MAGNETORESISTANCE OF METALLIC MULTILAYERS

Breakthroughs Being Applied

R. Coehoorn of Philips Research, Eindhoven, describes the background to a major new advance in field sensing materials for magnetic heads.

The expected advantages of applying novel multilayers which display a new effect called "giant magnetoresistance" (GMR) in magnetic read heads are a record high level of sensitivity for fields typically of less than 10^{-4} T, and a highly linear voltage *versus* field characteristic. The latter is very important for both analogue as well as digital recording, and its realization would lead to a significant simplification in magnetic head design. This short report summarises recent progress in exploiting the breakthroughs to develop magnetoresistive multilayer materials for magnetic head and other sensor applications.

Research on metallic multilayers with nanometre-scale layer thicknesses led to the discovery of the very large magnetoresistance effect in 1989. The first observations were made for systems with alternating iron and chromium layers (Fe/Cr multilayers) [1] and with alternating cobalt and copper layers (Co/Cu multilayers) [2]. Relative changes of the resistance of 100% at 4.2 K and 65% at room temperature have been found in fields on the order of 1 tesla.

The new effect was called giant magnetoresistance, referring to the extraordinarily large resistance changes for systems such as Fe/Cr and Co/Cu, or, alternatively, "spin valve magnetoresistance", referring to its microscopic origin. It is physically different to both the classical magnetoresistance effect and the anisotropic magnetoresistance (AMR) effect. The former is found for all metals and originates from curved electron paths in a magnetic field. It shows a quadratic field dependence, and leads for metals to relative resistance changes smaller than 1% in fields on the order of 1 T. AMR arises in magnetic alloys when the resistance depends upon the angle between the current and the magnetization directions. It is of relativistic origin and amounts to at most 3% for thin-film specimens at room temperature. The AMR effect is presently exploited in ultrasensitive thin-film field magnetic field sensors for magnetic read heads in recording equipment and for electronic compasses. The GMR effect has recently been shown to hold a high potential for application in low-field applications of the same types.

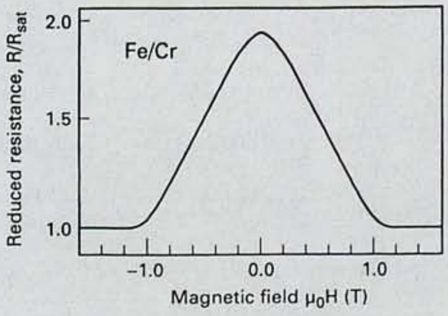


Fig. 1 — Magnetoresistance at 5 K of a 40 x (3.0 nm Fe/1.2 nm Cu) multilayer grown by MBE [4]. The magnetoresistance ratio defined as $R(H{=}0) - R_{sat}$, is 95%, where R_{sat} is the saturation value of the resistance at high fields and $R(H{=}0)$ is the resistance at zero field.

Antiferromagnetic Interlayer Coupling

The GMR effect is found in metallic multilayers which consist of alternating ferromagnetic (F) and non-ferromagnetic layers, where the angle between the magnetization directions of neighbouring F layers can be changed in a magnetic field. Antiferromagnetic interlayer exchange coupling is used in the case of Fe/Cr and Co/Cu multilayers [3]. For specific Cr or Cu thickness intervals, the Fe and Co layers, respectively, are alternately magnetized up and down. High fields, up to several teslas in some cases, are required to obtain a parallel alignment of the magnetization direction in all layers; the change of the relative magnetization direction is accompanied by a change in the resistance.

Fig. 1 shows the magnetoresistance curve for a multilayer consisting of 40 repetitions of 3.0 nm thick Fe layers and 1.2 nm thick Cr layers, grown by molecular beam epitaxy on a Ge(001) single crystal substrate in an ultrahigh vacuum chamber [4]. Fe and Cr have a body-centered cubic crystal structure, with a lattice mismatch of only 0.6%; Fe is lattice matched with the substrate within 1.3%. These are favourable conditions for the formation of a coherent superlattice, which has indeed been realised.

The occurrence of antiferromagnetic interlayer exchange coupling is an indication of relatively smooth interfaces, and of the absence of ferromagnetic bridges ("pinholes") across the very thin non-magnetic layers. Many multilayer systems have been found to display *oscillatory* interlayer exchange coupling together with variations in the magnetoresistance; multiple, superimposed, oscillations have been found in some systems. The periods of the oscillations depend on the superlattice growth direction and crystal structure, and can be explained in terms of the Fermi surface dimensions and topology of the non-ferromagnetic metal [5].

