Magnetism in Layered Structures

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Magnetic monolayers and multilayers exhibit a rich variety of unusual magnetic properties arising from their reduced dimensionality. Research on these layered systems is advancing our knowledge of nature and enhancing their technological exploitation.

Transition metal atoms such as Fe, Ni and Co carry a net magnetic moment as a result of their d-shells being unequally populated by spin-up and spin-down electrons. When these atoms join together to form a solid, an effective interaction between their magnetic moments arises by virtue of the Pauli exclusion principle (the so-called exchange interaction). This interaction — which typically concerns only neighbouring magnetic moments (*i.e.* it is of a short range) — may favour parallel or antiparallel alignment of the moments, depending on details of the electronic structure. Moreover, magnetic moments that are far apart are also linked together through the network of nearest neighbour links. As a result, when the temperature is low enough to quench out thermally induced disorder, the solid may exhibit macroscopic long range order (LRO). For parallel alignment, a macroscopic magnetization M arises, owing to alignment in the same direction of all individual spins within large regions of the solid, and the system is said to be in a *ferromagnetic* state.

Ferromagnetism is a very "fragile" and complex phenomenon. In most cases atoms in fact loose their moments in the solid state, where the occurrence of magnetic order is perhaps one of the most difficult problems in quantum mechanics. Even if the ground state (the state at a temperature T = 0 K) is magnetically ordered, any persistence of spontaneous magnetization at finite temperatures is strongly opposed by the entropy of disorder.

It turns out that a crucial parameter governing the occurrence of ferromagnetism at finite temperatures is the *dimensionality* of the system. Magnetism as we know it from our experience is essentially a consequence of the fact that our world is three-dimensional: in a two-dimensional world magnetism would be totally different.

The aim here is to review our theoretical understanding of magnetism in two dimensions and summarize the supporting experimental evidence. One possible physical realization of a 2-d system is a layered structure, the simplest consisting of a few atomic layers on top of a supporting bulk crystal. Despite the geometrical simplicity of this fundamental structure, see Fig. 1, its production represents a formidable problem in materials' science. A strict 2-d translational symmetry is only achieved when the interface between the adsorbed layer and the substrate is perfectly sharp, *i.e.* when layer-by-layer growth occurs. Unfortunately, competing processes such as interdiffusion or clustering leading to the formation of rough interfaces can also interfere.

The engineering of a number of truly 2-d epitaxial layers and multilayers (see Fig. 1) of some 3-d transition metals represents a recent success of materials' science. By "truly 2-d epitaxial" we mean that such systems approach the ideal geometric perfection shown in Fig. 1 over large areas (some 100 to 200 lattice constants). The present article deals with the magnetic properties of these systems.

The 2-d Magnetic Ground State

The magnetic ground state for an epitaxial monolayer essentially results

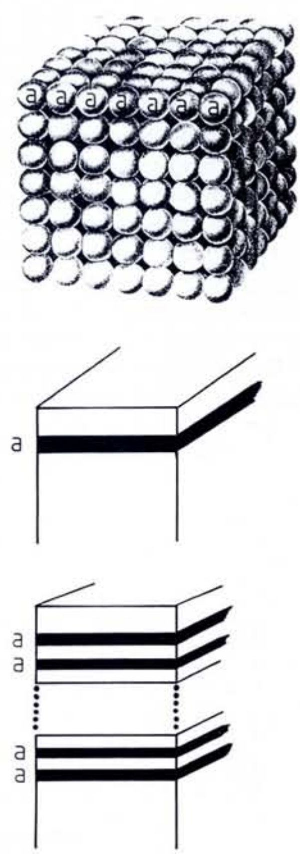

Fig. 1 — Three examples of layered structures. Upper — the magnetic epilayer (a) is vacuum terminated and grows on top of an ordered substrate as a perfect geometrical continuation of the substrate crystal structure. This growth mode, assumed in most computer simulations because of its geometrical perfection, implies that successive layers do not start to grow before the one being built is completed (so-called layer-by-layer growth). The deposition of the magnetic atoms on top of the substrate occurs typically in ultra high vacuum conditions (10^{-10} mbar range) in order to avoid contamination of the growing layer.

