

# Magnetism in Metals

A Symposium in Memory of Allan Mackintosh

Copenhagen, 26–29 August 1996

Invited Review Papers

*Edited by* D.F. McMorrow, J. Jensen and H. M. Rønnow

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## Preface

The Symposium on Magnetism in Metals took place at The Royal Danish Academy of Sciences and Letters in Copenhagen on 26-29 August 1996. The Symposium was conceived by Allan Mackintosh with the aim of bringing together a wide group of international experts in the field to discuss and review recent developments and new perspectives in magnetism research. The Programme Committee comprised Allan, Leo Falicov, Jens Als-Nielsen, Ole Krogh Andersen and myself, but Allan steered us skillfully in the choice of invited speakers. 1996 was to be the year of his sixtieth birthday and he was proud that Copenhagen had been chosen as that year's European Capital City of Culture. He had planned the programme, and had secured generous financial support for the meeting.

Alas, it was not to be. We were greatly saddened by the death of Leo Falicov in January 1995. The news of Allan's sudden death in December 1995 was a further dreadful shock for us all. We believed that Allan would have wanted us to go ahead with the Symposium, and Jette Mackintosh encouraged us to do so. I was asked to take over as Chairman of the Organising Committee: the programme was modified to include a memorial session on the first day, but otherwise was largely as Allan had planned.

The splendid conference room of the Royal Danish Academy was filled with more than 80 participants: the memorial session was a deeply moving occasion with reminiscences and tributes to Allan from his brother Ian and son Poul, and from some of his closest collaborators and friends, Hans Bjerrum Møller, Ole Krogh Andersen, Jens Jensen, Kurt Clausen and myself. The Symposium continued with the review talks, which are published in this volume. There were also a number of contributions presented as posters, which are not included here. These Proceedings begin with an obituary of Allan together with a list of his impressive range of scientific achievements and his publications.

Allan had also arranged a comprehensive social programme for the participants. The highlight of this was the magnificent Conference Banquet in Tivoli's Nimb Restaurant. We also toured the historic cathedral of Roskilde and the Viking Ship Museum before visiting Risø National Laboratory, our hosts for an excellent dinner.

The Symposium received generous financial support from the Royal Danish Academy of Sciences and Letters, the Carlsberg Foundation, the Novo Foundation, the Danish Natural Science Research Council and Risø National Laboratory, which we gratefully acknowledge.

The success of the Conference resulted from the collective efforts of many people. I would like to take this opportunity to thank Kurt Clausen and Ca Thi Studinski, who looked after all the financial affairs and the organisation of the participants, and Jens Als-Nielsen for the local arrangements at the Academy. Thanks go also to Des McMorrow, Jens Jensen and Henrik Rønnow for transforming the authors' manuscripts into this volume as a permanent record of the Symposium dedicated to Allan's memory.

Keith McEwen

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## Allan Roy Mackintosh, 1936–1995

Allan Roy Mackintosh, who died on 20 December 1995 as a result of a car accident in Denmark, devoted much of his life to the study of the behaviour of electrons in solids. He made major contributions to our understanding of the fundamental electrical and magnetic properties of the rare earth metals. In Denmark, where splendid traditions had been established in astronomy, atomic, nuclear and particle physics half a century earlier, Allan Mackintosh will be remembered for his successful efforts in developing modern solid state physics. Through the European Physical Society and, more recently, within the context of the European Union, he strove to improve the quality and efficiency of physics research through international collaboration.

Born in Nottingham on 22 January 1936, he was educated at Nottingham High School and Peterhouse, Cambridge. His doctoral research was carried out in the Cavendish Laboratory, under the supervision of Sir Brian Pippard, where he investigated the Fermi surface of metals, using ultrasonic attenuation methods. It was also in Cambridge that he met Jette, his Danish wife.

