



The Philips NatLab years: 1964–1994[☆]

P.J. van der Zaag^{a,*}, R. Coehoorn^b

^a Molecular Biophysics, Zernike Institute for Advanced Materials, University of Groningen, Nijenborgh 4, 9747 AG Groningen, the Netherlands

^b Group Molecular Materials and Nanosystems, Department of Applied Physics and Science Education, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, the Netherlands

ARTICLE INFO

Keywords:

Rare earth alloys and compounds
Permanent magnets
Metal hydrides
Magnetisation
Phase diagrams
Magneto-optical recording media

ABSTRACT

This article, written in honor of Jürgen Buschow on the occasion of his 90th birthday, discusses his work during a 30-year career at the Philips Research Laboratories in Eindhoven (The Netherlands), traditionally called the NatLab. We comment on this unique research environment and on Jürgen's way of working, and focus on his scientific work in the fields of permanent magnets, metal-hydrides for rechargeable batteries, and materials for magneto-optical recording, to which he made seminal and lasting contributions.

1. Introduction

In 1964, Prof. K.H.J. (Jürgen) Buschow joined the Philips Research Laboratories in Eindhoven, The Netherlands (traditionally called in Dutch the Natuurkundig Laboratorium, abbreviated as *NatLab*), after finalizing his PhD study in the Chemistry Department of the Vrije Universiteit in Amsterdam.

Philips Research was at that time organized in three divisions: Physics, Chemistry and Electrical Engineering. Yet, the key organizational units were groups ranging from 20 to 30 researchers, and working on specific fields such as magnetism, metals, theory, spectroscopy and optics (optical recording). Remarkable is that during his years at the NatLab, Jürgen Buschow always worked in the Metals group, in line with the focus of his research work on metallic compounds and their applications. While the Magnetism group at the NatLab focussed, at least until the 1980 s, on magnetic oxides, in particular on ferrites and their application in electronic components and magnetic recording heads. Fig. 1 shows a picture of the Metals group, taken in 1987. Jürgen's research during his 30-year career at Philips will be discussed in the next section.

In a typical research group, about half of the staff had a university degree. Most of these staff members were supported by a technical assistant. Based on the principles of prof. G.H. Holst [1] founder and first director of Philips Research, staff was typically transferred after 5–6 years in the job to take up development roles outside Philips' central research laboratories, within one of its product divisions. A small

percentage of the academic staff members was selected to stay on as principal scientist to form the scientific backbone of the NatLab and the respective research groups. Jürgen Buschow followed this career path and was promoted to principal scientist in 1976, followed by a promotion to research fellow in 1988. This last step was reserved for those principal scientists who through their work and inventions had a significant impact on the company's business, usually through successful innovations being applied in company products. Typically, fellows were those scientists who were considered the intellectual leader of their respective group and its field. In 1994, he retired from Philips Research and moved to the Van der Waals-Zeeman Institute of the Physics Department at the University of Amsterdam, having simultaneously a part-time professorship at the University of Leiden.

During his time at the NatLab, this industrial research laboratory was one of the three major Western industrial laboratories active in the electronics industry, comparable in scope and size to the IBM Labs and the AT&T Bell Laboratories. With the latter a staff exchange program existed, in which Jürgen Buschow took part: in 1977, he spent a year at Bell Labs, which led to several joint publications on hydrides composed of intermetallics containing rare-earth (RE) atoms. Quite some similarities existed between these labs, yet there were also differences. These are well-captured in a Philips Research report based on an exchange visit a few years later by dr. Peter Wierenga and dr. Denise Krol "*Do we want prizes or products?*". This report emphasized that research at the NatLab should focus on developing new products and markets for the Philips company. Throughout the years of its existence, from 1914 to 2023,

[☆] Contribution to a special issue of the Journal of Alloys and Compounds (2024), ed. A. Zhukov, V.G. Harris and E. Brück.

* Corresponding author.

E-mail address: p.j.van.der.zaag@rug.nl (P.J. van der Zaag).

research at the NatLab has led to a host of innovations and new products for the Philips company, such as ferrites, both soft and hard, LOCOS (local oxidation of silicon in IC manufacturing), the magnetic compact cassette system, magnetic resonance imaging (MRI) and various generations of optical disk technologies: CD, DVD and Blu-Ray. Also Jürgen Buschow's research led to new products for Philips, most notably in the fields of permanent magnets and hydride materials for rechargeable batteries.

2. Jürgen Buschow's research

Jürgen Buschow's research programs were focused on the physics and materials science of rare-earth based crystalline and amorphous intermetallic compounds, and on applications of such compounds in permanent magnets, metal hydrides for batteries and magneto-optical recording media. Fig. 2 gives an overview of the numbers of resulting publications (including Philips-internal reports and patents), grouped in five-year periods and subdivided in five categories. Note that, in view of the large total number of publications, a log-scale has been used. The figure shows that most publications were focused on fundamental properties. At the time, certainly in the 1960s, that fitted well to the research philosophy of the laboratory: fundamental research was expected to lead very likely to the discovery of novel phenomena and novel materials with improved properties or radically new functionality.

