Large enhancement of the perpendicular giant magnetoresistance in pseudorandom magnetic multilayers

J. Mathon
Department of Mathematics, City University, London EC1V 0HB, United Kingdom
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It is predicted that the current-perpendicular-to-plane giant magnetoresistance (CPP GMR) in magnetic multilayers with dimensions smaller than the electron mean free path is greatly enhanced by growing layers with thicknesses that follow a pseudorandom number sequence. The CPP GMR, calculated from the Kubo formula, is shown to increase exponentially with the multilayer thickness. This is due to Anderson localization, which is strong in zero field, but becomes weak in one of the spin channels when the layer moments are aligned in a saturating magnetic field. [S0163-1829(96)08326-9]

The giant magnetoresistance (GMR) effect in magnetic multilayers has received much attention.[1] The GMR effect, which is usually studied in the current-in-plane (CIP) geometry, occurs when the magnetizations of neighboring magnetic layers oriented spontaneously antiparallel are aligned parallel in a saturating magnetic field. The resistance decreases dramatically in an applied field and CIP GMR ratios as high as 200% have been reported for Fe/Cr (Ref. 2) superlattices. The dimensions of a magnetic multilayer in the direction of the current flow are usually macroscopic in the CIP geometry, i.e., much greater than the electron mean free path. The CIP GMR effect takes place, therefore, in the diffusive regime and is believed to be due to spin-dependent scattering from magnetic impurities that migrate into nonmagnetic spacer layers near the interfaces. Owing to reduced symmetry, rigorous theoretical treatment of CIP GMR in the diffusive limit is very difficult and a satisfactory microscopic theory of the effect is still lacking.[3]

An important development is the observation[4] of GMR in the current-perpendicular-to-plane (CPP) geometry. Electrons in the CPP geometry cross a large number of ferromagnet/spacer interfaces and are partially reflected at each interface from spin-dependent potential steps whose strengths depend on the offsets of the majority- and minority-spin bands in the magnetic and nonmagnetic layers. The role of multiple scattering from the interfaces is, therefore, much enhanced compared with the CIP geometry. In fact, Asano *et al.*[5] conclude, on the basis of their calculations for a single-orbital tight-binding model, that GMR in the CPP geometry is dominated by such band structure effects and impurity scattering is only of minor importance.

Schep *et al.*[6] have taken a further step and identified a situation accessible to experiment in which impurity scattering is negligible and hence rigorous calculations of GMR are possible. This is the ''clean'' limit in which all the dimensions of a multilayer sample are smaller than the mean free path for scattering from impurities. If one further assumes that the multilayer is periodic, transport is in the ballistic regime and the conductance (GMR) is obtained simply by counting the number of propagating states. Schep *et al.*[6] performed such a calculation for ballistic point contacts made from Co/Cu superlattices. They found using *ab initio* bands that CPP GMR in the absence of impurities can be as high as 120%. The CPP GMR they calculate is due, entirely, to scattering from perfect Co/Cu interfaces.

Although present experiments are in the diffusive limit, the ''clean'' regime identified by Schep *et al.*[6] will be realized in future devices.[7] It is, therefore, worthwhile to explore new possibilities this regime offers for GMR optimization. One of them is control over the arrangement of ferromagnet/nonmagnet interfaces which govern the CPP GMR. The only factor that determines the ballistic CPP GMR of a periodic superlattice[6] is the magnetic contrast of its unit cell. This in turn depends on the difference between the strengths of the scattering potentials for the majority- and minority-spin electrons at a ferromagnet/spacer interface. Since nature provides us with a limited number of ferromagnet/nonmagnet combinations, this places an upper bound on what can be achieved with conventional periodic superlattices. The purpose of this contribution is to show that the total magnetic contrast, and hence the CPP GMR of a layered structure, can be greatly enhanced by disordering the sequence of interfaces in a multilayer. This can be achieved by growing magnetic or nonmagnetic layers, or layers of both types, with thicknesses that follow a predetermined pseudorandom sequence. I shall refer to such multilayers as pseudorandom spin valves. It will be demonstrated that the CPP GMR of the pseudorandom spin valve increases exponentially with its thickness as long as the valve remains in the ''clean'' regime.