On leaving Cambridge in 1960, he became Associate Professor of Physics at Iowa State University. This move was to shape the direction of his future scientific career. The University's Ames Laboratory had begun to make single crystals of the rare earth metals. The chemical properties of these elements are very similar, and consequently they had only recently been separated into pure form. However, their physical properties, particularly their magnetic behaviour, are very diverse and were, at that time, unexplored territory for the inquisitive physicist. Allan Mackintosh took up this challenge and soon established himself as a leading expert in this new field. His major contributions included the discovery (together with his student Dan Gustafson), by an elegant positron annihilation experiment, that the number of  $4f$  electrons in cerium does not change significantly at the  $\alpha$ - $\gamma$  transition. Showing that Ce is a  $4f$  band metal disproved the then widely accepted promotional model, and was an early contribution to heavy-fermion physics.

In 1963, he spent a sabbatical at the Risø National Laboratory, in Denmark,

where a new research reactor had just become operational. Danish physicists led by Hans Bjerrum Møller were constructing a triple-axis neutron spectrometer to measure phonons in solids. Allan Mackintosh quickly realised the scientific potential of applying this technique to measure spin waves in the rare earth metals. This was the beginning of a most fruitful collaboration that was to contribute substantially to our understanding of rare earth magnetism, and which lasted until the last hours of Allan's life.

In 1966, Allan Mackintosh moved permanently to Denmark and became Research Professor at the Technical University, Lyngby, where he remained until 1970. He brought with him from Ames not only precious rare earth crystals but also T. L. Loucks' relativistic APW programs for performing electronic structure calculations. Allan used them to demonstrate the relevance of computing Fermi surfaces to describe magnetic ordering. He soon taught Danish students how to perform such calculations and asked them to compute the Fermi surfaces of the transition metals, whose complicated  $d$ -band sheets were currently being mapped out by the de Haas-van Alphen technique. It had been known only since 1964 that, unlike the localized  $4f$ -electrons in the rare earth metals, the magnetic electrons in the  $3d$ -transition metals contribute to the Fermi surface; the role of the Coulomb correlation between them was a much discussed topic. Allan had a deep understanding of the behaviour of electrons in metals, and a profound scepticism towards oversimplified theories. Leo Falicov was his life-long discussion partner and close friend. Allan's work not only helped to establish the boundaries of the usefulness of density-functional calculations for  $d$ - and  $f$ -bands systems, but also inspired his students to develop new computational methods.

In 1970 Allan became Professor of Experimental Solid State Physics at the University of Copenhagen. Soon afterwards, at the age of 35, he was appointed Director of the Risø National Laboratory. Prompted by the oil crisis, Denmark had embarked on a national debate about the development of nuclear power for electricity generation. In this frequently heated debate, Allan Mackintosh needed all his diplomatic skills to steer the discussion with factual rather than emotional persuasion.

After 1976, he returned to his Chair in Copenhagen, where he remained until his death. He made many more important contributions to the understanding of the magnetism of the rare earths, which led to him being awarded (jointly with Hans Bjerrum Møller) the prestigious Spedding Prize in 1986. He inspired and motivated all his collaborators and students, and his scientific papers, with their carefully constructed prose, are a pleasure to read. The culmination of his research was the publication (with Jens Jensen) of *Rare Earth Magnetism*, a superbly written exposition of the subject that has already become a classic text. His achievements were further recognised by his election in 1991 to Fellowship of the Royal Society

of London. In Denmark, he was made a Knight of the Dannebrog Order. He was also a Fellow of the Royal Danish Academy of Sciences and Letters, the Danish Academy of Technical Sciences, the Royal Norwegian Scientific Academy and the American Physical Society. Uppsala University awarded him an honorary doctorate of philosophy in 1980.

Although research was always Allan Mackintosh's main priority, his leadership skills were much in demand. His period as director of the Risø Laboratory has already been mentioned, and from 1986 to 1989 he directed NORDITA, with its close connections to the renowned Niels Bohr Institute. He also played an increasingly important role on the European physics scene. A strong believer in the need for international collaboration, he was President of the European Physical Society from 1980 to 1982. Later, he played an important role within the EU Large Scale Facilities programme: he emphasised scientific excellence as the principal criterion for funding.

