Magnetism in Europe

I.V. Mitchell

Introduction

The time has long since passed when it seemed that the only uses of permanent magnets were as compass needles to guide lost travelers or as a means to keep refrigerator doors firmly closed. Major developments in materials science over the years have widened the scope of applications to include such devices as small electric motors, loudspeakers, sensors, and actuators. Nevertheless, in most cases, the electromagnet reigned supreme and continued to be used for most of the applications where a magnetic field was required.

However, during the past 20 years a mini-revolution occurred with the development of new alloys based on samarium and cobalt (Sm-Co). The opportunities provided by this new generation of permanent magnets re-ignited the fires of competition and forced equipment manufacturers to take a fresh look at their products.

Unfortunately, at just about the same time that Sm-Co magnets were to be applied, the world supply of cobalt proved to be unreliable. Prices escalated rapidly and magnet users were left in a quandary. Manufacturers were understandably reluctant to commit themselves to incorporating the new high performance magnets into their products because they were wary of shortages and uncontrollable price fluctuations in the raw materials.

In response to this situation, the Commission of the European Communities (CEC) included cobalt substitution for permanent magnets in a program of research aimed at finding suitable materials to replace scarce and strategic raw materials. But, in late 1983, it was to be Sumitomo Special Metals Co. and General Motors Corp. which announced the breakthrough discovery of a remarkable new alloy based on neodymium, iron, and boron (Nd-Fe-B) that exhibited astonishing magnetic properties and set new standards for permanent magnet performance.

This opened up the prospect of cheap, high performance magnets which could find many kinds of applications—in various types of motors and generators, actuators, sensors, scanners, synchrotrons, and even magnetically levitated transportation systems.

Fully alert to the discovery of these "supermagnets," the CEC organized the first European workshop on "Nd-Fe Permanent Magnets: Their Present and Future Applications" in Brussels on October 25, 1984 and, as a direct consequence of the meeting, a plan of action was drawn up with a strong recommendation for a concerted European response in this field. At the same time, the Commission encouraged a small group of researchers to band together to prepare a detailed scientific proposal for a specific program of research. Their program proposal was submitted to the European Commission where it received the unanimous approval of its advisory committee for science and technology (CODEST).

In a little less than a year from the date of the workshop, Community funding was forthcoming from the program for a major collaborative research project on permanent magnets, and thus the Concerted European Action on Magnets (CEAM) was born. The scientific management of the project was placed in charge of the European Commission's research program on Advanced Materials (EURAM), which has particular expertise in high performance permanent magnets.

Purpose of CEAM

CEAM was a unique experiment in interdisciplinary collaborative research and development that spanned a three-year period (late 1985-late 1988). A veritable research network was created, based on a comprehensive study of neodymium-iron-boron (Nd-Fe-B) magnets.

CEAM linked 58 institutes throughout Europe, including most of the laboratories in the European Community working on this specialized subject. Industrial companies made up about one-third of the group with the others coming from universities and national laboratories. The project received funding of 2.5 million ECU (approximately $3 million).

Participation in CEAM was open to laboratories which had an active interest in the field and which wished to join the project when it was first established.

The primary objectives were:

- To develop high performance iron-based rare-earth permanent magnets and to design novel devices which exploit their exceptional properties;
- To generate European collaboration by the exchange of scientists and stimulate a new generation of researchers to undertake projects in applied magnetism of industrial relevance; and
- To provide a skills and information base to permit European industry to exploit the advanced magnets effectively.

Organization of CEAM

The research program was divided into the three broad areas—materials, magnet processing, and applications. The materials section was composed largely of physicists and chemists working on phase diagrams, searching for new alloys, and examining the intrinsic and extrinsic magnetic properties of rare earth alloys, with particular reference to those with the $Nd_2Fe_{14}B$ structure.

The magnet processing group primarily involved metallurgists and materials scientists and included significant industrial participation. This group was primarily concerned with the microstructure of magnet alloys and the numerous problems of magnet processing and stability.

The applications group focused on both electromagnetic and magnetostatic applications of the new magnets. Many of the participants in this group were electrical engineers and specialists in computer-aided design (CAD) working in industrial companies and universities. In all, more than 120 scientists from 58 laboratories directly participated in the project. Nine of the 12 EC Member States were represented, as well as Austria.

Organizationally, CEAM operated in four laboratories: the CNRS at Grenoble and Trinity College, Dublin for the materials group; Birmingham University for the magnet processing section; and the Technical University of Berlin for the applications group. All other participants acted as subcontractors of one or more of these coordinating institutes.