In contrast to periodic superlattices,[6] CPP transport in pseudorandom spin valves is not in the ballistic regime. This is because electrons are scattered from a disordered array of interfaces and the mean free path for this type of scattering is, therefore, only of the order of the distance between two neighboring interfaces, i.e., very short. Nevertheless, the CPP GMR of a pseudorandom spin valve can be calculated rigorously from the Kubo formula. The calculation will be described for a multilayer with alternating magnetic and nonmagnetic layers which is prepared according to the following prescription. A pseudorandom sequence of integers $\{M_i\}$ distributed uniformly over an interval $[M_{\min}, M_{\max}]$ is generated and the thicknesses M_i of the magnetic layers (measured in numbers of atomic planes) are chosen to follow this sequence. All the nonmagnetic layers have the same thickness of N atomic planes chosen so that the exchange cou-

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pling between any two neighboring magnetic layers is antiferromagnetic. The layer magnetizations in zero field are, therefore, antiparallel. I stress, that once such a multilayer is prepared, the position of each interface in it is known precisely. Any particular multilayer for which a calculation of the GMR is made can, therefore, be reproduced experimentally by growing the magnetic layers with the same known sequence $\{M_i\}$, and vice versa. In other words, the relevant quantity to be calculated is the sample specific GMR, which means that no statistical averaging is involved.

CPP GMR measurements are usually made in the two-terminal geometry.[4,3] I shall, therefore, calculate the CPP GMR of a pseudorandom spin valve sandwiched between two semi-infinite ''ideal'' lead wires with negligible resistances which play the role of contacts.[8,5]. Under these conditions, the conductance Γ^σ in a spin channel σ can be calculated for any configuration of the magnetic layers from the Kubo formula.[9,8,5,3]. The usual GMR ratio $R_{\rm GMR}$, defined in terms of the conductances for the ferromagnetic (FM) and antiferromagnetic (AF) configurations of the magnetic layers, is given by

$$R_{\rm GMR}=(\Gamma^{\uparrow}_{\rm FM}+\Gamma^{\downarrow}_{\rm FM}-2\Gamma^{\uparrow,\downarrow}_{\rm AF})/2\Gamma^{\uparrow,\downarrow}_{\rm AF}. \tag{1}$$

Since the wave vector $\vec{k}_\parallel$ parallel to the layers is conserved, the total conductance Γ^σ is the sum of partial conductances,

$$\Gamma^\sigma=\sum_{\vec{k}_\parallel}(e^2/h)\Gamma^\sigma(\vec{k}_\parallel), \tag{2}$$

where $\Gamma^\sigma(\vec{k}_\parallel)$ is the transmission coefficient in the channel $(\vec{k}_\parallel,\sigma)$ (Refs. 9,3) and the sum in Eq. (2) is over all $\vec{k}_\parallel$ from the two-dimensional Brillouin zone (BZ). The transmission coefficient $\Gamma^\sigma(\vec{k}_\parallel)$ for a general multiorbital tight-binding band structure is given by

$$\Gamma^\sigma(\vec{k}_\parallel)=4{\rm Tr}\{\tilde{G}^\sigma_{00}(\vec{k}_\parallel)t_{01}(\vec{k}_\parallel)\tilde{G}^\sigma_{11}(\vec{k}_\parallel)t_{10}(\vec{k}_\parallel) - {\rm Re}[\tilde{G}^\sigma_{01}(\vec{k}_\parallel)t_{10}(\vec{k}_\parallel)\tilde{G}^\sigma_{01}(\vec{k}_\parallel)t_{10})]\}. \tag{3}$$

Equation (3) is a straightforward generalization of the result obtained earlier by Lee and Fisher[8] for a single-orbital tight-binding model. The indices 0,1 in Eq. (3) label any two neighboring principal planes parallel to the layer structure, $\tilde{G}_{i,j}(\vec{k}_\parallel)=(1/2i)[G^-_{i,j}(\vec{k}_\parallel)-G^+_{i,j}(\vec{k}_\parallel)]$, and $G^-_{i,j}(\vec{k}_\parallel)$, $G^+_{i,j}(\vec{k}_\parallel)$ are the matrix elements between principal planes i,j of the advanced and retarded one-electron Green's functions evaluated at the Fermi energy E_F. Similarly, t_{01} is the tight-binding hopping matrix between the principal planes 0,1. Because of the current conservation, the choice of the planes 0,1 is arbitrary. Finally, the trace is taken over all the orbital indices that are contained implicitly in the principal layer indices 0,1.