Fig. 2a gives the measured thickness dependence of the interlayer exchange coupling for a Co/Cu/Co (100) oriented sandwich system [6] grown by molecular beam epitaxy, and Fig. 2b shows the accompanying variation of the magnetoresistance in sputtered (100) oriented multilayers [7]. The rapid oscillations in the coupling strength above 2 nm Cu thickness are probably not resolved in the magnetoresistance owing to small variations in thickness and to the presence of some interface roughness.

Two-Current Model

Crucial ingredients of the theory of GMR are i) spin-dependent electron scattering in the bulk of the magnetic layers and/or at the interfaces, and ii) an electron mean free path for at least one spin direction which is larger than, or of the same order of magnitude as, the multilayer repetition period. Fig. 3 shows

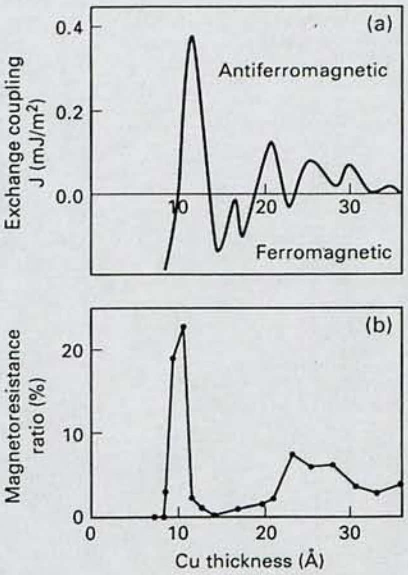


Fig. 2 – Oscillatory interlayer exchange coupling. a, upper) Dependence on the Cu layer thickness of the interlayer exchange coupling between 6 nm Co layers deposited by MBE on a Cu(100) single crystal [6]. The coupling constant J is proportional to the energy which is required to change the magnetization directions from parallel to antiparallel. The exchange coupling can be viewed as a superimposition of two oscillations, with periods 1.4 and 0.46 nm.
b) Dependence on the Cu layer thickness of the magnetoresistance at 5 K of (100) oriented 25 x (1.6 nm Co/Cu) multilayers. Deposition by DC magnetron sputtering on a 20 nm Cu underlayer. Comparison with a) shows that the magnetoresistance is only observed for antiferromagnetically coupled systems.

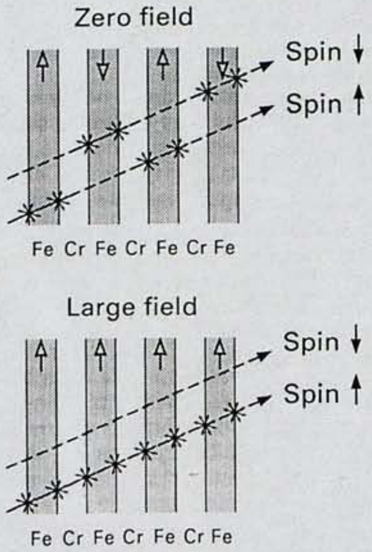


Fig. 3 – Two-current model: schematic illustration of electron scattering at the interfaces in Fe/Cr multilayers with antiferromagnetic interlayer exchange coupling. Dashed lines are electron trajectories and fanned arrows indicate relatively large diffusive scattering probabilities at the interfaces; arrows in the Fe layers indicate the magnetization directions. The probability for diffusive scattering at an interface with magnetization "up" is much larger for spin-up than for spin-down electrons, and vice versa if the magnetization direction of the Fe layer is "down".

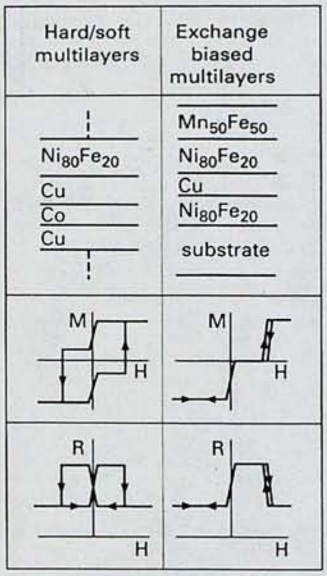
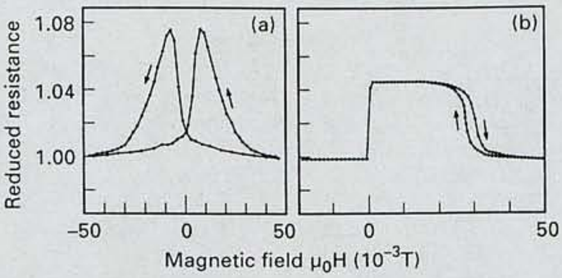


