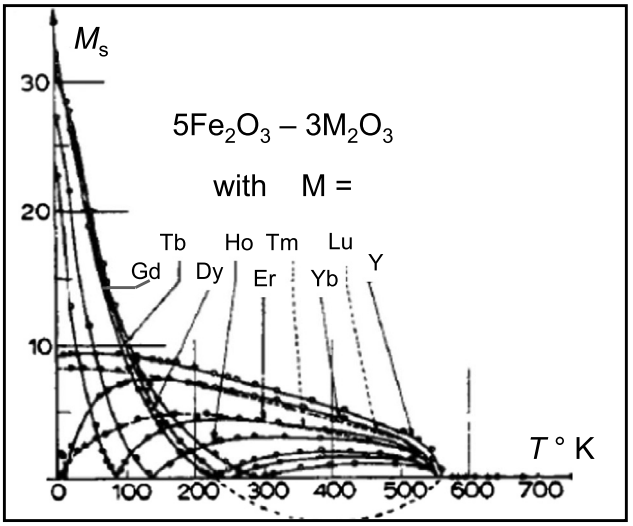

Fig. 11. Temperature dependence of the spontaneous magnetization of the second series of rare-earth garnet ferrites measured by Pauthenet and fitted with Néel: *Les propriétés magnétiques des ferrites d'yttrium et de terres rares de formule* $5Fe_2O_3 \cdot 3M_2O_3$ [91,92].

change, the ions being too far apart, but rather from the superexchange described by Kramers about 20 years before [95]. This interpretation of the ordering temperatures of rare-earth garnets in terms of the currently well-known superexchange was, at the time, rather revolutionary, as this was its first experimental confirmation. Those rare-earth ferrites were also a first example of ferromagnetic-like insulators of great interest for applications (*e.g.*, for devices using magnets under microwaves) and were patented, but not early enough [96]. The discovery of ferrimagnetism, and in particular of the garnets with their important potential applications, completed the discovery of antiferromagnetism, leading to Néel's attribution of the Nobel Prize.

Later on, Néel calculated the magnetic properties of some pure rare-earth elements [97], whose purifications below a percent became possible in the USA after an unsuccessful attempt in France. Those 4f transition metals and their alloys became later one of the most important fields of research of his laboratory, under the supervision of R. Lemaire.

5.6. The birth of nano-magnetism: nanoparticles and rock magnetism, multilayers and magnetic recording

Before the above brief overview of Néel's ferrimagnetism, we have occasionally used the terms "small particles," "nanoparticles," or "magnetic films". This is because the experimental and theoretical studies of those objects punctuated Néel's works ever since the beginning of the 1940s.

Regarding "magnetic nanoparticles," most of Néel's works were motivated by the interpretation of rock magnetism, in which he started to be interested as early as 1931, when he was hired by the university of Clermont-Ferrand with the title of professor, before resigning and going back to Strasbourg to complete his thesis. His interest in this subject came from an important discovery of Bruhnes, who had directed the observatory of Clermont-Ferrand around 1900: some of the large lava slabs of the temple of Mercury, at the top of the Puy-de-Dôme, had a remanent magnetization directed in the opposite direction to the others, in disagreement with both the direction of the Earth magnetic field and the magnetization of the underlying lava.

For Néel, this was obviously an exciting phenomenon that deserved to be deepened; but, for lack of means, he could not afford it back then. It was more than twenty years later, in the 1950s, that he could really tackle this problem. Indeed he had all the basics for it, as recalled in the next two sentences. At temperatures smaller than intra-particle Curie temperatures, the ensemble of atomic moments of each particle is equivalent to a single collective "giant moment" equal to their sum if these interactions are ferromagnetic (see, *e.g.*, [48]). If the interactions are antiferromagnetic, the collective moment results from the lack of full compensation and is approximately given by the square root of the number of moments participating in the surface roughness of the particles.