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Materials

The primary goals of the materials research group established within CEAM were:

- To achieve a fuller understanding of the magnetic properties of $Nd_2Fe_{14}B$ and related phases;
- To improve magnetic properties by controlling the microstructure and chemical composition; and
- To search for new rare-earth iron-rich phases likely to have potential as permanent magnets.

All these areas have produced noteworthy scientific achievements that are providing a valuable input to the industrial developments taking place in CEAM and elsewhere.

New Phases

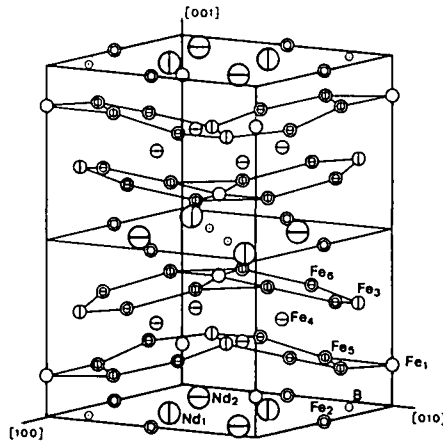
A coordinated search for novel rare-earth iron compounds was carried out by more than a quarter of the CEAM participants. One approach, which proved fruitful, was to overquench a ternary composition by melt-spinning and then crystallize it from the amorphous state. Another was to produce a polycrystalline sample from the melt, and then subject it to thermomagnetic and microprobe analysis. Hundreds of compositions were prepared, and many new stable and metastable phases were discovered.

One particularly promising material was obtained with melt spun ribbons of $R_4Fe_{77}B_{19}$, which initially crystallize to an Fe_3B phase that contains a little of the rare-earth element. With further development, this material could be used to manufacture inexpensive bonded magnets with a modest energy product but good corrosion resistance.

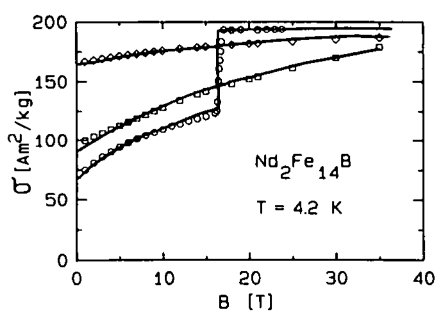
Another useful family likely to find a niche for certain applications are the pseudo-binaries $Sm(Fe_{12-x}M_x)$, where M = Ti, V, Si, etc., which crystallize in the tetragonal $ThMn_{12}$ structure. The best of them show Curie temperatures and anisotropy similar to those of $Nd_2Fe_{14}B$, but with somewhat lower magnetization.

Crystal Growth and Structure

A prerequisite for a proper understand ing of the intrinsic magnetic properties of the $Nd_2Fe_{14}B$ family of compounds is the availability of high quality single-crystal samples. Crystals of $R_2Fe_{14}B$ and $R_2Co_{14}B$ were successfully grown in controlled atmospheres for different rare-earth elements (R) in Grenoble and Amsterdam using a sophisticated Czochralski technique. These were made available to other laboratories for fundamental magnetic investigations.



Crystal model shows the tetragonal unit cell of $Nd_2Fe_{14}B$.



Magnetization curves of a $Nd_2Fe_{14}B$ crystal along the three principal crystallographic directions.

The first determination of the complex structure of $Nd_2Fe_{14}B$ by x-ray diffraction on a single crystal was a milestone and confirmed an earlier powder diffraction determination by a U.S. group. This layered structure has two inequivalent Nd sites, six iron sites, and one boron site. Sheets of Nd and B are sandwiched between double close-packed iron layers. It has now been clearly established that the low symmetry of the neodymium sites is the origin of the strong magnetic anisotropy, which leads to hysteresis and useful hard magnetic properties. Although single crystals do not exhibit hysteresis, they do permit accurate measurement of the anisotropy.

Phase Relations and Microstructure

In the real world, magnets have a microstructure composed of several phases. To optimize the microstructure, the various phases that coexist in a state of equilibrium must be clearly identified as a function of the composition and temperature. Ideally, the nonmagnetic phases should be avoided, controlled or reduced as far as possible to maintain the quality of the magnet. Careful constitutional investigations of the Nd-Fe-B system by varying the $Nd_2Fe_{14}B$ composition, revealed a small region with two-phase magnets. These magnets are composed of grains of $Nd_2Fe_{14}B$ (Φ phase) surrounded by a thin, nonmagnetic Nd-rich layer. The technical spin-off is that these magnets exhibit superior hard magnetic properties compared with conventional three-phase magnets, which contain a further nonmagnetic component, $Nd_{1+\epsilon}Fe_4B_4$ (η phase).