A strong element of the Philips Research infrastructure was, firstly, an excellent facility for fabricating intermetallic compounds, often

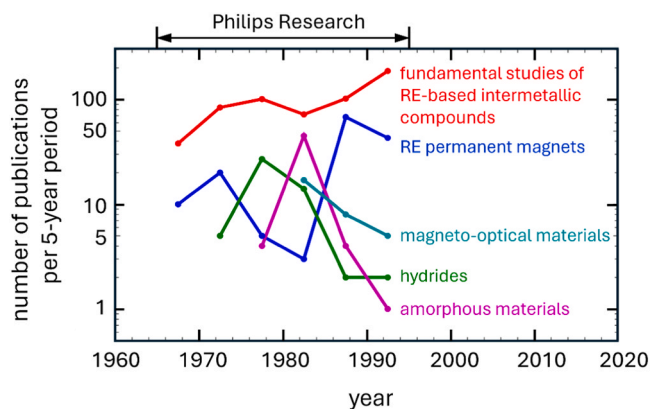


Fig. 2. Publications of Jürgen Buschow in the period 1964 – 1994 during his years at Philips Research, including internal reports and 21 patents, grouped in five-year periods and subdivided into five categories. The total number of disseminations included is 867. One should note that a strict subdivision into categories is not always possible, due to overlapping research themes within a single publication. Source: Ref. 2 (period 1964 – 1989) and Web of Science (period 1991 – 1994).

enabled by arc-melting of a powder mixture of the elemental materials, followed by prolonged heating at temperatures well above 1000 °C in order to approach a thermodynamic equilibrium composition. Other



Fig. 1. Picture of the Metals group at Philips Research (1987), with Jürgen Buschow at the right of the bottom row. Second row, far left: the group leader, ir. Marcel Brouha. Bottom row, third from left: ing. Dick de Mooij, technical assistant, with whom Jürgen Buschow collaborated intensively for more than a decade. Bottom row, far left: one of the authors (RC).

strong elements included characterization tools such as X-ray diffraction equipment, various types of magnetometers, differential scanning calorimetry, and scanning and transmission electron microscopy. Furthermore, fundamental results could readily create the momentum for intensive collaborations with fellow-researchers, also outside the NatLab in one of Philips' product divisions, leading to novel technologies. In addition, Jürgen built up intensive collaborations with university and public-institute laboratories, where specialized equipment such as nuclear magnetic resonance (NMR), electron spin resonance (ESR), Mössbauer spectroscopy, neutron diffraction, single-crystal growth, positron annihilation, and high magnetic fields studies was available. We focus here on three research topics with a strong impact on applications: permanent magnets, hydrides and magneto-optical materials.

2.1. Permanent magnets

Magnetic materials are key components in a wide range of products of diversified electronics companies such as Philips at that time, with applications, for example, in electromotors, magnetic shielding in television tubes, magnetic actuators, magnetic read-write heads for data storage, and magnetic tape recording media. Already in the 1930 s, the NatLab developed a world-wide reputation with the development of 'alnico' hard-magnetic magnet materials [3]. Permanent magnets, also called 'hard-magnetic materials', have a magnetization that can only be reversed when applying a large magnetic field. Alnico magnets are based on iron to which aluminium (Al), nickel (Ni) and cobalt (Co) has been added. The most important success was undoubtedly the discovery and development, in the 1940 s and 1950 s, of relatively inexpensive soft-magnetic and hard-magnetic ferrites. Both classes of materials were based on newly discovered crystal structures. These were formed by iron and oxygen and elements such as nickel (Ni), manganese (Mn) and zinc (Zn), for producing soft-magnetic ferrites, or barium (Ba) and strontium (Sr), for producing hard-magnetic ferrites [4].

In the 1960 s, researchers in the US [5,6] opened the wide research field of the physics, chemistry, materials science and applications of intermetallic compounds formed by combining rare-earth metal atoms with magnetic or non-magnetic transition-metal atoms, or with s/p bonded metal atoms. The RE-series consists of the 15 lanthanide elements, ranging from lanthanum (La) to lutetium (Lu). With increasing atomic number, the 4f orbitals are filled, leading to localized magnetic moments that typically follow Hund's rules. The chemical valence and the atomic volumes vary relatively little across the RE series (with interesting exceptions for nearly-closed-shell or nearly half-filled-shell atoms such as cerium (Ce), europium (Eu) and ytterbium (Yb)). Gradually it became clear that it is therefore often possible to finetune the magnetic properties of RE-based intermetallic compounds without a change of the crystal structure.

This emerging research field was assigned to Jürgen Buschow when he entered the NatLab in 1964 after having completed a PhD study on the optical properties of ionized molecular materials. Such a change of research topic was quite common, and was in line with the principles according to which the laboratory's first director, prof. G.H. Holst, had organized the laboratory [1]. He soon focused on permanent magnet materials of the type SmCo_5 , with samarium as the rare-earth element, and entered a world-wide race for ever larger values of the so-called BH_{max} energy product. The energy product is a measure of the quality of a permanent magnet. It is defined as the externally supplied energy, per unit volume of the magnetic material, that is required to fully reverse the magnetization direction of an optimally shaped magnet, followed by magnetization reversal to the original direction. It follows from a measurement of the magnetization (M) and the magnetic induction (B) as a function of the applied magnetic field (H) under negative field conditions. In 1969, various authors, including Jürgen Buschow, reported almost simultaneously similarly high record values of the BH_{max} product. The Philips work led to 20.2 MG.Oe (MegaGauss.Oersted, the c.g.s. unit used conventionally at that time), corresponding to 164 kJm^{-3} [7].