Middle — the magnetic layer (a) is sandwiched between the substrate and an overlayer, often used to protect the "delicate" magnetic atoms from contamination after exposure to air. The overlayer can also be used to amplify the magnetic signal from (a), as in the case of neutron total reflection experiments (see text).

Lower — many layers (a) are stacked one top of each other, separated typically by non-magnetic buffer layers. This structure is called a multilayer and is often found in technological applications as a way of achieving a sizeable amount of magnetic matter while maintaining the unique magnetic properties arising from the layer (a) being very thin.

Professor Danilo Pescia will shortly take up his position in experimental physics at the RWTH, D-5100 Aachen to which he was appointed in April 1990. A graduate of the ETH, Zürich where he took a Ph.D. in 1983 in solid state physics, he spent three years as an Oppenheimer Fellow at the Cavendish Laboratory, Cambridge, UK, and four years as a research scientist at KFA, Jülich. He plans to pursue his research in surface physics and thin films.

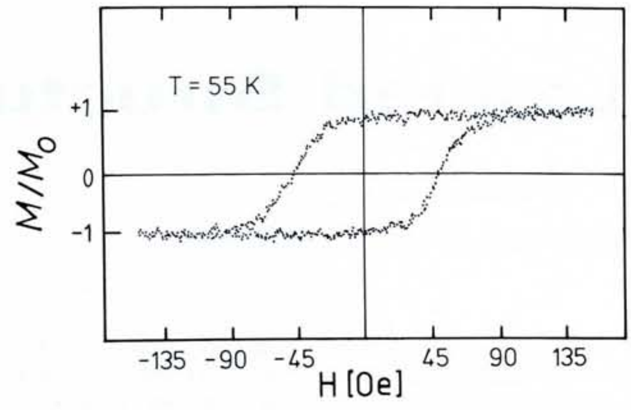

Fig. 2 — Hysteresis curve M(H) at 55 K of a one monolayer (1 ML) thick Co epitaxial film on the surface of a Cu(100) single crystal. The monolayer is coated with a 3 nm layer of Cu, which was also found to grow layer-by-layer on the Co film. The magnetization is normalized to M_0, the value of M at large fields. The technique used to record M(H) is based on the Kerr effect.

from competition between two mechanisms. One is overlap between the magnetic 3-d wave functions of the adsorbate atoms and the non-magnetic wave functions of the substrate. This overlap broadens the magnetic bands so the density of states at the Fermi level may fall below the value necessary for the Stoner criterion to be fulfilled.

The Stoner criterion places a lower limit on the density of states at the Fermi level, below which the formation of a ferromagnetic moment is no longer energetically favoured with respect to the non-magnetic state. In this case the epilayer is ''dead''. On the other hand, the number of nearest neighbours within the adsorbed layer is lower than in the bulk, thus reducing the width of the d-bands. This acts to *increase* the density of states at the Fermi level, thus favouring ferromagnetism and a magnetically ''alive'' epilayer.

A rigorous computational effort, led in particular by A.J. Freeman of Northwestern University, USA, shows that for most monolayers of the common bulk magnets, the state that is magnetically ordered has the lowest energy, *i.e.* there are no ''dead'' layers. The magnetic moments μ of the monolayers are sizeable — at least as large as in bulk — owing to the reduced band width. For a Cr monolayer, for instance, a giant enhancement (**four times** the bulk value of 0.59 μ_B where μ_B is the Bohr magnetron) is predicted.

Studies of 2-d magnetism have also revealed considerable complexity. For example, S. Bluegel and P.H. Dederichs at KFA Jülich discovered, using numerical techniques, the existence — besides the non-magnetic and ferromagnetic solutions — of a third stable energy minimum. This is the $c(2\times2)$ antiferromagnetic state which requires neighbouring spins to orient themselves in opposite directions.

The wealth of theoretical results — all achieved thanks to the latest generation of supercomputers — is now being put under scrutiny by a series of experimental tests.

''Dead'' or ferromagnetic?

A consensus has emerged among experimental physicists that thin films of well known bulk ferromagnets like Fe, Co, Ni and Gd indeed have a ferromagnetic ground state down to an actual monolayer. This is exemplified by Co films on Cu(100) where the response of to an applied magnetic field *H* is a macroscopic magnetization *M(H)* which follows an *hysteresis* curve (Fig. 2). It is particularly important to note that at zero applied magnetic field a spontaneous magnetization persists — a clear signature of long-range ferromagnetic order over macroscopic distances.