For multilayers with nonmagnetic layers made of Cu, a single-orbital tight-binding model should already contain all the interesting physics since for every $\vec{k}_\parallel$ there is only one band at the Cu FS. The Kubo formula will, therefore, be evaluated for a simple cubic (sc) single-orbital tight-binding band with nearest-neighbor hopping and (001) orientation of the layers. In that case, $t_{01}=t_{10}=t$ is a scalar and each principal layer consists of a single atomic plane. The Green's functions are calculated assuming that the electrons are noninteracting in the nonmagnetic spacer and experience exchange-split one-electron potentials in the ferromagnet. Although this is not essential, the same atomic potentials in the leads and nonmagnetic layers are assumed and the same hopping t in all the layers is used.

An efficient way[8] to calculate the matrix elements G_{01}, G_{00}, and G_{11} is to set first the hopping t_{01} between the planes 0 and 1 equal to zero. This creates an imaginary cleavage plane which separates the infinite sample into two independent left and right overlayers on semi-infinite ideal leads. It follows from Dyson's equation that the required matrix elements of the Green's function for the connected sample are given by $G_{00}=(1-t^2g_{00}g_{11})^{-1}g_{00}$, $G_{11}=(1-t^2g_{00}g_{11})^{-1}g_{11}$, and $G_{01}=G_{10}=tg_{00}G_{11}$, where g_{00} and g_{11} are the surface matrix elements of the Green's functions for the isolated left and right overlayers. It is convenient to pass the cleavage plane between the first and second atomic planes of the right ideal lead. The right overlayer is, therefore, just a semi-infinite ideal lead and its surface Green's function g_{11} is known analytically.[10] The left overlayer consists of a semi-infinite ideal lead, the pseudorandom spin valve, and one atomic plane of the right ideal lead. The surface Green's function of the left overlayer is generated recursively[8,11] from the surface Green's function of the left semi-infinite ideal lead. All the atomic planes of the left overlayer are deposited one by one on the left lead and the Green's function is updated after each deposition from the Dyson equation

$$g^\sigma_{\rm new}(\vec{k}_\parallel)=[E_F-V^\sigma-w(\vec{k}_\parallel)-t^2g^\sigma_{\rm old}(\vec{k}_\parallel)]^{-1}, \tag{4}$$

where $w(\vec{k}_\parallel)$ is the in-plane dispersion common to all the atomic planes and V^σ is the local atomic potential depending on the type of an atomic plane which is being deposited. The local potential in each atomic plane of the ferromagnetic layers is given by $V^{\uparrow,\downarrow}=V^{\rm FM}\mp\Delta/2$, where $V^{\rm FM}$ is the atomic level in the ferromagnet and Δ is the exchange splitting. The potential in each atomic plane of the nonmagnetic spacer is $V^\sigma=V^{\rm sp}$. I recall that the thicknesses of the magnetic layers follow a pseudorandom sequence $\{M_i\}$ whereas the nonmagnetic spacer layers are all deposited with the same thickness of N atomic planes. The last recursion step is the deposition of a single atomic plane of the ideal lead, which gives the exact one-electron Green's function g_{00} of the left overlayer.

An example of a sequence of potentials seen by the majority- and minority-spin electrons in the FM and AF configurations of a pseudorandom spin valve is shown in Fig. 1. (The atomic potentials in a tight-binding model are, of course, defined on discrete atomic planes; the solid lines in Fig. 1 are, therefore, only a guide to the eye.) The potentials are measured in units $2t=1$, where t is the hopping integral. They were chosen to model approximately the situation in Co/Cu. Because there is a very good match between the bands in Co and Cu for the majority-spin electrons,[6] the potential step at the spacer/ferromagnet interface for the majority-spin carriers was chosen to be small $V^\uparrow-V^{\rm sp}=0.2$. A much larger potential step $V^\downarrow-V^{\rm sp}=0.6$ was chosen to simulate a mismatch between the bands in Co and Cu in the minority-spin channel.[6] The Fermi energy in the spacer, measured from the bottom of the band, was set at

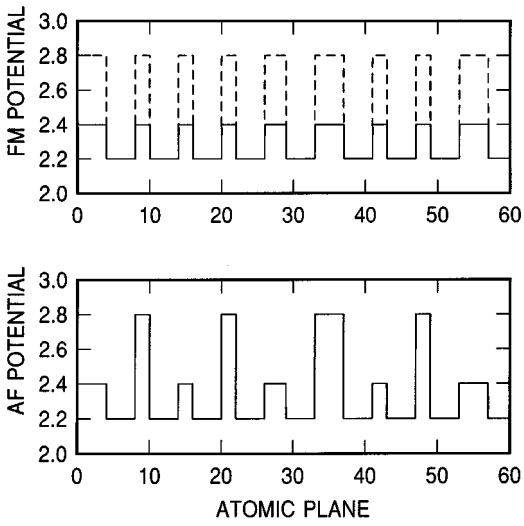

FIG. 1. Pseudorandom potentials seen by the minority-spin (dashed line) and majority-spin (solid line) electrons in the ferromagnetic (FM) configuration, and the potential seen by electrons of either spin orientation in the antiferromagnetic (AF) configuration. The parameters are given in the text.