For a time, this was a world record [8]. The best Alnico and ferrite magnets were surpassed by a factor of ~ 2 and 7, respectively [3]. Fig. 3 shows the published magnetization curve. Interestingly, the length of the communications paper in which this was published was only little more than one page. This work also led to various important patents on SmCo_5 -type and related permanent magnet materials, to numerous extensive crystallographic and phase diagram studies, to highly cited review papers on rare-earth based intermetallic compounds [9,10] and to Philips products based on these materials.

In the 1970 s, the focus in the research program on RE-intermetallics shifted to hydrides for battery applications (see below). Furthermore, the program was extended to include amorphous RE-based materials, made by rapid quenching techniques. One application of amorphous materials is their use as a precursor for novel materials, which can be obtained after controlled heating above the glass temperature. Interestingly, the slow kinetics of the crystallization process can lead to the formation of thermodynamically metastable crystal structures, or extended ranges of solubility. An important result was reported in 1984, when researchers of General Motors (US) used a rapid quenching technique to fabricate nanocrystalline magnet materials of an entirely new ternary compound, $\text{Nd}_2\text{Fe}_{14}\text{B}$ [11]. The compound was actually found to be thermodynamically stable at room temperature. The rapid quenching technique led to a nanocrystalline microstructure with favorable permanent magnet properties. Independently, researchers of the Sumitomo Company (Japan) demonstrated $\text{Nd}_2\text{Fe}_{14}\text{B}$ -based magnets that were fabricated using the more conventional powder-metallurgical method [12].

These breakthroughs urged an immediate change of the program at Philips Research on RE-intermetallics. Rare-earth-based permanent magnets were still of key importance to Philips. An important application was found in compact disc (CD) players, in which a Sm-Co type permanent magnet with coil system was used as an actuator around the optical lens system, to assure a good focus of the laser beam and good track following. The use of much lighter Nd-Fe-B magnets could make the response much faster. Also applications in MRI magnets for patient imaging in hospitals were envisaged. The unexpected finding of an entirely novel ternary compound, with a quite complex crystal structure (68 atoms per unit cell), suggested that many more of such not yet discovered materials might be 'beyond the horizon'. Whether such novel materials would then also provide the required combination of properties, including a large saturation magnetization, a large uniaxial magnetocrystalline anisotropy and a magnetic ordering (Curie) temperature well above room temperature, was yet to be unveiled.

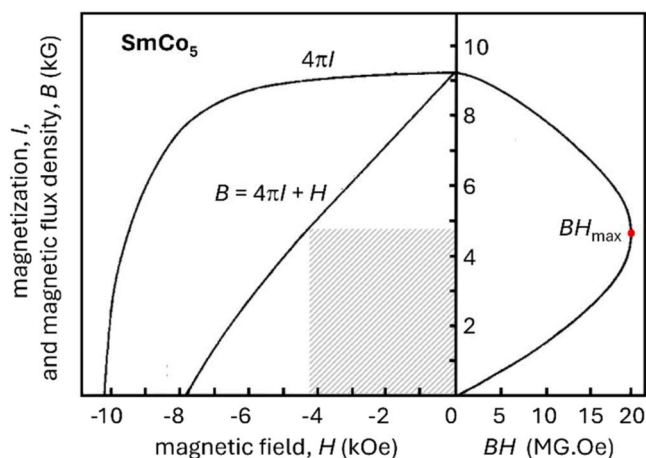


Fig. 3. Magnetization curve of a world-record SmCo_5 permanent magnet, measured in c.g.s. units. I is the intensity of magnetization, B is the magnetic induction (or flux density) and H is the applied magnetic field. The maximum area under the $B(H)$ curve (dashed, 20.2 MG.Oe) is a measure of the magnet's quality. Figure adapted from ref. 7.

The new research program delivered within a few years indeed various new classes of compounds with properties that were suitable for permanent magnet properties: $\text{Nd}_2\text{Fe}_{14}\text{C}$ -based materials, materials of the type $\text{RFe}_{12-x}\text{M}_x$, with R a rare-earth metal atom and M a transition metal or s/p-metal atom, and materials of the type $\text{R}_2\text{Fe}_{17}(\text{C,N})_x$. A review was presented in 1991 [13]. Unfortunately, no improvement with respect to the favorable combination of properties of the $\text{R}_2\text{Fe}_{14}\text{B}$ -type compounds could be found. Even today, Nd-Fe-B type magnets hold the record of the highest energy product, around 460 kJ/m^3 [14].