The key to monolayer ferromagnetism is the recent development (see *e.g.* [1]) of techniques capable of measuring the magnetic properties of a very small amount of matter (10^{14} atoms instead of 10^{22} for a standard bulk sample). Fig. 2 perhaps represents a textbook example of this development, where the technique used to determine *M* exploited the Kerr magneto-optic effect. Some 100 years old, this technique relies on the intensity of reflected light being slightly affected by the magnetic state of the reflecting mirror. The dependence is very small, and the reflected light usually samples a volume that is ≈ 200 atomic layers deep. Sensitivity to a monolayer was never suspected before it was demonstrated in 1985 by S. Bader at the Argonne National Laboratory, USA.

Magnetic moments

While a string of experimental techniques — including the Kerr effect — are able to distinguish between ferromagnetic order and magnetically ''dead'' layers, the majority of them are not suitable either for determining the absolute value of μ or for detecting antiferromagnetism. An ''unconventional'' alternative is to resort to neutrons as a probe. The interaction of neutrons with magnetic moments is known exactly so they can be used to measure ferromagnetic properties as well as the absolute value of μ in bulk samples. The problem is again one of achieving the required sensitivity to a monolayer given that neutrons typically sample more than 1 mm of matter. At glancing incidence, however, neutrons are totally reflected so the probing depth can be kept to within the first 100 atomic layers from the surface.

By combining this enhanced surface sensitivity with the quantum interference of the neutron waves reflected at the interfaces of the sandwich structures illustrated in Fig. 1, R.F. Willis and coworkers at the University of Cambridge, UK were able to measure systematically the thickness dependence of μ for thin Co-films on Cu(100). In line with theoretical results, the magnetic moment turns out to be at least comparable with the bulk one down to the monolayer range (Fig. 3).

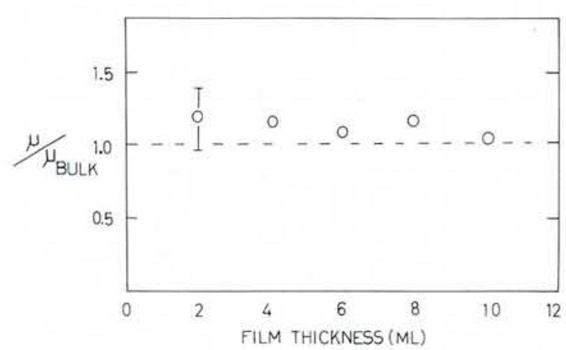

Fig. 3 — Magnetic moments μ of thin Co films sandwiched between Cu films as a function of the thickness of the Co, at 4 K, showing that the bulk value of 1.8 μ_B is maintained even at very small thicknesses. The experimental technique used to measure μ involved the total reflection of spin polarized neutrons.

Perpendicular magnetization

The driving force behind research on layered structures is the prospect of very thin films being perpendicularly magnetized as these would find important applications. Perpendicular magnetization appeals particularly to the magnetic recording industry because it offers an increased storage density. Indeed, reversible perpendicular recording in very thin films is on the verge of large-scale commercialization (of course, for technical applications the magnetic signal from the very thin layer is suitably amplified by multilayering).

Perpendicular magnetism was suggested originally by L. Néel in the 1950's and confirmed recently for model systems by J.G. Gay and R. Richter using numerical calculations. In thin films, the direction of the ground state magnetization is the result of two competing mechanisms. On the one hand, dipole interaction tends to keep the magnetization in the plane of the film, in order to minimize the magnetostatic energy arising from stray magnetic fields associated with the perpendicular magnetization. On the other hand, perpendicular magnetization may lower the energy of the system *via* spin-orbit interaction, relativistic in origin, which allows the spin to "sense" the broken translational symmetry of a thin film. Numerical calculations predict that if the film is sufficiently thin, the magnetic anisotropy arising from the spin-orbit coupling might overcome the dipole interaction and give rise to a perpendicular magnetization.