Magnetic Properties

A precise description of the magnetic anisotropy and the coupling between magnetic moments of the rare-earth and iron sublattices is important if advances are to be made in understanding hard magnetic properties. Such a description is now beginning to emerge after systematic and painstaking examination of the $R_2Fe_{14}B$ series using techniques such as nuclear magnetic resonance (NMR), Mössbauer spectroscopy, neutron diffraction, and high-field magnetization measurements.

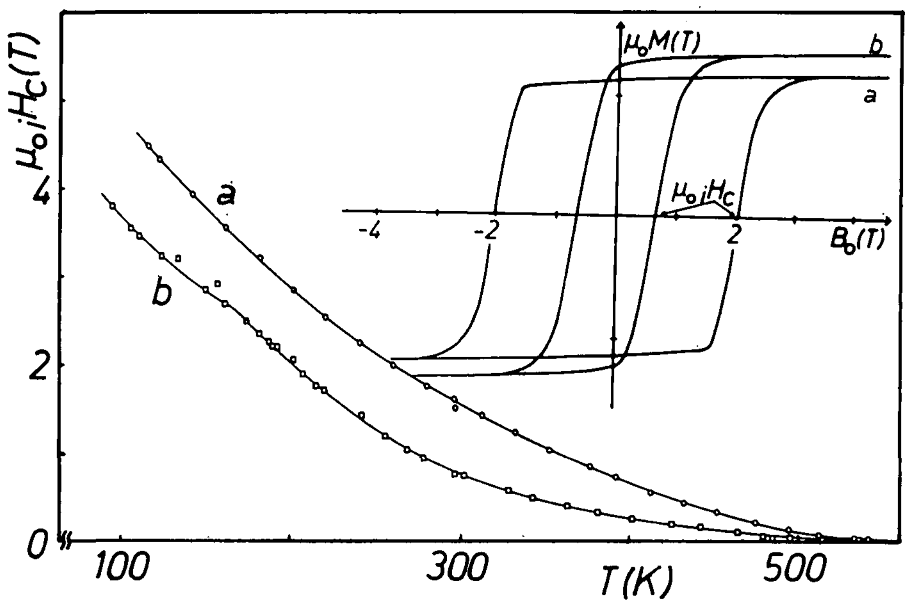
The magnetic anisotropy due to the rare earth is usually described in terms of a crystal-field model. A reliable set of crystal-field parameters has now been deduced for the whole $R_2Fe_{14}B$ series as a result of collaborative work. In particular, the complex magnetic structures in $Nd_2Fe_{14}B$ and $Ho_2Fe_{14}B$ at low temperatures, the temperature dependence of the magnetic anisotropy, and the transitions in high magnetic fields are now well understood.

It was possible to exploit the unique high magnetic field facilities in laboratories in Grenoble and Amsterdam for studies on single crystals, permitting the evaluation of the magnetic coupling and crystal-field parameters. Through such means, consistent sets of coupling parameters for different rare-earth-iron and rare-earth-cobalt families of alloys were derived.

Atomic-Scale Magnetism

An essential ingredient for understanding magnetic phenomena is a detailed knowledge of the magnetism at the level of individual atoms. Magnetic moments and electric field gradients are probed on an atomic scale via hyperfine interactions, measured by NMR or

Mössbauer spectroscopy. Site preferences of elements substituted for iron have been determined, and magnetic order has been successfully inferred on an atomic scale. From hyperfine measurements and experiments involving the diffraction of polarized neutrons, the magnitude of the iron moments on different lattice sites are found to range from 1.9 to 2.5 Bohr magnetons (μ_B). The electric field gradients at the two rare-earth sites in $Gd_2Fe_{14}B$ provide a direct measurement of the second-order crystal-field parameters.



Temperature dependence of coercivity of Nd-Fe-B magnets (a) with and (b) without Dy additions. Inset: The corresponding hysteresis loops at 296 K. (Photo: Kirchmayr—Vienna University.)

Coercivity

CEAM proved to be an excellent forum for constructive discussion of the controversial subject of coercivity mechanisms. A permanent magnet is distinguished from other types of magnetic material by its hysteresis loop and coercivity. It is this which enables it to remain magnetized in the presence of an opposing magnetic field. Two conditions must be fulfilled for appreciable coercivity to exist—the material should possess a strong intrinsic anisotropy, and, at the same time, have an appropriate microstructure.