However, an unexpected finding led to a novel principle for designing permanent magnets. In a search for developing Nd-Fe-B materials with a decreased Nd-content, which are potentially less costly, Jürgen Buschow and his collaborator Dick de Mooij found a nanocomposite material in which the magnetization is mainly due to the soft-magnetic compound Fe_3B , whereas the magnetic hardness is mainly due to a small volume fraction of $\text{Nd}_2\text{Fe}_{14}\text{B}$ [15]. In conventional magnets the easy magnetization direction is fixed and already determined during the fabrication process by grain-orientation in a large magnetic field. However, the easy magnetization direction of these novel materials can still be reoriented in any required direction after fabrication. It was recognized that this arises due to the exchange interaction between the two phases and the small size of the crystallites [16]. The materials were made by rapid quenching followed by an anneal process. As shown in Fig. 4, careful control of the composition is required. The figure also shows that, surprisingly, a $\text{Nd}_2\text{Fe}_{14}\text{B}$ -containing material can be formed by such a process in a part of the phase diagram in which $\text{Nd}_2\text{Fe}_{14}\text{B}$ is not thermodynamically stable. This was the first example of such a type of material, for which the term ‘exchange spring magnet’ has been coined in the literature [17]. This is even today a subject of continued research. Possible applications include permanent magnets with complicated three-dimensional distributions of the magnetization direction.

2.2. Metal hydrides

Fully unexpectedly, the development at the NatLab of SmCo_5 permanent magnets led in 1970 to a discovery that had at least a similarly significant impact. During the production process of the magnets the properties were found to be deteriorated by the uptake of hydrogen [18]. However, it was also found that the material releases hydrogen very easily, and that the uptake takes place in high quantities. Such materials would be well suited as a hydrogen-storage medium. It was soon after discovered that among isomorphous materials the compound LaNi_5 was the most suitable material for that purpose [19]. At room

temperature, the pressure at which reversible absorption/desorption takes place is only 2.5 bar. The final composition is approximately $\text{LaNi}_5\text{H}_{6.7}$.

Jürgen Buschow contributed importantly to the development of these hydride materials by studying the sensitivity of the charging/discharging isotherms upon a partial replacement of the nickel and lanthanum atoms by other transition metal and rare-earth metal atoms. The ease of hydrogen uptake is usually studied by measuring at a fixed temperature the relation between the hydrogen pressure and the amount of hydrogen that is stored in the material. Fig. 5(a) and (b) show some representative results, taken from ref. 20. In panel (a), all studied alternative transition metal elements that replace Ni were found to lead to a smaller hydrogen uptake capacity. Panel (b) shows the effect of replacing lanthanum by various other RE elements. The maximum hydrogen uptakes remains the same, but the required pressure increases. These studies led to various technology-oriented patents. Jürgen

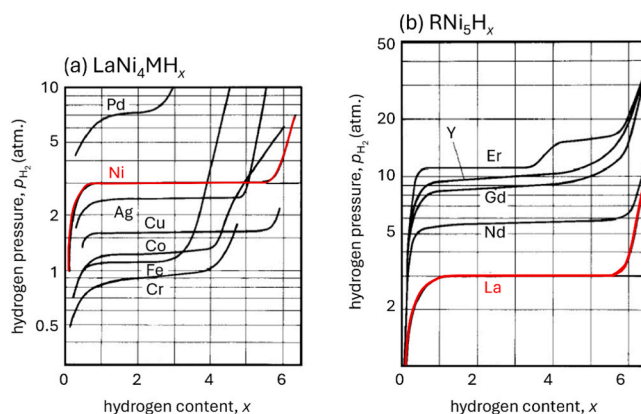


Fig. 5. Isotherms at 40°C for various LaNi_5 -type hydrides, showing the relation between the hydrogen pressure and the amount of hydrogen (the number of hydrogen atoms per formula unit, x), that is stored in the material at a fixed temperature. (a) Isotherms for various LaNi_4M compounds, with M as indicated in the figure. For all alternative systems studied, the maximum hydrogen uptake is less than for the $\text{M} = \text{Ni}$ reference system (red curve). (b) Isotherms for various RNi_5 compounds, with R the rare earth elements La, Nd, Gd and Er, or Y (no RE element, but chemically very similar). For all alternative systems studied, the required pressure is larger than for the lanthanum reference case (red curves). Figure adapted from ref. 20.