An example of the intensive research on perpendicular magnetisation is given in Fig. 4: ultrathin epitaxial fcc-Fe films on Cu(100) are remanently magnetized perpendicular to the film plane. Using such elementary units of matter, F. Meier and coworkers at the ETH Zürich have been able to perform reversible perpendicular recording. It also transpires that perpendicular magnetization depends on fine details of the electronic structure: Co-films of similar thickness — see Fig. 2 — are, in fact, magnetized in-plane.

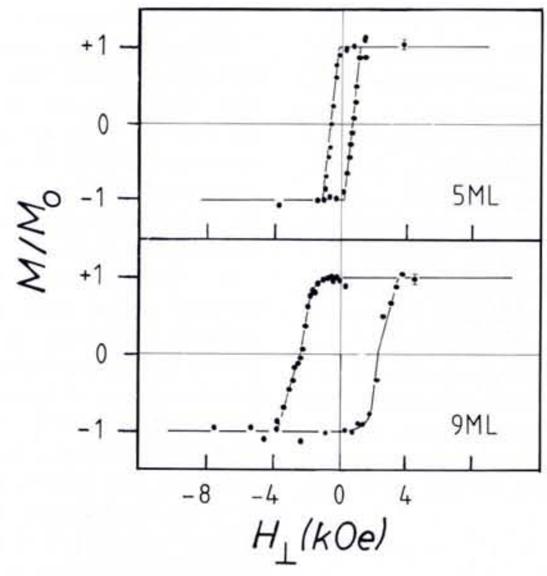

Fig. 4 — Hysteresis curves of vacuum terminated Fe-films (5 and 9 monolayers thick) grown layer-by-layer on Cu(100), recorded at 30 K. The magnetic field is applied perpendicular to the film surface, and the remanence at zero field H is a proof of perpendicular spontaneous magnetization. The technique used to obtain the magnetization curves relied on the emission of spin polarized electrons when the Fe-films are irradiated with ultraviolet light. (The spin polarization of the emitted electrons is proportional to the magnetization.)

So-called spin polarized electron spectroscopies are ideally suited to measuring the magnetic properties of very thin films because they combine a direct determination of M with an extreme sensitivity to thin surface layers (they typically probe surface layers that are only 2-4 atoms thick).

2-d Magnetism at Finite Temperatures

Having established both theoretically and experimentally the ordered nature of the magnetic ground state in epitaxial monolayers, the question arises as to the response of the system to temperature increases. This can be addressed using a criterion suggested by L.D. Landau. Let the spin at the site i of N spins in a volume V deviate by an amount Δ_i from the ground state value S as a consequence of a temperature increase. The stability of long range order is evaluated by examining the thermodynamic limit of the mean square fluctuation per spin

$$\sigma^2 = \lim_{N,V\to\infty} \sigma^2(N,V)$$

with $\sigma^2(N,V) = (1/N)\sum_i <\Delta_i^2>$

where $< >$ means the thermal average. If $\sigma^2(N,V)$ remains finite for $N,V \to \infty$ the ground state spontaneous magnetization is stable against temperature increases and the system has an order parameter $M(T)$. If instead $\sigma^2(N,V) \to \infty$, in the thermodynamic limit the long range order is regarded as unstable, *i.e.* it is destroyed at any finite temperature by the occurrence of strong fluctuations in the spins.

In three dimensions, σ^2 turns out to be finite: this is the theoretical reason why ferromagnetism exists in our world as we know it. In two dimensions however, σ^2 diverges: 2-d long-range order of any kind (magnetic as well as crystalline or superconducting) is unstable against a temperature increase. The absence of $M(T)$ in 2-d systems has been proved rigorously for pure exchange interactions by the celebrated Mermin-Wagner theorem and is the key to the understanding of physics in two dimensions. In fact, the very mathematics which forces σ^2 to diverge ensures that minute deviations from the pure exchange interaction (such as the "small" spin-orbit coupling and dipole interaction, which are typically two to three orders of magnitude smaller than for exchange) are enough to install the full magnetic moment at appreciable temperatures. This extraordinary property is unusual in 3-d systems, where thermodynamics is essentially governed by the exchange interaction.

The full consequences of the highly non-linear response of a 2-d magnetic system to small disturbances are wide ranging and partly unexplored, especially at the experimental level. Two important examples are given in the following sections.