$E_F = 0.8$ (also in units $2t = 1$). This value was chosen so that the perpendicular energy $E_\perp = E_F - E_\parallel$ of electrons incident at small angles on the interface lies above the top of the potential relief. This again mimics the situation in Co/Cu(001).[6] Finally, four atomic planes were used for each spacer layer and the thickness of the magnetic layers was made to fluctuate between two and four atomic planes.

Taking the pseudorandom sequence of potentials shown in Fig. 1 as a typical example, the transmission coefficients for the majority- and minority-spin electrons in the FM and AF configurations of the magnetic layers were evaluated from Eqs. (3) and (4) for structures containing in total of up to 500 atomic planes. The CPP GMR was then determined from Eqs. (1) and (2). Convergence in the sum over $\vec{k}_\parallel$ was achieved for about 2000 points in the irreducible segment of the two-dimensional BZ. Since there are no approximations in the recursion method based on repeated application of Eq. (4), the CPP GMR is evaluated from the Kubo formula exactly.

The natural logarithm of the calculated CPP GMR is plotted in Fig. 2(a) (circles) against the thickness of the valve which is measured in the number of repeats $N_{\rm rpt}$ of a ''unit cell'' consisting of two magnetic and two nonmagnetic layers. The CPP GMR of an equivalent periodic superlattice with the same potentials is also shown in Fig. 2(a) (squares). For the superlattice, the thickness M of all the magnetic layers is taken to be the mean of the random distribution ($M = 3$). The average ''unit cell'' of the pseudorandom spin valve and the unit cell of the equivalent superlattice thus contain the same number of atomic planes (i.e., 14). It is clear from Fig. 2(a) that the CPP GMR of the pseudorandom spin valve increases exponentially with the valve thickness. The ballistic CPP GMR of the equivalent ordered superlattice is, on the other hand, independent of the thickness. It can be seen from Fig. 2(a) that, even for a relatively thin pseudorandom valve $N_{\rm rpt} \approx 20$ (about 300 atomic planes), a huge enhancement of the CPP GMR by a factor of almost 5000 is

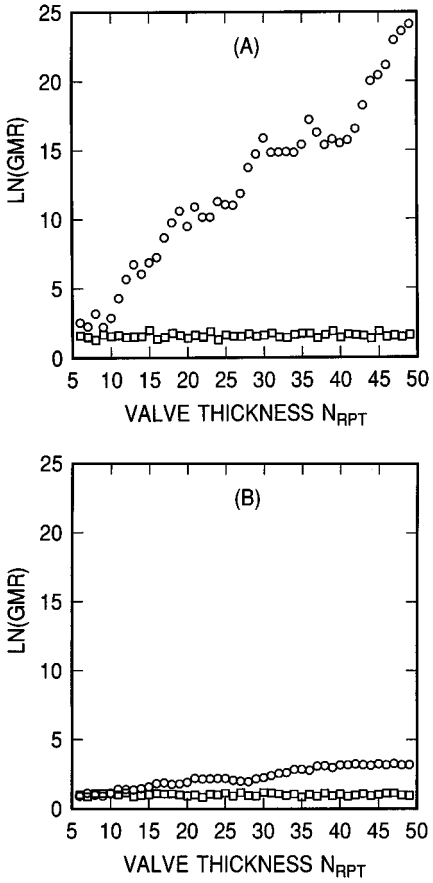

FIG. 2. Dependences of the logarithm of the CPP GMR ratio of a pseudorandom spin valve (circles) and of an equivalent periodic superlattice (squares) on the thickness of the structure $N_{\rm rpt}$: (a) $N = 4$, $M_{\rm min} = 2$, $M_{\rm max} = 4$; (b) $N = 5$, $M_{\rm min} = 5$, $M_{\rm max} = 6$; the heights of the potentials are as in Fig. 1.

achieved. Calculations for other choices of the potentials and for different pseudorandom sequences of layer thicknesses all lead to qualitatively the same behavior. Moreover, an exponential enhancement of the CPP GMR is obtained not only for a pseudorandom sequence of magnetic layer thicknesses but also when the thicknesses of the nonmagnetic layers or the thicknesses of both types of layer are randomized.