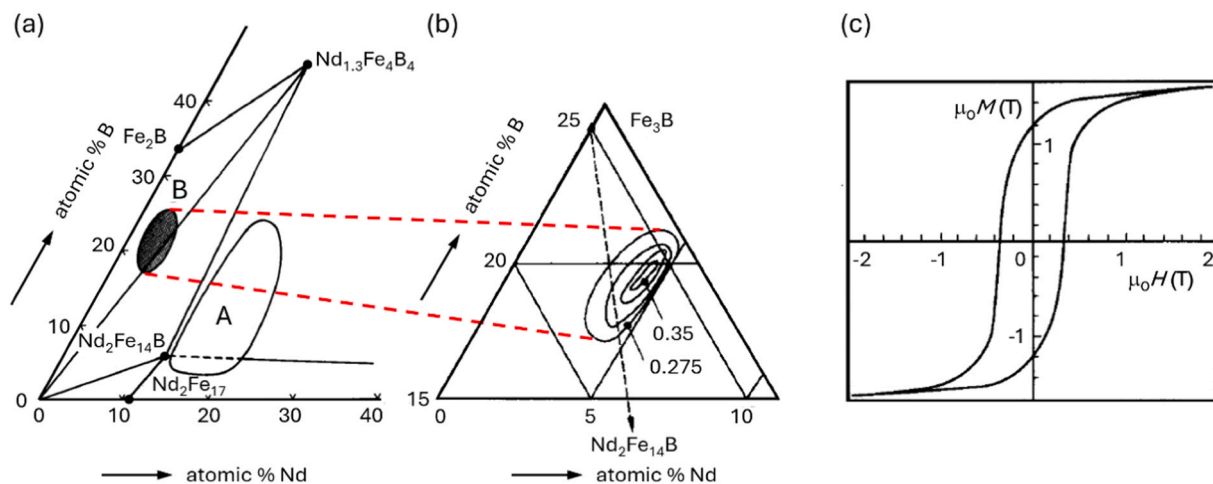


Fig. 4. (a) Fe-rich part of the ternary Nd-Fe-B phase diagram, showing the composition region A of conventional $\text{Nd}_2\text{Fe}_{14}\text{B}$ magnets and the region B where nanocomposite $\text{Fe}_3\text{B}:\text{Nd}_2\text{Fe}_{14}\text{B}$ magnets can be fabricated. (b) Contours of equal coercive field (expressed as $\mu_0 H_c$ in units T) of the nanocomposite magnets. (c) Magnetization (M) as a function of the applied field (H) of optimized nanocomposite magnets, with a composition $\text{Nd}_{4.5}\text{Fe}_{7.7}\text{B}_{18.5}$. The coercive field is 0.35 T . Panel (a) adapted from ref. 16a. Panels (b) and (c) adapted from ref. 16b.

furthermore studied the effects of hydrogen uptake on the magnetic properties of magnetic intermetallic compounds and the mechanism of hydrogen absorption. That work was carried out in part during his stay in the AT&T Bell Laboratories in 1977 [21], resulting in the view that upon hydride formation ‘complexes’ between the rare-earth and the hydrogen atoms are formed.

The application for which this discovery had the most impact was rechargeable batteries. Many properties of the new hydrides were favorable. For example LaNi_5 -hydrides remain electrically conducting when charged. And repeated charging and discharging leads to crumbling of the material (‘decrepitation’), caused by swelling upon absorption by approximately 25 %. As a result, a fine powder is formed with a large surface area to volume ratio, which allows fast charging and discharging. A well-cited short review of the technical development of such batteries, focusing also on more practical aspects such as the sorption kinetics, the change of the storage capacity by repeated cycling, and the assembly of the battery, was written by Willems and Buschow [22]. At that time, finetuning had led to an optimized composition $\text{La}_{0.8}\text{Nd}_{0.2}\text{Ni}_{2.5}\text{Co}_{2.4}\text{Si}_{0.1}$. In 2001, authors of a review paper in Nature reported that more than one billion of LaNi_5 -hydride type batteries were sold per year [23].

2.3. Magneto-optical recording media

Another area where Jürgen Buschow’s expertise led to significant impact and success was in materials for magneto-optical recording media. In the early 1980 s, Buschow and his collaborator Piet van Engen were searching for suitable materials for magneto-optical (MO) recording devices to enable read-write optical recording, in addition to the read-only CD, which Philips would launch as a product in 1982. This prompted them to search for ferromagnetic materials with a high magneto-optical Kerr rotation, i.e. a large rotation of the polarization direction of light upon reflection at the magnetic material surface, when such a material is magnetized in a direction perpendicular to the surface. The sign of the rotation angle depends then on the sign of the magnetization direction angle (up or down), thus enabling the read-out of a magnetic bit structure. They explored the magneto-optical properties of the class of ferromagnetic Heusler alloys, motivated by the notion that the number of intermetallic compounds for which the Kerr rotation had been studied was quite limited at that time. Kerr rotation is a relativistic effect, and requires combining magnetic atoms such Mn, Co, Fe or Ni with heavy atoms with a large spin-orbit interaction for the valence

electrons. Heusler alloys represented a particularly interesting class of intermetallics as the constituents can be varied widely. Buschow and Van Engen soon discovered that the Heusler alloy PtMnSb shows a record-high Kerr effect, with at room temperature a rotation angle of -1.27° at a wavelength of 720 nm [24]. The photon energy dependence of the Kerr rotation is shown in Fig. 6(a). Moreover, this material showed a Curie temperature (magnetic ordering temperature) T_C of 582 K, well above room temperature. A further practical manufacturing advantage was that PtMnSb retained its crystal structure and the high Kerr effect after heating to several hundreds of K above the Curie point.

The high Kerr effect of PtMnSb results from a combination of reasonably high values of the off-diagonal elements and rather low values of the diagonal elements of the dielectric tensor. Substitution of each of the three constituent atoms with related magnetic or non-magnetic atoms was tried, but was found to diminish greatly the magneto-optical effect [24]. An explanation of the large Kerr rotation came from band structure calculations done in collaboration with prof. R.A de Groot of the Radboud University (Nijmegen). In a highly cited first publication (over 5000 times according to Google Scholar to date) [25], the authors reported that magnetic Heusler alloys with a Cl_B crystal structure can be half-metallic semiconductors: there is a band gap for electrons of the minority-spin electrons, while there is no band gap for the majority-spin electrons. The effect was demonstrated for the compound NiMnSb . The authors wrote “we have the remarkable situation here that the conduction electrons at the Fermi level are 100 % spin polarized. This property may exist for *some* of the conduction electrons in other ferromagnets, ..., but in the present materials the unusual situation exists that the spin polarization entails *all* of the conduction electrons” [25].