2-d Curie temperature T_2

In bulk materials the 3-d Curie temperature T_3 is essentially given by the exchange energy J times the number of nearest neighbours z, which amounts to T_3 being in the 1000 K range for the classical ferromagnets Fe, Co and Ni. The 2-d Curie temperature, T_2 for pure exchange ferromagnets is exactly 0 K. The relative strength λ of the dipole interaction with respect to the exchange energy amounts to $\approx 10^{-2}$. According to "conventional wisdom", any order induced by the dipole interaction would be destroyed at temperatures exceeding $T \approx \lambda zJ \approx 10$ K! In fact, T_2 is given by:

$$T_2 \approx zJ/(1+c_1 \ln [c_2/\lambda])$$

where c_1 and c_2 are constants. T_2 approaches 0 K as $\lambda \to 0$. The logarithmic term, however, ensures that T_2 varies very slowly with λ and for typical values of $\lambda \approx 10^{-2}$, T_2 is essentially given by the product zJ. The 2-d Curie temperature T_2 is therefore reduced with respect to T_3 owing to the reduced number of nearest neighbours. Nevertheless, it remains in the 100 K range, *i.e.* one to two order of magnitudes higher than expected from "conventional wisdom". The presence of long-range order at appreciable temperatures on the order of zJ is experimentally well documented for Fe, Co, Ni and Gd monolayers.

Phase transitions

A system exhibiting long-range macroscopic order undergoes a phase transition, with LRO disappearing sharply at a well-defined critical temperature T_c. Physicists have formulated the so-called universality hypothesis for the thermodynamics of the phase transition: measurable thermodynamic quantities should show power law singularities with universal critical exponents. "Universal" means that the critical exponents depend only on a small number of parameters, such as the dimensionality of the system or the degree of freedom of the individual spins, but do not depend on either the strength of the exchange interaction or most material constants.

The validity of the hypothesis is well supported by the results of numerous experiments on 3-d systems. It is legitimate to ask how far its validity extends to two-dimensional systems, where LRO is switched on by interactions other than exchange. The answer is not straightforward.

One of the most famous models for ferromagnetic order in statistical physics is the 2-d Ising model. The Ising model consists of a net, with 2-d translational symmetry of spins interacting *via* exchange. The peculiarity of these spins is that all of them can only point in one of two opposite directions. This mathematical abstraction makes the problem exactly solvable and results indeed show power law singularities, *e.g.*,

$$M \sim (1-T/T_c)^{\beta}$$

where the critical exponent is independent or J and μ (*e.g.* $\beta = 1/8$ exactly).

The same holds true for similar mathematical 2-d models with discrete symmetry — *i.e.* β lies within the range $0.10 < \beta < 0.16$, the last digit depending on the exact symmetry of the problem.

The key role of dimensionality is also confirmed by these models: *e.g.* β in 2-d systems is clearly lower than both the bulk value of 0.38 and the value of approximately 0.8 for the surface of a semi-infinite solid. These mathematical models neatly support the validity of the universality hypothesis in two-dimensional systems.

In "real" systems, however, the isotropy of the pure exchange interaction is broken by very small disturbances so that the individual spins have a larger degree of freedom than in the mathematical models. It is not *a priori* evident that discrete mathematical models have any significance for "real" systems. The response of a real system in the vicinity of the phase transition to minute disturbances of the exchange interaction is, however, once again nothing less than extraordinary.

Consider the system over large spatial scales — which are the relevant ones for the thermodynamics of the phase transition. It has been demonstrated by Pokrovskii and by Kadanoff and their coworkers that the strength of these disturbances in the large scale effective Hamiltonian grows exponentially: a symmetry breaking field, no matter how small, will force the system away from the isotropic exchange behaviour into a state of broken symmetry in which only a discrete number of directions are actually allowed. At the phase transition, any real 2-d system is therefore equivalent to some system with discrete symmetry so truly 2-d systems should show power law singularities with critical exponents independent of such parameters as J, the strength of the symmetry breaking field, and even the film thickness.