Nd-Fe-B magnets prepared by sintering powders are composed of individual magnetic grains (typically 5μm) disseminated in a nonmagnetic matrix. When the alloy is subjected to an external field opposed to the direction of magnetization, a domain of reversed magnetization will spontaneously nucleate and grow exponentially as the field exceeds a critical value. In contrast, Nd-Fe-B magnets prepared from ribbons obtained by quenching the melt have a different microstructure composed of tiny magnetic grains (around 20 nm) surrounded by a very thin (~2 nm) intergranular phase. In this case, a reverse domain may nucleate, but its surface—the domain wall—has a tendency to be pinned at magnetic inhomogeneities such as the grain boundaries. It can grow only when the field is large enough to overcome the pinning energy. Hysteresis loops and associated microstructure establish the relative importance of nucleation and pinning mechanisms in the sintered and rapidly quenched magnets.

Chemical Substitutions

Nd-Fe-B magnets with approximate composition $Nd_{15}B_{77}B_8$ are ideal for room temperature applications, but their coercivity falls off rapidly at about 100°C, well below the Curie temperature (T_c = 310°C). This makes them unsuitable for use in electrical machines where temperatures can rise to 150 or 200°C. A comprehensive study of the effects of chemical substitution in $Nd_2Fe_{14}B$ has been undertaken to find a way to optimize the material for different applications. Cobalt is effective at raising the Curie temperature but does not normally improve coercivity. Anisotropy is best enhanced by replacing part of the Nd by Dy or Tb. Additions of small amounts of Al, Nb, Ge and Mo have been shown to be effective in improving the coercivity at elevated temperatures. The effect of many of these substitutions on intrinsic magnetic properties and microstructure has been clarified during the course of the program. A range of compositions optimized for various applications is now at hand.

Magnet Processing

Through the active collaborations set in motion by CEAM, considerable experience was gained in the processing and manufacture of Nd-Fe-B magnets. The magnets now produced are comparable in quality to any produced elsewhere in the world. Much of the work of the magnet processing group has concentrated on certain critical aspects of the processing and production of magnets from the basic alloy, $Nd_{16}Fe_{76}B_8$. In the light of the results from the materials group, other compositions, which give improved coercivities, are being studied.

Achievements can be divided broadly into the following areas:

- Characterization of the microstructure of the cast ingots;
- Development of the hydrogen decrepitation process as an economical powder production route;
- Measurement of the physical properties and characterization of microstructure of sintered magnets;
- Study of the corrosion behavior of sintered magnets and development of corrosion protection strategies; and
- Production, characterization and magnet fabrication of melt-spun material.

Industrial application is already under way in a number of these areas.

Primary Materials

The starting material for magnet manufacture is the Nd-Fe-B alloy in the form of cast ingots. The state of the ingot is found to be related to the final magnetic properties of sintered magnets made from it. Standard cast ingots fabricated by two primary alloy producers have been circulated within the magnet processing group for thorough characterization and assessment. Controlled solidification experiments have been carried out on the cast ingot material in order to study phase equilibria and preferred growth directions.

A milestone has been the establishment of optimum casting conditions needed to minimize the free-iron content, which has a deleterious effect on

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the milling characteristics. Cast ingots also include large regions of Nd-rich material and these are shown, by means of optical metallography and scanning electron microscopy, to have a complex microstructure. Much effort has been put into unraveling this structure since it is now thought to be the key to understanding both the coercivity and corrosion properties of sintered magnets.

Powder Metallurgy Sintered Magnets

One processing route for Nd-Fe-B magnets involves sintering finely milled and oriented metal powder. A significant achievement in CEAM is the successful transfer of the hydrogen decrepitation process (HD) from the laboratory to industrial scale for fabricating sintered Nd-Fe-B magnets. The cast ingot can be readily broken up into fine material by exposure to hydrogen at a moderate pressure (~1 bar) at room temperature. The hydrided powder, which contains about 0.4 wt.% hydrogen, is milled, aligned, and pressed to form a green compact. The hydrogen is removed completely during subsequent vacuum sintering. The advantages are that the milling times are considerably reduced compared with the standard procedures and there is a very significant reduction in sintering temperature. Finer grain sizes and hence higher coercivities are also achieved using the HD-process, now established as a viable commercial route for manufacturing sintered Nd-Fe-B magnets. This technology is now being pursued by magnet manufacturers in the European Community and elsewhere. In addition, hydrogen absorption/desorption spectroscopy has been successfully used to characterize both ingot material and sintered magnets.