In the same paper, the authors showed that also PtMnSb would be expected to be a half-metallic semiconductor, provided that the spin-orbit splitting of the Pt-5d states could be neglected. In subsequent work [26], they showed that due to a large spin-orbit splitting of the top of the minority spin valence band a more complex situation arises (see Fig. 6(b)). Optical excitations from the occupied $m = -1$ states near the Fermi energy are not cancelled by excitations from the $m = +1$ states, as these are empty. This unbalance was argued to explain the observed large Kerr rotation.

Practical applications of PtMnSb for magneto-optical recording were hampered by difficulties in developing appropriate thin-film materials. However, this work strongly boosted the field of spin-polarized transport, which flourished in the 1990 s after the discovery of the giant-magnetoresistance effect [27,28]. Both worldwide and also at Philips

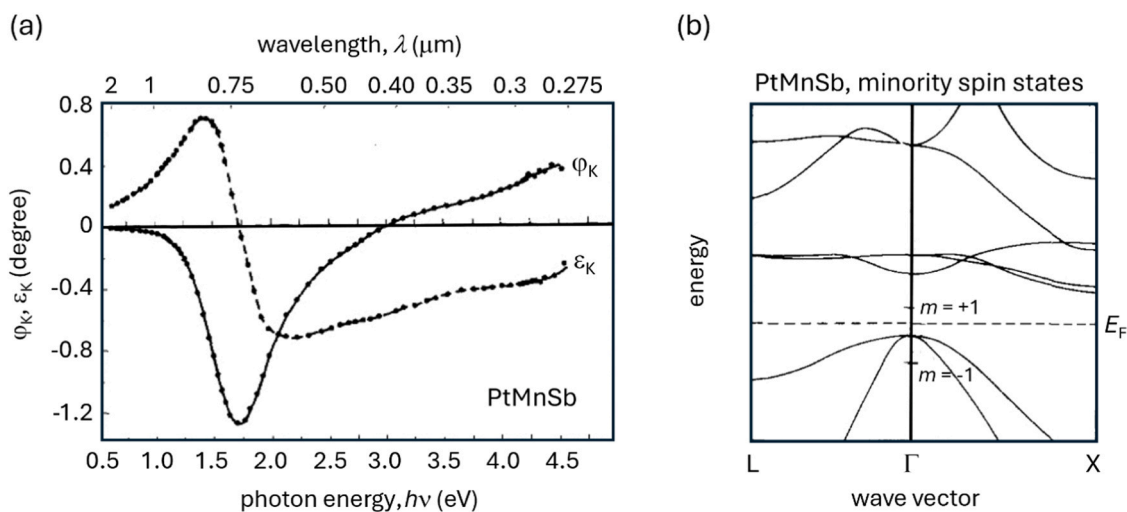


Fig. 6. (a) Photon energy dependence of the magneto-optical Kerr rotation (ϕ_K) and ellipticity (ϵ_K) for PtMnSb . (b) Calculated band structure for minority-spin electrons in PtMnSb from the center of the Brillouin zone (Γ -point) to the L and X points, neglecting spin-orbit interaction. Dashed line: Fermi energy E_F . The figure indicates the spin-orbit splitting of the top of the valence band to $m = \pm 1$ states. Figure (a) adapted from ref. 24. Figure (b) adapted from ref. 25.

Research extensive work was done, on artificial systems in the form of metallic multilayers and on ultrathin layers of magnetite Fe_3O_4 , as electron transport in Fe_3O_4 is also spin-polarized [29]. Unfortunately, maintaining this property in ultrathin layers of Fe_3O_4 and especially for electron transport across an interface has proven to be difficult [30].

3. Jürgen Buschow's way of working and impact

Fig. 2 shows that Jürgen Buschow had a phenomenal scientific output over the years, culminating to a nearly one paper per week output in the last decade of his career at the Philips' NatLab. This productivity reflected Jürgen's strong drive, and was also the result of his knowledge on intermetallic compounds, the excellent facilities at the NatLab for making these materials, and Jürgen's extensive network in the academic community. Also the number of Jürgen Buschow's granted patent filings, 21 during his Philips career, was quite exceptional compared to most of his peers in the laboratory. Half of these filings (10) concerns permanent magnets and their manufacturing. There are four on metal hydrides for batteries, two on magnetic optical recording media and three on invar alloys, a topic that we did not discuss here.

In various roles, Jürgen established and maintained strong academic-industrial relationships. Not just by collaborating intensively with many academic groups worldwide, as discussed above. But also as a supervisor of various PhD students, who did the daily work in Eindhoven and obtained their degree at the universities of Delft, Leiden or Amsterdam. And as a part-time professor at the University of Leiden, active participant of various European projects, (co)organizer of conferences, editor of the Journal of Less-Common Metals (after 1991 the Journal of Alloys and Compounds), and editor of many volumes of the Handbook of Magnetic Materials. This was stimulated and facilitated by the NatLab management, who in line with one of G. Holst's rules/policies [1] recognized that having such relationships would be beneficial. A strong 'ecosystem' would reinforce current projects, provide researchers with new ideas and would prove helpful in attracting young talented researchers.