Allan Mackintosh took great pleasure in music, travelling and his comprehensive collection of malt whiskies. He disguised his enjoyment of sport and physical activity behind a facade of feigned mediocrity. On hill-walking holidays, he divided his energies between humorous discourses as to the pointlessness of climbing the next hill, and making certain that he was the first to the top. Numerous friends will also remember the warmth of the welcome extended to them by Allan and Jette at their home in Denmark.

In later years, his keen interest in the history of physics led him to investigate the mutual influence of Ernest Rutherford and Niels Bohr. He revealed important, but often overlooked, achievements of less well-known scientists, such as John Vincent Atanasoff's key role in the invention of the computer. In one of his last papers, he showed the contribution of Charles Ellis to the discovery of the neutrino. It is symbolic of his interest in the past as well as the future of physics, that he spent the last day of his life selecting experiments to be carried out in 1996 at Risø under an EU - financed programme, and then gave an eloquent seminar on the discovery of the neutrino some 60 years ago.

We have lost one of the finest physicists in magnetism and neutron scattering: the tragic nature of Allan Mackintosh's sudden death makes this loss all the more acute amongst his world-wide circle of friends and colleagues.

## Allan Mackintosh's scientific achievements

Allan worked at Iowa State University between 1960 and 1966, and became interested in the rare earths and electronic structure calculations. During this period, he and his students made the following main contributions:

- (a) The discovery of magnetic superzones [4].
- (b) The first systematic studies of conduction-electron scattering by localized moments [5].
- (c) The demonstration by positron annihilation that the number of  $4f$  electrons in Ce does not change significantly at the  $\alpha$ - $\gamma$  transition [15]. This disproved the validity of the then generally accepted "promotional" model. Since the  $4f$  electrons are itinerant at low temperatures, and have very large masses, this was one of the first contributions to heavy-electron physics. Throughout his life, Allan returned to the problem of understanding the transition between and the duality of the itinerant and localized character of the  $4f$  electrons in Ce and Pr. He motivated his former students in the field of electronic structure calculations to push the border of validity of the itinerant picture [64,80] and, shortly before his death, he and his experimental colleagues succeeded in observing in Pr a new magnetic excitation of itinerant character [79–81].
- (d) The observation of the positive or negative magnetoresistance associated with changes of the magnetic structures in Ho and Dy [16], which allowed a systematic study of the effect of magnetic superzones and spin-wave scattering, and revealed intermediate phases, later identified as helifans [73].
- (e) The determination of the spin-wave energy gap in Tb from resistivity measurements [12]. The deduced value was close to that later measured by neutron scattering [39,42].
- (f) The first direct observation, by positron annihilation, and interpretation, by electronic structure calculation, of the highly distorted Fermi surfaces in the heavy rare earths [23].

In 1963, Allan Mackintosh spent a sabbatical year at Risø and initiated the study of rare earth magnetism in Denmark. On returning permanently to Europe in 1966, he began a long-lasting study, with Hans Bjerrum Møller and their colleagues, of the spin waves in Tb. These experiments resulted in the following advances:

- (a) The first complete study of the spin-wave spectrum for any magnetic system [35,42], allowing the deduction of the magnon density of states, the ther-

modynamic properties, and the detailed form of the exchange interaction in reciprocal and real space, in the ferromagnetic phase.

- (b) Measurements of the magnons in the helical phase, the first detailed studies of the excitations of an incommensurate system [25]. Such phasons, or Goldstone modes, have since proved to be of interest in a number of incommensurate systems. From the dispersion relations, the exchange and its temperature dependence were deduced, clarifying the driving mechanism for the helical-ferromagnetic transition.
- (c) Measurement of the single-ion anisotropy parameters, distinguishing between the crystal-field and magnetoelastic contributions [39].
- (d) Experimental demonstration of the “frozen-lattice” effect, that the energy gap does not vanish when the hexagonal anisotropy is cancelled by a magnetic field [39].
- (e) The observation of anisotropic two-ion coupling [43].
- (f) Detailed studies of the interactions of magnons with phonons [25], with magnetic impurities [28], with each other [42], and with conduction electrons [50].