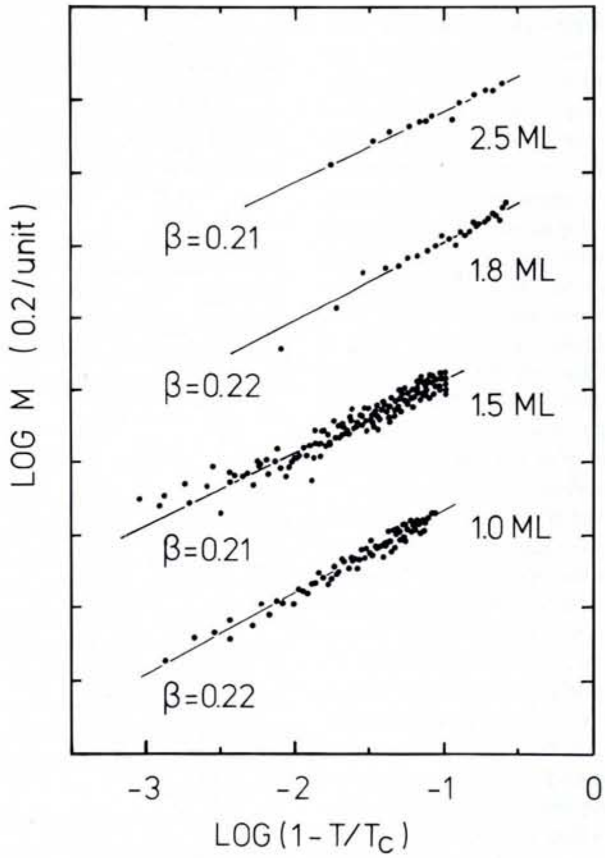

Fig. 5 — Temperature dependence of the magnetization, determined using spin polarized electron spectroscopies, in Fe films of various thicknesses grown layer-by-layer on top of a Au(100) single crystal. M(T) is plotted as log M versus log ($1-T/T_c$) to emphasise the power law dependence for all thicknesses. In contrast to β which remains constant, the 2-d Curie temperature T_2 depends strongly on the thickness (T_2 for a monolayer is about room temperature; T_2 for the 2.5 ML thick film is ≈ 500 K).

It should be noted that the value of the critical exponent derived from the slope of the straight lines is about 0.2, i.e. slightly outside the range expected theoretically. More precise manufacturing of the thin films is probably still required in order to realize the very demanding range of theoretical values (owing to the geometric perfection in the plane of the film being "only" ≈ 100 lattice constants, magnetic fluctuations are limited to this maximum size and the transition is slightly rounded. The determination of β is therefore subject to some uncertainty).

On the theoretical side, the role of the dipole interaction in the critical region is not understood at all so the "true" critical exponent may well fall outside the range 0.10–0.16 range.

This behaviour is precisely that which was found experimentally for the order parameter M in a recent systematic study of the phase transition in thin Fe films on Au(100). Fig. 5 shows the temperature dependence of the magnetization in the vicinity of the transition temperature, T_c, for films of various thicknesses. A log-log scale is used where power laws would be represented by straight lines whose slope is the critical exponent β. From the Figure, the power law nature of the vanishing of M and the independence of β on the film thickness are evident. Thin Fe films on Au(100) therefore represent a physical realization of a 2-d form of universality.

Conclusions

Research on the magnetic properties of layered structures has produced a wealth of exciting new results and this paper by no means offers an exhaustive compilation. For instance, we have largely neglected multilayers of the types illustrated in Fig. 1.

The complexity of multilayers has been highlighted in recent experiments by P. Grünberg, C. Carbone and coworkers at Jülich on Fe-Cr systems which revealed a magnetic coupling between the Fe layers *through* the Cr. As a result of this coupling an entire new class of phenomena arises, not anticipated by the present discussion of single layers.

The essential message however, summarized by Fig. 5, is that truly 2-d magnetic systems are now within the reach of modern experimental physics.

FURTHER READING

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Germar R. *et al.*, *Appl. Phys. A* **47** (1988) 393.
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Ground state magnetic properties:
Freeman A.J. et al., in *Polarized Electrons in Surface Physics*, Ed. R. Feder (World Scientific, Singapore) 1985.