Both authors of this contribution have experienced Jürgen's stimulating role as a mentor for new colleagues at the beginning of their careers at Philips NatLab. One of us (PJvdZ) joined the NatLab in 1990 in what were turbulent times. The Philips company went through a comprehensive reorganization. Consequently, the projects that were assigned to young researchers were much more focused on subjects expected to have a quick return on investment for the company. It was quite a shock to be assigned to work on the "conventional" topic of soft-magnetic MnZn ferrites. Yet, interestingly, a predecessor had still managed to minimize the energy loss upon ac-cycling, by studying sintered materials with grain sizes smaller than the domain wall width of about 4 μm . It was previously thought that in such a situation an unfavorable complex domain structure would be formed. However, a neutron depolarization study showed this not to be the case: the grains were found to be monodomain below a size of 4 μm and transitioned from two-domain (just above 4 μm) to a closure-domain structure (above 12 μm). After the required internal-Philips review, Jürgen Buschow browsed friendly through it in his cigar-smoke filled room, and decided that he could accept it for publication. Given the circumstances, having quickly a first paper as principle author published was an important step. Thanks to an invitation from Jürgen this work and its implications would later be presented at the European Conference on Magnetic Materials and Applications [31]. So Jürgen has been very effective in helping young researchers entering the field of magnetism, and to get people's attention for their research as he felt needed.

The second author of this contribution (RC) was already inspired before joining the NatLab by Jürgen Buschow's work on half-metallic ferromagnets, as discussed in Section 2.3. That stimulated his interest in a position at Philips Research. As a direct colleague of Jürgen in the Metals group, since 1985, he was readily introduced by him into the subject of permanent magnets and into the European network Concerted

European Action on Magnets (CEAM), formed by top-experts. Jürgen taught him to highly value a research strategy combining physics and materials science based on comparative studies for large numbers of materials systems, instead of just one or a few systems. This way, he profited strongly from such studies by Jürgen and his collaborators on the properties of rare-earth intermetallic compounds when developing a systematic theoretical understanding of their properties by using electronic structure calculations [32]. Fruitful collaborations led also to novel permanent magnet materials based on $\text{Nd}_2\text{Fe}_{14}\text{C}$ and Fe_3B , as described in Section 2. RC also greatly acknowledges the opportunity that Jürgen offered him to write a review in a book chapter on spin-valve type magnetic multilayers showing the giant magnetoresistance effect, with applications to read heads for magnetic tape and disk recording [33].

4. Summary

In this short communication, we have highlighted some of the key contributions that Jürgen Buschow made in the fields of permanent magnets, metal hydrides and magneto-optical recording media during his 30-years career at the Philips NatLab. We have tried to put this into context with the way of working at the NatLab. The works described in this paper illustrate how Jürgen Buschow thrived in this environment to become the scientist with the highest number of publications in the NatLab's history, and a top inventor who contributed to a wide range of technologies. In addition, he was an inspiring colleague and mentor, establishing strong industrial-academic relationships. His work has largely contributed to the impact of Philips' NatLab research on science as well as on the electronics industry, for many decades.

CRediT authorship contribution statement

P.J.van der Zaag: Writing – original draft, Writing – review & editing. **R. Coehoorn:** Writing – original draft, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability

Data will be made available on request.

Acknowledgement

The authors thank their former Philips NatLab colleague prof. F. Roozeboom for reviewing the manuscript and for providing useful comments.

References

- [1] (a) H.B.G. Casimir, Gilles Holst, pioneer of industrial research in the Netherlands, *Philips Tech. Rev.* **40** (1982) 121.(b)ibid. Haphazard Reality: Half a Century of Science, Harper & Row, 1984. In this autobiography, prof. Casimir, one of Holst's successors, summarizes the principles according to which Holst had organized the NatLab in the form of a set of ten rules.
- [2] Bookseries "Verzameling van Publikaties en Octrooien van Prof. dr. K.H.J. Buschow", Philips Electronics N.V., 1994.
- [3] U. Enz, Magnetism and magnetic materials: historical developments and present role in industry and technology, in: E.P. Wohlfarth (Ed.), Ch. 1 in *Ferromagnetic Materials*, vol. 3, North Holland Publishing Company, Amsterdam, 1982.
- [4] J. Smit, H.P.J. Wijn, Ferrites, Philips Technical Library, 1959.
- [5] E.A. Nesbitt, H.J. Williams, J.H. Wernick, R.C. Sherwood, Magnetic moments of intermetallic compounds of transition and rare-earth elements, *J. Appl. Phys.* **33** (1962) 1674.