The rocks that Néel studied contained naturally-sintered ferromagnetic or ferrimagnetic nanoparticles sometimes coupled by dipolar interactions (as was always the case in his interpretation of the Rayleigh law [40–42]), [43], or in his new type of permanent magnets [50] where inter-particle exchange and dipolar interactions dominated thermal fluctuations). For the study of rock magnetism, Néel considered in detail the opposite situation of more or less distant particles, in which case the temperature is larger than interparticle interactions, when the magnetic moment of each particle fluctuates in temperature. He called "superparamagnetism" the case of ferromagnetic nanoparticles [98,68,99,100] and "superantiferromagnetism" the case of antiferromagnetic nanoparticles [101–105,63].

Not surprisingly, such nanoparticle fluctuations generally freeze below a "blocking temperature" given by the single-particle Arrhenius law of section 5.4, leading to a set of specific properties such as the time-dependent thermo-remnant

magnetization *i.e.* the magnetization that each particle acquires when it is cooled in a magnetic field. Together with Néel's description of magnetic irreversibility in terms of thermally activated energy barrier distributions (section 5.4), those concepts were obviously very useful in the study of terracotta [98,68] but also, later, in the study of spins glasses [71,106, 134].

Néel's first explanation [68,107,99,97] of the magnetic memory of "terracotta" and "lava lands" observed by Émile Thellier [108] in the 1940s, came out in the 1950s. He also helped geophysicists to understand the reality of the inversions of the earth magnetic field, explaining for example how Thellier's results on the magnetic memory of rocks [109,128] could be interpreted as resulting from the presence in rocks of single-domain magnetic particles precipitated during the cooling of lava in the presence of the Earth's magnetic field. Being at room temperature at the time of their discovery, *i.e.* below their blocking temperature, they give rise to thermo-remanent magnetizations oriented in the direction of the Earth field at the time of their cooling, with possibly very slow magnetic after-effect persisting at geological timescales.

However, understanding theoretically the inner mechanisms that govern these inversions was a serious challenge. Interestingly, Néel was in a way at the origin of a significant advance in this direction when, by the end of the seventies, he triggered the interest of P. Nozières on this subject. The latter constructed a mathematical model for the dynamical properties of the Earth's dynamo, based on both mechanical and electromagnetic degrees of freedom [110]. In particular, he found two very different timescales: a fast one for each magnetic field inversion and a slow one for their periodicity, the ratio between the two being in full agreement with observations! Néel's rather simple, but basic and very useful notions published in the above-mentioned series of papers in the *Comptes rendus*, were followed by a review paper – probably invited – in Japan. The calculation of the prefactor τ_0 of Néel's single-particle relaxation law having been, much later, improved by Brown [111], this relaxation law was called the Néel–Brown model after this model was checked for the first time in 1997 using a 25-nm single Co nanoparticle.

In the case of temperatures smaller than inter-particle dipolar interactions, the glassy nature of a possible dipolar "supermagnetic order" was not really considered by Néel, even if it underlay his early interpretation of the Rayleigh law [40–43]. It is currently admitted that interparticle random dipolar fields should lead to a "super spin-glass" order. The question of the random fields, just a tool for Néel, was later extensively studied, not only in the spin-glass context, but also in the completely different context of diluted antiferromagnets and random anisotropy magnets in the 1980s – see, in particular, the famous and simple Imry and Ma's argument [112], which was challenged by several authors and then rehabilitated [113]. Numerical and experimental tests were done later, *e.g.*, in rare-earth-based random anisotropy systems where the ratio of the random field by the exchange can be varied regularly from one system to another one.

The next step in this field of micro/nano-magnetism was the study of "layers and multilayers," a field that became, as it is well known, extremely important and popular after the discovery of the giant magnetoresistance of Fe/Cr multilayers by Albert Fert [114] and Peter Grünberg [115], and the considerable prospects of their applications. When Néel started working in this field, in 1962, he had many ideas that led him to study theoretically and experimentally, with his colleagues, several basic and applied problems relative to these new systems. Note that he had previously studied surface magnetism with, *e.g.*, his concepts of surface anisotropy [116] resulting from the suppression of all the neighbouring atoms on one side of the sample surface – what we now call "symmetry-breaking surface anisotropy".

Clearly, the experimental techniques for thin-film elaboration and characterization were far from being as developed as they were 30 years later but, nevertheless, they had given important results on thin-films magnetism which led, in particular, to the introduction of several new concepts such as, besides the "symmetry-breaking surface anisotropy," those of "exchange anisotropy," "interlayer couplings," and "surface roughness" (see below).