In recent years, Allan Mackintosh turned his attention to holmium. Together with Jens Jensen, he explained old observations and predicted new effects. The major results are:

- (a) The discovery of the helifan structures [73]. These structures, which are stable in a range of intermediate fields, have many interesting features. This work has solved mysteries in the magnetization, neutron diffraction and transport properties of Ho which have been unexplained for decades, and has excited wide interest.
- (b) The investigation of the effects of commensurability on magnetic excitations [69]. They established that commensurability causes an energy gap in the spin-wave spectrum of Ho at  $\mathbf{q} = \mathbf{0}$ , and that the dipole interaction produces a discontinuity in the dispersion relation, and thereby stabilizes the cone structure at low temperatures. The long-standing mystery of the stability of the cone structure was thereby solved.

The culmination of Allan’s research in the rare earths was the book, co-authored with Jens Jensen [75], which contains many predictions of new effects and suggestions for experimental studies. These have given a considerable stimulus to rare earth research, and a number of such studies have already been initiated.

His most recent research was in examining a possible breakdown of the *standard model* of rare earth magnetism in praseodymium, where a new magnetic excitation was observed [79,81]. This new form of magnetic excitation is interpreted as arising from the dynamical response of the conduction electrons, and is thus the first observed example of a propagating paramagnon.

In 1966 at the Technical University in Lyngby, Allan Mackintosh initiated what later became a school of electronic structure calculation. He and his students were among the first to calculate the electronic structures of transition and rare earth metals, and to demonstrate that the Fermi surfaces obtained with what he called the standard single-particle potential, agree in detail with experimental results [26,27,29,37,41]. For the fcc transition metals Rh, Pd, Ir, and Pt, the excellent agreement with the extremal areas of orbits on the Fermi surface measured via the de Haas–van Alphen effect, allowed him to deduce for each orbit, the mass enhancement due to electron–phonon and electron–electron interactions, and thus to provide the first reliable estimate of the spin-fluctuation enhancement in Pd [26].

The nearly perfect agreement of the complicated Fermi surfaces obtained with this Slater-exchange potential was a strong hint that the local approximation (LDA) to density-functional formalism proposed three years earlier by Kohn and Sham for ab initio computation of ground-state properties might work. This motivated Allan’s former students to develop methods for charge- and spin-selfconsistent LDA calculations. In 1975, as a first application, Allan Mackintosh et al. applied the LMTO method with the standard potential to the heavy hcp transition metals [47]. Apart from demonstrating again excellent agreement with de Haas–van Alphen measurements, they proved the efficiency of the new method and showed how it made the complicated relativistic hcp band structures intelligible in terms of “canonical” *s*-, *p*-, and *d*-bands.

During the second half of the seventies, LDA-LMTO calculations were performed for all elemental *3d*, *4d*, and *5d* metals and the cohesive and magnetic ground-state properties obtained ab initio were surprisingly accurate. In a review of the electronic structure of transition metals, Mackintosh and Andersen [53] explained the LDA bands and their relation to the pressure-volume curves, the crystal structures, and the occurrence of itinerant magnetism. Furthermore, they reviewed the experimental and computed Fermi-surface and optical properties. For the much studied noble metals, Mackintosh et al. [55] investigated whether a local potential exists, which will reproduce not only the experimental ground-state properties, but also the band structures. The answer was: No, but almost.

The influence of Allan Mackintosh on the field of electronic structure calculations went far beyond the research papers he authored. He continued to provoke and motivate his large, international electronic-structure family, but usually refused to put his name on the publications. He preferred writing reviews in which

he also pointed out what to do next [58,64,70,80].

Allan Mackintosh also made important contributions to the history of both physics [82] and computing [67,72].

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