- [6] K. Strnat, G. Hoffer, J.C. Olson, W. Ostertag, J.J. Becker, A family of new cobalt-based permanent magnet materials, *J. Appl. Phys.* 38 (1967) 1001–1002.
- [7] F.F. Westendorp, K.H.J. Buschow, Permanent magnets with energy products of 20 million gauss oersteds, *Sol. State Comm.* 7 (1969) 639.
- [8] J.H.N. van Vucht, Intermetallic compounds: background and results of twenty years of research, *Philips Techn. Rev.* 36 (1976) 136.
- [9] K.H.J. Buschow, Intermetallic compounds of rare-earth and 3d transition metals, *Rep. Prog. Phys.* 40 (1977) (1977) 1179.
- [10] K.H.J. Buschow, Rare Earth compounds, Ch. 4, in: E.P. Wohlfarth (Ed.), in *Ferromagnetic Materials*, vol. 1, North Holland Publishing Company, Amsterdam, 1980.
- [11] J.J. Croat, J.F. Herbst, R.W. Lee, F.E. Pinkerton, Pr-Fe and Nd-Fe-based materials: a new class of high-performance permanent magnets, *J. Appl. Phys.* 55 (1984) 2078.
- [12] M. Sagawa, S. Fujimura, N. Togawa, H. Yamamoto, Y. Matsuura, New material for permanent magnets on a base of Nd and Fe, *J. Appl. Phys.* 55 (1984) 2083.
- [13] K.H.J. Buschow, New developments in hard magnetic materials, *Rep. Prog. Phys.* 54 (1991) 1123.
- [14] R. Skomski, P. Manchanda, P. Kumar, B. Balamurugan, A. Kashyap, D.J. Sellmyer, Predicting the future of permanent-magnet materials, *IEEE Trans. Magn.* 49 (2013) 3215.
- [15] D.B. de Mooij and K.H.J. Buschow, *Magnetic Material Comprising Iron, Boron and A Rare Earth Material*, US patent US4935074A (1986).
- [16] (a) K.H.J. Buschow, D.B. De Mooij, R. Coehoorn, Metastable ferromagnetic materials for permanent magnets, *J. Less Comm. Met.* 145 (601) (1988);
(b) R. Coehoorn, D.B. de Mooij, C. de Waard, Meltspun permanent magnet materials containing Fe₃B as the main phase, *J. Magn. Magn. Mater.* 101 (1989) 1989.
- [17] E.F. Kneller, R. Hawig, The exchange-spring magnet: a new material principle for permanent magnets, *IEEE Trans. Magn.* 27 (1991) 3588.
- [18] H. Zijlstra, F.F. Westendorp, Influence of hydrogen on the magnetic properties of SmCo₅, *Sol. State Comm.* 7 (1969) 857.
- [19] J.H.N. van Vucht, F.A. Kuijpers, H.C.A.M. Bruning, Reversible room temperature absorption of large quantities of hydrogen by intermetallic compounds, *Philips Res. Rep.* 25 (1970) 133.
- [20] H.H. Van Mal, K.H.J. Buschow, A.R. Miedema, Hydrogen absorption in LaNi₅ and related compounds: experimental observations and their explanation, *J. Less Comm. Met.* 35 (1974) 65.
- [21] K.H.J. Buschow, R.L. Cohen, K.M. West, What is the mechanism of hydrogen absorption in rare-earth intermetallics? *J. Appl. Phys.* 48 (1977) 5289.
- [22] J.J.G. Willems, K.H.J. Buschow, From permanent magnets to rechargeable hydride electrodes, *J. Less Comm. Met.* 129 (1987) 13.
- [23] L. Schlapbach, A. Züttel, Hydrogen-storage materials for mobile applications, *Nature* 414 (2001) 352.
- [24] P.G. van Engen, K.H.J. Buschow, R. Jongebreur, M. Erman, PtMnSb, a material with very high magneto-optical Kerr effect, *Appl. Phys. Lett.* 42 (1983) 202.
- [25] R.A. de Groot, F.M. Mueller, P.G. van Engen, K.H.J. Buschow, New class of materials: half-metallic ferromagnets, *Phys. Rev. Lett.* 50 (1983) 2024.
- [26] R.A. de Groot, K.H.J. Buschow, Recent developments in halfmetallic magnetism, *J. Magn. Magn. Mater.* 54 (1986) 1377.
- [27] M.N. Baibich, J.M. Broto, A. Fert, F. Nguyen Van Dau, F. Petroff, P. Etienne, G. Creuzet, A. Friederich, J. Chazelas, Giant magnetoresistance of (001)Fe/(001)Cr magnetic superlattices, *Phys. Rev. Lett.* 61 (1988) 2472–2475.
- [28] G. Binasch, P. Grünberg, F. Saurenbach, W. Zinn, Enhanced magnetoresistance in layered magnetic structures with antiferromagnetic interlayer exchange, *Phys. Rev. B* 39 (1989) 4828.
- [29] P.J. van der Zaag, P.J.H. Bloemen, J.M. Gaines, R.M. Wolf, P.A.A. van der Heijden, R.J.M. van de Veerdonk, W.J.M. de Jonge, On the construction of an Fe₃O₄-based all-oxide spin valve, *J. Magn. Magn. Mater.* 211 (2000) 301.
- [30] P.J. van der Zaag, O. Fitchorova, A. Sokolov, V.G. Harris, Ferrite films: deposition methods and properties in view of applications. in *Modern Ferrites: Basic Principles, Processing and Properties 1*, Wiley, 2022, pp. 295–411.
- [31] P.J. van der Zaag, New views on the dissipation in soft magnetic ferrites, *J. Magn. Magn. Mater.* 196-197 (1999) 315.
- [32] R. Coehoorn, First principles band structure calculations for rare earth-transition metal compounds: magnetization, hyperfine parameters and magnetocrystalline anisotropy, *J. Magn. Magn. Mater.* 99 (1991) 55.
- [33] R. Coehoorn, Giant magnetoresistance and magnetic interactions in exchange-biased spin-valves, Ch. 1, in: K.H.J. Buschow (Ed.), in *Ferromagnetic Materials*, vol. 15, North Holland Publishing Company, Amsterdam, 2003.