Rapid Quench Processing

A simpler processing route involves direct casting of the melt onto a rapidly rotating copper wheel. This method tends to produce randomly oriented crystallites, and the magnets exhibit inferior properties to those produced by powder metallurgy. Melt-spun ribbon produced under a variety of conditions has been characterized magnetically and microstructurally by electron microscopy. The very fine grain sizes (~20 nm) are correlated with wheel speed and subsequent annealing treatments. Promising results are obtained when small amounts of other elements such as Si and Nb are added to the basic alloy; there are significant changes in the grain morphology and substantial enhancements in the magnetic properties. Melt-spun ribbon has been used to make magnets by a variety of means, including compression molding, hot pressing, and die-upset forging. The ribbon material is also being used successfully to produce Nd-Fe-B-based injection-molded magnets. Such magnets have found important industrial applications.

Corrosion Protection

An impediment to the wider adoption of Nd-Fe-B based magnets is their susceptibility to certain forms of corrosion. This is caused in part by the excessive reactivity of regions of Nd-rich material in the magnet microstructure. They are especially prone to corrosion in humid atmospheres, where, under similar conditions, they show approximately four times the activity of mild steel. Common corrosive products are oxides and hydrogen. To better understand the processes involved, gravimetric measurements have been employed to characterize the oxidation behavior of the milled powder, and some of the oxidation products have now been successfully identified. Oxidation properties are correlated with the particle size distribution of the powders. Various coating procedures to protect finished magnets from corrosive environments have been developed. It has been found that pretreatment prior to coating is essential for good adherence of the protective layer.

Physical Properties and Data

Absolute confidence in the magnetic measurements carried out in the various laboratories was obviously vital to the inter-comparative exercises in the program. Standard magnets were prepared and circulated among the participants and the magnetic measurements collated and compared. In general, very satisfactory agreement was obtained among the various laboratories. Furthermore, these measurements are now being correlated with the microstructure of the magnets. To place the work in context, many measurements were also carried out on commercial Nd-Fe-B magnets from suppliers throughout Europe, the United States, and Japan.

Applications

Research and development in this group centered on applying Nd-Fe-B magnets in rotating machines and static devices. The technical achievements can be divided broadly into:

- New and improved design and engineering capabilities;
- Design and construction of a range of electrical machines; and
- Design and construction of prototypes of static devices such as hexapoles, wigglers, undulators, clamping devices, and magnetic resonance image scanners.

There is a general consensus that Nd-Fe-B will become the material of choice for many new permanent magnet machines.

Design Capabilities

Members of the applications group have successfully evolved and demonstrated new and innovative design capabilities. Good design is the key to efficient use of Nd-Fe-B, exploiting the excellent magnetic properties while simultaneously minimizing the corrosion susceptibility and compensating for the temperature dependence of the properties of the present generation of magnets.

Sophisticated computer-aided-design (CAD) procedures have been developed to calculate complex magnetic fields, based on both analytical techniques and finite-element numerical methods. A wide range of electrical machines and static devices have been designed which exploit the superior properties of Nd-Fe-B magnets. Details of improved magnet specifications and properties coming from CEAM and elsewhere are taken into account to refine the characteristics of the prototypes.

Engineering Capabilities

The high magnetization of Nd-Fe-B and its comparatively low cost make permanent magnet excitation particularly attractive in large electrical machines. However, handling and assembly of even small magnetized parts is difficult and large magnetized components present an added safety hazard as well as severe risk of damage during handling. For these reasons, assembly strategies and fixing procedures are being developed and experience accumulated through the construction of prototypes.

The preferred strategy is to magnetize the material after assembling the device. Investigations into post-assembly methods of magnetization are producing encouraging results. The high field needed to magnetize Nd-Fe-B makes it difficult to produce the complex patterns of magnetization required for some types of small motors. Nevertheless, examples of miniature brushless dc and stepper motors have been constructed which demonstrate the benefit of using the new material. Alternative magnetization methods are being actively pursued. The sensitivity of the material to high temperatures calls for increased attention to the thermal aspects of a design.