Fig. 4 — Magnetoresistive multilayers with high sensitivity: a, upper) illustrations of hard/soft (left) and exchange-biased (right) systems which have been proposed, together with schematic representations of the magnetization M(H) and resistance R(H) responses in a magnetic field H.
b, lower) The corresponding experimentally observed room-temperature magnetoresistance: 30 x (2 nm Co/5 nm Cu/3 nm $Ni_{80}Fe_{20}$/5 nm Cu) hard/soft multilayer (left) and an exchange-biased (8 nm/2.5 nm Cu/6 nm $Ni_{80}Fe_{20}$/8 nm MnFe) multilayer (right).



schematically the effect of spin-dependent interface scattering on the spin-up and spin-down electron currents in Fe/Cr multilayers. The probability for diffuse scattering at an interface between Cr and an Fe layer with magnetization "up" is much larger for spin-up than for spin-down electrons. The reverse is true if the magnetization direction of the Fe layer is "down".

The application of the two-current model, with non-interacting parallel spin-up and spin-down currents, leads to the following resistivities for the high-field (parallel – P) and low-field (antiparallel – AP) cases:

$$\rho_P = (1/\rho_\uparrow + 1/\rho_\uparrow)^{-1} \text{ and } \rho_{AP} = (\rho_\uparrow + 1/\rho_\downarrow)/4$$

Here $\rho_\uparrow$ and $\rho_\downarrow$ are the resistivities in the two separate spin channels in the parallel case. For Fe/Cr, $\rho_\downarrow << \rho_\uparrow$ where we have assumed that for both spin directions, the mean free path is much larger than the repetition period; it is easily seen that $\rho_{AP} \geq \rho_P$.

Iron and chromium form a nearly ideal pair of metals because their electronic structures are almost identical for minority spin electrons, whereas their majority spin electronic structures are highly dissimilar. Recent electronic structure calculations have shown that this is not only true for bulk crystals, or at ideal interfaces, but also at more realistic non-ideal chemically mixed interfaces. A similar situation holds for majority spin electrons In Co/Cu multilayers. Both systems therefore exhibit the required large difference between the scattering probabilities of spin-up and spin-down electrons. Additional advantages of Fe/Cr and Co/Cu systems are that both metals are almost perfectly lattice matched, leading to coherent superlattices with small probabilities of spin-independent scattering at lattice imperfections.

One of the issues currently being debated is whether or not the spin-dependent scattering occurs predominantly in the bulk of the layers, or at the interfaces. Various experiments have shown that even atomically thick additional layers at the interfaces may drastically alter the GMR [8]. It is of interest to note that diffusive (as opposed to reflective) scattering at the interfaces, arising from imperfections at the interfaces, may increase the GMR if the scattering is strongly spin-selective. Indeed, there are observations of an optimum degree of interface roughness for Fe/Cr multilayers [9]; more perfect or more diffuse interfaces being less ideal.

Sophisticated Models

More sophisticated theoretical models for the GMR have been developed, and make use of the Boltzmann transport equation or the Landauer-Buttiker formalism [10]. Several approaches have been proposed to include quantum effects [11]. One of the interesting predictions is that the GMR of "current perpendicular to plane" (CPP) systems should be much larger than that of the commonly used "current in plane" (CIP) systems. This was, in fact, recently observed by several groups [12] (for example, by comparing the CPP magnetoresistance for Fe/Cr pillar-shaped microstructures with the CIP magnetoresistance). The larger temperature dependence of the magnetoresistance in the CPP geometry has been attributed to a more pronounced influence of spin-flip scattering at high temperature owing to wavelike fluctuations of the magnetization direction (magnons), which lead to mixing of the two spin currents. Studies of the CPP magnetoresistance are expected to help clarify the role of electron-magnon scattering, which together with electron-phonon scattering leads to a decrease with temperature of the magnetoresistance.