When, in 1962, he tackled this issue, Néel determined the magnetic charge distributions and magnetic energy of a tri-layer system made up of two ferromagnetic layers separated by a non-magnetic spacer [117]. He then modelled different types of possible couplings between the two ferromagnetic films [118]: an obvious antiparallel coupling resulting from the closure of the dipolar field going from one ferromagnetic layer to the next one and another one, more demanding, associated with interfaces roughness – "like an orange skin" as it was already written. Here, he transposed his ideas of 3D cavities at the origin of his dispersion fields, to 2D roughness. These basic results were supported by an intense experimental activity in his group with characterizations, magnetic measurements, Kerr observations of different layered systems – in general tri-layers. Interestingly, this led to the observation of a third, unknown, coupling mechanism with a metallic intermediate layer and below a certain thickness (*e.g.*, Cr or Pd) [119,120]. This mechanism was later attributed to the already published but apparently not yet very well-known RKKY interaction.

Néel's group also elaborated and studied multi-layers – this is the term that they already used – made up of four magnetic layers [121], which constitutes a significant achievement. Those first studies, surprisingly ahead of their time, were followed by several others with the aim of using trilayer systems as magnetic memories [122,123]. It cannot have escaped anyone that some of those structures were very similar to those more recently used in spintronic applications. However, at the time, the films were thicker and of lower quality. For example, the thicknesses of the outside layers were of the order of 1000 Å and those of intermediate ones above 100 Å. The roughness was also much more important. Finally, these studies, precursors of several aspects of micro/nano-magnetism, never included magnetoresistance studies.

One cannot finish this section without mentioning the discovery of the "antiferromagnetic hysteresis," currently known as "exchange bias" or "exchange anisotropy". This phenomenon is characterized by a shift, in the direction opposite to the initial saturation field, of the hysteresis loops of a ferromagnetic film coupled with an antiferromagnetic one. It was

observed for the first time [124] on a film of Co with an epitaxial CoO layer – due to a light surface oxidation – and promptly interpreted by Néel [125,126], showing in detail how the rotation of the magnetization of the ferromagnetic film is delayed by the antiferromagnetic layer, which is much more robust against a magnetic field. This phenomenon has been used in spin-valve magnetic memories [127] to enable the reversal of only one of the two ferromagnetic films of the junction, the other one being pinned by the antiferromagnetic layer.

6. Conclusion

We hope that this relatively short overview of Néel's works, with a particular emphasis on his CRAS publications, offers a global insight into his whole career and the driving forces behind it. Coming after Curie, Langevin, and Weiss, his immediate predecessors, he really laid the foundations of classical magnetism, a science so deeply rooted in the past and still rich in modern developments, and of which he is considered as one of the fathers. The most important of Néel's breakthroughs, which conditioned his entire career, were a consequence of an essential assumption that he made at its beginning: the molecular field is not a simple static mean field, but it can fluctuate in time (thermal fluctuations) and in space (different magnetic environments).

As indicated in the beginning of this paper, this assumption exploited Heisenberg's discovery of short-range exchange interactions [3], showing that Néel's classical magnetism is, in fact, based on quantum mechanics – a theory that he was sometimes accused of not believing in, an accusation that we see is false. Having not been trained in quantum mechanics, as was often the case at that time, Néel simply followed his common sense, of which he was not lacking. In fact, his way of working generally began with a simplification, as much as possible realistic, of the problem so as to fit in with his ideas and with possible calculations; then a mathematical analysis, rigorous or with clearly indicated approximations; and finally a confrontation with experiments. His ideas were generally carefully argued, as shown for example with his local molecular field fluctuations where, using probabilistic calculations, he started by showing how the fluctuations grow when the number of neighbours decreases.

By publishing an important part of his work in the CRAS, Néel contributed greatly to the influence of this journal, continuing a tradition dating back to the 17th century that he himself tried to pass on to his colleagues and co-workers, as it can be seen in the reference list. The basic nature of Néel's CRAS publications makes them highly relevant for a long time to come, even if today old founding works are not always quoted as they should be… giving, maybe, some reason to write this paper.

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