Small Machines

In computer peripherals, such as printers and storage devices, the trend is to small packages, low power consumption, high efficiency, and low cost. Consequently, drives need to have small rotor inertia, small motor mass, and small volume. Moreover, there is an increasing demand for low-noise electric motors. A natural solution is to use small Nd-Fe-B magnets with slotless stators and either cup-type or disk-type air gap windings. The rotor inertia can be appreciably reduced, which leads to the desired mass and volume reductions. Due to the good demagnetization of Nd-Fe-B magnets, the flux density in the air gap remains high, resulting in a low noise motor with high torque.

However, the high air gap flux density can cause saturation effects, and therefore a finite element approach is needed to establish optimum design criteria. An alternative approach, currently under investigation by members of the group, is to design a motor where the Nd-Fe-B magnets are buried in the rotor. This novel configuration results in an air gap field higher than in the magnet itself and virtually eliminates demagnetization effects. This type of motor is ideal for applications requiring high output per unit volume. The desirability of direct-drive motors is clear because gearboxes and pulleys can be replaced thanks to the increased torque generated by the new type of permanent magnet machines.

Industrial Machines

Current projects include investigating the potential for Nd-Fe-B magnets in various categories of permanent magnet excited machines for industrial applications ranging from axis- and spindle-drives for machine tools to the electrodynamic braking of high-speed railway vehicles. Although electronically commutated brushless dc motors, which are likely to be a significant growth technology for European industry, feature strongly in the group's activities, detailed design studies are also being made on brushed dc motors, line-start synchronous motors, stepper motors, and permanent magnet generators. Nd-Fe-B is being considered for radial- and axial-field designs of both slotted and slotless configurations.

Prototype machines have been constructed to validate, among other things, thermal characteristics under both peak and continuous performance. This is particularly important for currently available grades of Nd-Fe-B. Among the advantages found for machines equipped with Nd-Fe-B are improved performance factors such as higher torque per frame size, improved efficiency, and better dynamic response. In some cases, motors are being evaluated in drive systems using pulse width modulation supply modules and incorporating torque, velocity, and position control.

Static Devices

Permanent magnets play an important role in static applications. Common examples included clamping or holding devices for ferrous materials, loudspeaker and headphone elements, focusing and steering of charged particles, and medical diagnosis equipment such as the magnetic resonance imaging (MRI) scanner. In CEAM, efficient permanent magnet devices are being designed which take advantage of the superior properties of Nd-Fe-B.

A new range of workholding tools has been developed. The magnet cost per unit of attractive force for Nd-Fe-B is much better than for earlier designs based on traditional magnetic materials.

A new hexapole device using Nd-Fe-B magnets is under construction for use in an electric cyclotron resonance ion source. Here, ions are produced in a plasma which is contained in a "magnetic bottle." Traditionally, the magnetic bottle has been created by a hexapolar field of permanent magnets and an axial field of electric coils. Using Nd-Fe-B magnets, stronger hexapole fields can be obtained—and hence denser plasma contained—with the added advantage that the electrical coils and power supplies can be dispensed with. This allows construction of a more powerful ion source which can be easily mounted on the high voltage platforms used in nuclear physics facilities.

Another noteworthy achievement is the design of a permanent magnet configuration for whole-body MRI using Nd-Fe-B material. This requires approximately 5 tons of magnet in place of the 100 tons needed if ferrite magnets are used. The device is designed to produce in its center volume a very homogeneous magnetic field (uniform to 1 part in 10,000) with a strength of 0.2 tesla. This permanent-magnet-based MRI dispenses with a helium refrigeration infrastructure and therefore offers an attractive alternative to the more costly superconducting magnet systems presently used by hospitals to make proton resonance images for medical diagnostic examinations.

Conclusion

Apart from its technical achievements, which during a three-year time span led to more than 460 scientific publications and eight patent applications, CEAM has been of great service to the R&D Community. A new generation of young technicians and researchers are being trained in the field of applied magnetism, which is helping to revitalize a rather small and fragmented industry that is critical to the economic and strategic well-being of Europe. CEAM is also helping to create a "snowball" effect, allowing new results to move rapidly into industrial development and manufacture. And as far as advanced magnetic technology is concerned, CEAM is providing a European dimension to the subject, which is indispensable in the face of fierce worldwide competition.

CEAM's underlying principle of free exchange of information and expertise across a wide range of disciplines is encouraging genuine pan-European cooperation. Once established, these links are not easily broken and lead to new collaborations—as evidenced by the number of joint project proposals received in recent EC R&D programs. Its successes and the way skills and expertise across so many frontiers, political as well as disciplinary, were brought—and kept—together, is a fine example of what can be achieved in Europe. CEAM will serve in the future as a benchmark to judge other projects.

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