The physical mechanism that causes an exponential growth of the CPP GMR is Anderson localization.[12] In the absence of impurities, conduction takes place in independent one-dimensional $(\vec{k}_\parallel, \sigma)$ channels. Electrons in each channel see the same pseudorandom sequence of potentials but they sample it with different perpendicular energies $E_\perp = E_F - E_\parallel$. The conductance in every channel satisfies[12] $\Gamma^\sigma(\vec{k}_\parallel) \propto \exp[-L/\lambda^\sigma(\vec{k}_\parallel)]$, where L is the length of the channel (valve thickness) and $\lambda^\sigma(\vec{k}_\parallel)$ is the localization length. An exponential decrease of the conductance of a one-dimensional disordered conductor is not only predicted theoretically but has recently been observed at room temperature in very thin gold wires.[13]

The key property of a pseudorandom spin valve is that the degree of disorder in it, and hence the degree of electron localization, is controlled by an applied magnetic field. It can be seen from Fig. 1 that the degree of disorder is large for carriers of either spin orientation in the AF configuration

(and also for the minority-spin carriers in the FM configuration) but becomes small in the majority-spin channel when the magnetic moments are aligned parallel in an applied field. The localization length in a typical $\vec{k}_{\|}$ channel is, therefore, short in the AF configuration for both spin orientations but becomes long for the majority-spin electrons in the FM configuration. It follows that the magnetic contrast of each $\vec{k}_{\|}$ channel increases exponentially with its length. Although localization lengths fluctuate from channel to channel and the magnetic contrast of some channels may, therefore, increase only very slowly, the results shown in Fig. 2(a) demonstrate that the weight of such low-contrast channels is negligible in the sum over $\vec{k}_{\|}$ which determines the GMR ratio.

Because the disorder in a pseudorandom spin valve is imposed and, hence, reproducible, it can be manipulated to optimize the GMR ratio. Given the constraint that the total thickness of the pseudorandom spin valve should remain smaller than the mean free path for scattering from impurities, one can formulate several simple rules for GMR optimization. First, the magnetic contrast of an individual ferromagnet/spacer interface should be high, which is the case, e.g., for Co/Cu or Fe/Cr. The thickness of the layers that are not subject to fluctuations should be small since this maximizes the number of repeats, N_{rpt}. The mean thickness of the layers whose thicknesses are made to fluctuate should be also small since it again maximizes N_{rpt}. On the other hand, fluctuations in the thickness, $\Delta M = M_{\max} - M_{\min}$, should be large compared with the mean M. In fact, the ratio $\Delta M/M$ determines the rate of increase of GMR with the valve thickness. This is illustrated in Fig. 2(b) where the CPP GMR of a pseudorandom valve with the same heights of potentials as in Fig. 1 but with $M_{\min}=5$, $M_{\max}=6$, and $N=5$ is compared with that of an equivalent periodic superlattice ($N=5$, $M=5$). The exponential growth of the GMR in Fig. 2(b) is now so slow that an enhancement over the periodic superlattice is only by a factor of 2–3 for $N_{\mathrm{rpt}} \approx 20$. This example serves to illustrate that Anderson localization effects would not be observable in conventional superlattices in which fluctuations of the order of one atomic plane occur spontaneously due to terrace formation. To achieve the desirable very large enhancement of the CPP GMR, one has to grow deliberately multilayers with large fluctuations in layer thicknesses.

The predicted enhancement of the CPP GMR should be observable in pseudorandom nanostructure valves sandwiched between macroscopic contacts, provided the valve dimensions are smaller than the mean free path for scattering from random imperfections (other than the interfaces). This is the experimental situation to which the present calculation based on the Kubo formula strictly applies. A long mean free path is, therefore, desirable but the present model calculations indicate that several tens of nanometers should be sufficient for a large enhancement. This is within the reach of current growing techniques.

An interesting question arises whether the enhancement of the CPP GMR persists in pseudorandom valves with transverse dimensions greater than the mean free path. Scattering from imperfections leads to mixing of $\vec{k}_{\|}$ channels which might eventually destroy one-dimensional localization. However, what the upper bound is, if any, on the valve diameter is difficult to estimate theoretically. As a first step, it would, therefore, be interesting to measure the CPP GMR of pseudorandom valves with macroscopic transverse dimensions since they can be readily manufactured by the existing techniques. It is, however, to be expected that a very large enhancement can only be achieved in the ''clean'' regime, i.e., with nanostructure valves.

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