Low-Field Systems

In spite of the high magnetoresistance ratio for Fe/Cr and Co/Cu systems, their potential for applications in magnetic sensors is limited owing to the very high switching fields. The Earth's magnetic field corresponds to a magnetic induction of about 0.5 x 10^{-4} T, which is of the same order of magnitude as stray fields from magnetic tapes or disks. Fig. 4a shows schematically two methods which have been proposed to reach high sensitivity at low fields. In "hard/soft" multilayers [13] alternating soft magnetic permalloy ($Ni_{80}Fe_{20}$) layers and relatively hard magnetic Co layers are separated by Cu layers (the Cu layers are relatively thick (5 nm) in order to reduce the interlayer exchange coupling). It is very fortunate that permalloy, well known for its high magnetic permeability and low magnetostriction, is well matched to Co and Cu, both electronically and structurally. Fig. 4b demonstrates that switching of the permalloy layer occurs in a small field interval $\mu_0\Delta H \approx 10^{-2}$ T, resulting in a 8% resistance change. At higher fields, the Co layers also switch, and the resistance decreases.

Permalloy/Cu/permalloy/MnFe layered systems have also been proposed for sensor applications [14]. As shown in Fig. 4b, relative resistance changes of 4-5% can be obtained in field (magnetic induction) intervals smaller than 10^{-3} T. Fig. 4a illustrates that the first permalloy layer switches at round zero field, whereas the second layer switches only after the application of a relatively large positive field, its magnetization loop being shifted due to the exchange interaction with the antiferromagnetic 8 nm thick $Fe_{80}Mn_{20}$ alloy layer. This intriguing effect is called "exchange biasing" and is believed to be caused by the creation of a specific domain structure in the antiferromagnet during growth on the second permalloy layer in a magnetic field.

The search for more sensitive magnetoresistive thin film materials is strongly stimulated by the continuing miniaturization of magnetic recording heads for digital audio and video recording and for data storag Magnetoresistive elements in such heads today measure 10-100 μm, but future [illegible]creases of storage density will probably require 1 μm structures. A European consortium formed by three companies (Philips, Thomson and Siemens), four universities (Paris-Sud, Strasbourg, Eindhoven, and Erlangen) and one institute (KFA Jülich) has started a three-year EC-funded ESPRIT Basic Science project on magnetoresistive multilayers. The aims are to elucidate further the new physics of these structures, as well as to take up the challenge of preparing ultra-sensitive materials for industrial applications.

[1] Baibich M.N., *et al.*, *Phys. Rev. Lett.* **61** (1988) 2472; Binasch G., *et al.*, *Phys. Rev. B* **39** (1989) 4828.

[2] Mosca D.H., *et al.*, *J. Magn. Mag. Mater.* **94** (1991) L1; Parkin S.S.P., Bahdra R. & Roche K.P., *Phys. Rev. Lett.* **66** (1991) 2152.

[3] Grünberg P., *et al.*, *Phys. Rev. Lett.* **57** (1986) 2442; Parkin S.S.P., *ibid.*, **64** (1990) 2304; *67* (1991) 3598.

[4] Folkerts W., Hoving W. & Coerne W., *J. Appl. Phys.* **71** (1992) 362.

[5] Bruno P. & Chappert C., *Phys. Rev. Lett.* *67* (1991) 1602.

[6] Johnson M.T., *et al.*, *Phys. Rev. Lett.* **68** (1992) 2688.

[7] Coehorn R., *et al.*, in *Magnetism and Structure in Systems of Reduced Dimension* (Plenum, New York) 1993 (to be published)

[8] Baumgart P., *et al.*, *J. Appl. Phys.* **69** (1991) 4792; Parkin S.S.P., *Appl. Phys.Lett.* **61** (1992) 1358.

[9] Petroff F., *et al.*, *J. Magn. Mag. Mater.* **93** (1991) 95.

[10] Camley R.E. & Barnas J., *Phys. Rev. Lett.* **63** (1989) 664; Bauer G.W., *ibid.*, *69* (1992) 1676.

[11] Levy P.M., Zhang S. & Fert A., *et al.*, *Phys. Rev. B* **43** (1990) 1643.

[12] Pratt W.P., *et al.*, *Phys. Rev. Lett.* **66** (1991) 3060; Gijs M.A.M. & Okada M., *Phys. Rev. B* **46** (1992) 2908.

[13] Shinjo B. & Yamamoto H., *J. Phys. Soc. Japan* **59** (1990) 3061.

[14] Dieny B., *et al.*, *J. Appl. Phys.* **69** (1991) 4774.