2-d magnetism at finite temperatures:
Pokrovskii V.L. *et al.*, in *Spin Waves and Magnetic Excitations*, Modern Problems in Condensed Matter Sciences, Vol. **22.2**, Eds. A.S. Borovik-Romanov, S.K. Sinha (North-Holland, Amsterdam) 1988.

Course Restructuring

An interim report of tertiary level physics education in the UK recommends:
— remedial teaching of mathematics in universities and polytechnics;
— a 30% reduction in content of the three year undergraduate course;
— an additional year which could either be a fourth year to a three year core course, or a more intensive one year course that may or may not form part of a Masters of Physics preceding the doctorate programme.

The report was prompted by the changeover to a shallow, broader school syllabus, and a concern that UK physics degrees compare poorly with continental counterparts at a time when mobility at a European level is being encouraged. It was prepared by a joint working party, chaired by Professor Chambers of the Physics Department, University of Bristol, made up of representatives from the Institute of Physics, the Standing Committee of University Professors and the Committee of the Heads of Polytechnics. The report was released in April to British universities to allow them to submit appropriate proposals for the next round of government funding.

Financial aspects will dominate further discussions on how to implement the recommendations. Meanwhile, the working party will consider the 'reduced content' syllabus in time to make a final report in September 1990.

Another report dealing partly with physics education is due to be released next month. This time it is the Action Committee on Science and Technology that is examining scientific manpower needs in the UK. Meanwhile, the EPS's very own Action Committee for Physics Education is preparing a major survey at the European level. It will mirror in many vital respects the British considerations.

Paul Scherrer's 100th Anniversary

More than 400 former students of Professor Paul Scherrer met on 3 February 1990 at the ETH, Zürich to celebrate the 100th anniversary of his birth. Among them were about 90 physicists who wrote their Ph.D. theses under his supervision while he was professor of experimental physics at the ETH from 1920 to 1960. They came from all over the world to pay tribute to an excellent teacher and to a man with many outstanding human qualities, enthusiasm, drive and humour. It was very gratifying that his daughter Mrs. Ines Jucker-Scherrer was also with us.

But who was Paul Scherrer and why does he mean so much to Swiss physicists? In a small country like Switzerland, physics could only develop to a high international standard if in the critical era of the twenties when physics was undergoing revolutionary changes, a competent, enthusiastic and dynamic person held the key position of professor of experimental physics at the ETH. Switzerland was indeed fortunate that in 1920 Paul Scherrer, together with Professor Debye, was called to Zürich to succeed Professor Weiss. He built up a successful institute and in the 40 years of his activity many young Ph.D. students obtained a solid training in physics to allow them to find important positions all over the world. Scherrer's initiative in 1927 led to the appointment of Wolfgang Pauli as professor of theoretical physics at the ETH, thus making Zürich one of the leading centres in Europe.

Scherrer was not only a leader of physics in Switzerland but also an "internationalist" who took on the rôle of an international mediator. For example, he organized the first international conferences that took place in Zürich after each World War. Scherrer was also one of the founders of CERN and was instrumental in the choice of Geneva for its site.

Everybody was in a happy and somewhat nostalgic mood and wanted to hear lively, exiting and enthusiastic lectures in the style of Scherrer. This was accomplished by the programme of lectures put together by an organizing committee of former assistants under Kurt Alder.

The main speakers were O. Huber, W.J. Merz, F. Boehm, J.-P. Blaser and H. Albers.

A highlight of the occasion was the talk by Victor Weisskopf who became a personal friend of Scherrer when he was Pauli's assistant in the 1930's. The title — "The great art to make physics understandable" — was appropriate since Scherrer's main aim was to teach students good physics using clear and lively lectures and demonstrations. It was for this reason that Scherrer's lectures were attended by many students, not only of physics but also from other faculties. Movies taken in the 1940's of a number of entertaining skiing, swimming and hiking excursions made by Scherrer's institute completed a memorable and highly enjoyable evening.

W.J. Merz, *Zürich*

Akzo Research Joins

Akzo Research Laboratories Arnhem, the main central research laboratories of Akzo Nederland bv, is to become an Associate Member of EPS. The managerial contact is Ir. E. van Andel, Director of Corporate Research, and the scientific contact is Ir. R.O. van Hasselt, Head of the Department of Applied Physics that is very active in aspects of polymer physics related to fibre technologies.